THE DISPOSAL OF RADIOACTIVE FISSION GASES BY ADSORPTION

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Abstract

In the operation of nuclear reactors, nuclear fuel reprocessing plants, and in-pile experiments, special provision must be made for disposal of gaseous fission products to prevent contamination of the atmosphere to an unacceptable degree. A disposal process is described in which the noble gas fission products, krypton and xenon, are delayed relative to the sweep gas by physical adsorption as they pass through an adsorbent such as activated charcoal. A theoretical expression describing this process has been developed, using a theoretical plate analysis, and has been verified experimentally. The retention time for a gas present in trace concentration is proportional to the amount of charcoal in the adsorber and to the adsorption coefficient which is evaluated experimentally for a particular combination of materials and conditions. The retention time is inversely proportional to the volume flow rate of the sweep gas. The retention times of experimental adsorbers have been measured by a radioactive tracer technique using krypton-85 and xenon-133 to typify fission gases. Retention times have been measured for various adsorbents including different grades of activated charcoal, silica gel, activated alumina, and molecular sieve materials. Activated charcoal was the most effective adsorbent. Retention times were measured using helium, hydrogen, argon, nitrogen, oxygen, air, CO2, Freon-12 and krypton as sweep gases, individually. Retention time decreases logarithmically as temperature increases. Water vapor reduces the retention time for krypton on charcoal as does CO2. Krypton interferes negligibly up to 1000 microns partial pressure. Provision must be made for the dissipation of heat from radioactive decay. Ignition temperatures for charcoal in oxygen were determined and methods of prevention and control of charcoal fires were investigated. Experimental results described have been used to design off-gas adsorber systems for in-pile experiments and to analyze the performance of an adsorber for a homogeneous circulating fuel reactor.

Introduction

In the operation of nuclear reactors, nuclear fuel reprocessing plants, and in-pile experiments, special provision must be made for the disposal of gaseous radioactive fission products to prevent contamination of the atmosphere. Figure 1 shows the important fission gas isotopes. Their half-lives range from a few minutes up to ten years. Among the krypton isotopes the longest-lived is krypton-85, approximately ten years, but the next longest is only four hours. The longest-lived xenon isotopes are Xe-131m and Xe-133, having half-lives of 12 days and 5.27 days, respectively; the shorter-lived one being more abundant. The longestlived iodine isotope is I-131, having a half-life of eight days.

When a reactor accident occurs, all the radioactive isotopes shown in Figure 1 can appear in the gas system, even after removal of particulate matter. In reactors and in-pile experiments having circulating fuel, fission gases are released having the entire range of half-lives all the way down to a few seconds. By the time the off-gas reaches the disposal system, the age is such that only isotopes having half-lives greater than a few minutes are present to an appreciable extent. When solid fuels from reactors and in-pile experiments are dissolved after aging, only ten year Kr-85 and five and twelve day xenon and eight day iodine require consideration.

Various methods have been used to handle the noble gases, krypton and xenon. They have been vented to the atmosphere (1) or stored in large containers for decay. Solvent extraction has been applied successfully (2). This paper is concerned with the application of the process of dynamic adsorption of noble gas fission products.

Radioactive iodine vapor has been removed from air streams by caustic scrubbers or by hot silver reactors. Using I-131 tracer, we have found that it is removed at least 99.9% by charcoal adsorbers. This work, which is of a preliminary nature, is reported in ORNL-CF-58-5-59 (3), and more detailed studies are currently in progress.

Dynamic Adsorption

In the process of dynamic adsorption, the noble fission gases, krypton and xenon, are physically adsorbed from a moving sweep gas, by a material such as activated charcoal in a manner similar to that used in gas chromatography. Adsorption equilibrium exists at every point within the adsorber. Cnly a small fraction of the krypton and xenon remain in the moving gas phase, and as a result, they are delayed relative to the sweep gas. The delay times are characteristics of the adsorber and may be measured experimentally. In some applications the adsorber is regenerated continuously by radioactive decay of the adsorbed fission gases.

We have applied a radioactive tracer technique to measure retention times. A short pulse of radioactive krypton-85 and/or xenon-133 is injected into the flowing gas stream at the entrance of the adsorber. The concentration of radioactive gas is measured at the outlet of the adsorber as a function of time. Figure 2 shows a typical elution graph for a mixture of Kr and Xe. The ordinate is the relative concentration of radioactive gas and the abscissa is the time after injection. For each of the two elements, krypton and xenon, there is a characteristic breakthrough time, and a time to peak which for symmetrical elution curves is the average time for the passage of individual fission gas atoms through the adsorber. The breakthrough time is used for conservative design; however, the average retention time is easier to predict.

Theory

The transport of fission gases through an adsorber may be treated theoretically as shown in Figure 3 by considering a small element of length of the adsorber, dy. The rate of change of concentration of a fission gas, dP/dt, in the element dy is proportional to the concentration gradient, dP/dy, and to the volume flow rate, F, and is inversely proportional to

ISOTOPE	HALF-LIFE	ISOTOPE	HALF-LIFE
KRYPTON Kr ^{83m} 85m	114 MIN	XENON Xe ¹³⁵ 137	9.13 HR
Kr ^{85m}	4.36 H R	Xe ¹³⁷	3.9 MIN
Kr ⁸⁵	10.27 YR	Xe ¹³⁸	17 MIN
Kr ⁸⁷	78 MIN	10DINE	12.6 HR
Kr ⁸⁸	2.77 HR	1 ¹³⁰	
Kr ⁸⁹	3.18 MIN	131	8.05 DAYS
XENON		132	2.4 HR
Xe ^{131m}	12 DAYS	133	20.8 HR
Xe ^{133m}	2.3 DAYS	134	52.5 MIN
Xe ¹³³	5.27 DAYS	135 136	6.68 HR
Xe ^{135m}	15.6 MIN	1.00	1.43 MIN

Fig. 1—Fission products important in reactor off-gas streams.



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Fig. 3—Theoretical equations for transport of fission gases through an adsorber.



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Fig. 4—Theoretical plate equations for fission gas transport.

the equilibrium adsorption coefficient, k, and to the unit loading of the adsorber m/L.

- P = concentration or partial pressure of fission gas in the gas phase (atm).
- t = time at which P is observed (min).
- F = flow rate of sweep gas (cc/min).
- L = length of adsorber (cm).
- m = amount of adsorbent in adsorber (gm).
- k = either dynamic or equilibrium adsorption coefficient for fission gas on adsorbent (cc.atm/gm.atm or cc(STP)/gm.atm, respectively). Although these sets of units are somewhat dissimilar, the two coefficients, as employed herein, are theoretically equal numerically.
- y = distance from entrance of adsorber at which P is
 observed (cm).
- f function which describes arbitrary variation of P at inlet of adsorber.

In the theoretical derivation k is the equilibrium adsorption coefficient relating the amount of fission gas adsorbed to the local partial pressure of fission gas. In the application of experimental results k is the dynamic adsorption coefficient which relates the average retention time to m/F. In the experimental test, f_0 describes an instantaneous pulse of tadioactive fission gas. t is the time after injection of the radioactive pulse. The solution in Figure 3 states that the pulse will emerge unchanged in shape, but delayed by a time, km/F. This simple treatment yields a useful value for the time to peak. However, it indicates erroneously that there will be no distortion of the pulse as it travels through the adsorber, because this mathematical treatment ignores pulse broadening processes.

We have used a theoretical plate model to treat dynamic adsorption while providing for the pulse broadening processes (4). In this analysis the adsorber is imagined to be divided into a number of theoretical chambers, N, with adsorption equilibrium in each one. The differential equation in Figure 4 describes the concentration of a fission gas as a function of time in the first one of these theoretical chambers, after injection of an instantaneous pulse of fission gas. The symbols have the same significance as in Figure 3. The quantity N is the number of theoretical chambers in an adsorber having an amount of adsorbent, m. The rate of change of fission gas concentration is proportional to the flow rate, F, and to the instantaneous concentration, P, and is inversely proportional to the adsorption coefficient, k, and to the weight of adsorbent in a theoretical chamber, m/N. The solution for the fission gas concentration at the outlet of the last chamber is given by the second expression in Figure 4. The time to peak concentration at the outlet is shown in the third expression and is similar to the last equation in Figure 3. The time to peak or average retention time is proportional to km/F, and is relatively insensitive to the number of theoretical chambers, N. The second equation in Figure 4 describes the shape of the elution curve and may be used to predict the breakthrough time.

Figure 5 compares the theoretical plate equation with experimental data. The theoretical curve has been fitted to the experimental curve by selecting values for k and N. The fit of the curves in the break-through region indicates the usefulness of the theoretical plate model. Using this analysis, experimental data may be reduced to two elementary parameters, k and N.

Experiments

The information on retention of fission gases under various conditions has been obtained experimentally using apparatus as shown in Figure 6. A constant flow of sweep gas, in this case oxygen, passes through an adsorber, a flow meter and past a Geiger-Muller tube. The radioactive tracer is injected at the krypton holder and the counting rate, which is proportional to concentration of tracer, is observed as a function of time by the G-M counter. Figure 7 shows the adsorbers on the right, the counters in the middle background, and instruments for registering counting rates on the left. Figure 8 shows an end-window Geiger-Muller counter tube equipped with a gas cell. Using apparatus of this type, elution curves shown in Figure 9 were obtained. Here the relative concentration of radioactive krypton in the effluent gas is plotted versus time after injection of the krypton pulse at the entrance of the adsorber. The adsorber contained 782 grams of charcoal and the sweep gas was oxygen at a flow of 250 cc/min. Elution curves are shown at four different temperatures. The lower the temperature, the longer is the retention time. Refrigerated beds would usually be more desirable except for possible failure of refrigeration. These data are analyzed to obtain the adsorption coefficient k as a function of temperature as shown in Figure 10, which also includes some other data.

The efficiency of various adsorbents has been investigated using this same technique. In Figure 11 activated charcoal is compared with silica gel, Driocel, activated alumina, and porous glass. Various grades of activated charcoal are compared in Figure 12. Columbia Grade G is slightly superior; however, the samples are all about the same with respect to k. Figure 13 shows that the performance of Linde Air Products Molecular Sieve materials is not as good as that of charcoal but where the combustibility of charcoal is a hazard, these materials are the best choice if moisture can be excluded. Sieve Material 5A is superior to the others. LA has too small a pore size to admit krypton. Columbia Grade G activated charcoal has been used for most of the tests described below.

Figure 14 shows the effects of various sweep gases. Krypton is retained more efficiently in the presence of helium than with other gases, because helium is adsorbed to a lesser degree. Freon-12 is not suggested as a reactor sweep gas but it is used to illustrate the effect of a strongly adsorbed gas.

Figure 15 shows the effect of krypton partial pressure in oxygen with the total pressure constant at one atmosphere. In practical off-gas systems, the fission gas partial pressure does not ordinarily exceed 1 millimeter of mercury and the efficiency is not significantly affected. Figure 16 shows that small amounts of CO₂ cause a substantial reduction in the retention time. This effect can be quite important in the event of an accidental fire. One of our charcoal adsorbers serving a 5 MW reactor was ignited by an explosion of radiolytic deuterium in oxygen. Although the fire was put out without interrupting reactor operation, the combustion product, CO₂, reduced the retention time by approximately a factor of two, temporarily, and resulted in a measurable but not hazardous release of radioactive gases. The adsorber was restored to normal service after the CO₂ had been displaced.

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Fig. 5—Typical distribution of fission gas activity in experimental effluent gas stream.

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Fig. 6-Schematic diagram of equipment for measuring retention times.



Fig. 7—Photograph of equipment for measuring retention times.



Fig. 8—End window GM tube, equipped with gas cell, for measuring Kr^{85} and Xe^{133} .



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Fig. 10—Effect of temperature on dynamic adsorption coefficient for charcoal.

Fig. 9—Effect of charcoal temperature on elution curves.



Fig. 11-Dynamic adsorption coefficients for miscellaneous adsorbents.



Fig. 12—Dynamic adsorption coefficients for various activated charcoals.



Fig. 13-Dynamic adsorption coefficients for Linde molecular sieves.



Fig. 14 — Dynamic adsorption coefficients for columbia G activated carbon with various sweep gases.



Water vapor has an effect similar to that of CO₂. Figure 17 shows that the efficiency of charcoal may be reduced to one-half by adsorbed moisture. Charcoal in equilibrium with air at 50% relative humidity contains 5% water by weight.

The adsorption coefficient, k, shown in the preceding figures, is used for evaluating the average retention time, t_{max} . This quantity may be converted to the more useful breakthrough time, t_b , by the data contained in Figure 18, which applies to a 0.75 inch diameter charcoal adsorber with 0₂ at 250 cc/min. The ratio of t_b to t_{max} approaches 90% for very long adsorbers. At 10 or 20 feet this ratio runs from 75 to 80%. For less than 5 feet ratios are less than 50%.

The adsorber must be fully packed. Figure 19 shows the performance of a horizontal adsorber which has only 3% of void space. The resulting tunnel provides a "short circuit" for the gas and the breakthrough time was only one-tenth of that ordinarily expected. It is desirable to install adsorbers vertically.

The adsorption coefficients for xenon have been determined for only a few conditions because only short-lived tracers are available. Xenon is retained about 10 to 30 times longer than krypton, depending upon temperature, as shown in Figure 20.

Design of Practical Adsorbers

With the theoretical results previously described and with the experimental data, we are now prepared to design a practical adsorber for krypton and xenon fission gases. In some applications it is useful to stop the flow of gas as soon as radioactivity is detected; and the retention time required is the time necessary to stop the flow of gas. In other cases it is necessary to remove fission gases from a continuously flowing gas stream. The adsorber delays the passage of radioactive krypton and xenon until they have decayed sufficiently except for Kr-85. The retention time required depends upon the concentration of fission gases entering the adsorber and upon the allowable concentration in the exit gases. The production of the various isotopes of krypton and xenon may be evaluated using ORNL-2127 (5) by Blomeke and Todd. The allowable emission of fission gas isotopes depends upon the location of the plant and the environmental conditions. It may be limited by the permissible external or internal doses to personnel in the plant or in the surrounding community. Interference with sensitive radiation detection instruments may be the limiting factor. The report ORNL-CF-58-12-10 (6) will assist in evaluating the allowable and initial emission rates.

From the ratio of input concentration to allowable output concentration and the half-life we calculate the decay time needed for each isotope and select the longest decay time required. From the last expression in Figure 4, using the approximation (N - 1)/N = 1, which is valid for large N, we obtain the amount of adsorbent required.

$$m = \frac{Ft_{b}}{kt_{b}}/t_{max}$$

- m = amount of adsorbent needed to provide the required breakthrough time (gm).
- F = flow rate of sweep gas (cc/min).







Fig. 19—Effect of void space in horizontal charcoal adsorber.

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Fig. 20—Dynamic adsorption coefficients for krypton and xenon on charcoal with oxygen or nitrogen sweep gas.



Fig. 21-Model of fission gas adsorption system used on 5 Mw fluid fuel reactor.

- k = dynamic adsorption coefficient obtained by retention time experiments under the conditions for which the adsorber is being designed (cc atm/gm atm).
- tb[#] breakthrough time. Selected to provide time for defay of radioactive gases or for emergency shutdown of gas flow (min).
- t_max = average retention time for fission gas atoms passing through adsorber. This quantity is strictly speaking the retention time for the greatest proportion of fission gas atoms and is very nearly identical to the average retention time (min).

Heat of radioactive decay may be removed by using small diameter pipes to contain the adsorbent and by immersing the pipes in cooling water. The pipes should be smaller near the entrance to the adsorber where the heat load is greatest and may be larger in diameter near the outlet. High temperatures interfere with adsorption of the fission gases and in the presence of an oxidizing gas can cause a fire hazard. We have observed ignition temperatures for charcoal in oxygen as low as 290° C. It is necessary to provide for measurement of the temperature of the adsorber so that if undesirable conditions occur, corrective action may be taken. A fire may be controlled safely by diverting the flow of the oxidizing gas to an alternate adsorber. This procedure has been tested experimentally as reported in ORNL-CF-58-6-6 (7), and was applied successfully to a fire which occurred accidentally in one of two charcoal adsorbers which were handling the fission gases in oxygen from a 5 MW homogeneous reactor.

Refrigerated adsorbers may be used to advantage where the size of the adsorber is important as with mobile reactors; however, it is necessary to take several precautions in their use. Failure of the refrigeration system produces an unsafe condition in which radioactive gases are desorbed as the adsorber warms up. In the applications where such a release cannot be tolerated, several alternatives are available, most of which require stopping the flow of radioactive gas from the reactor. The adsorber system may be sealed off in an emergency and designed for the very high pressures which may result or be provided with an expansion volume in the adsorber system inside the shielding enclosure. Emergency stand-by refrigeration systems with an emergency power source may be provided or an auxiliary adsorber system into which the gas from the main adsorber can be allowed to expand may be used. Another hazard is the possibility of explosion in the adsorber if oxidizing gases, even in trace concentrations, are exposed to the charcoal at very low temperatures. Allowance must be made for the fact that the heat released by radioactive beta decay of the fission gases will cause the central temperature of the adsorber to be higher than the wall temperature, and the refrigeration system must have capacity to carry away this heat of beta decay.

Applications

We have applied this information to the design of several adsorbers. In a fused salt in-pile loop experiment we used a 34 pound charcoal adsorber with a gas flow rate of 20 scfm of air at 26°C. It provided a one minute retention time for krypton, sufficient for scramming the reactor and for stopping the flow of air, leaving the krypton and xenon in the charcoal to decay. The procedures for designing a refrigerated adsorber for a similar application are given in ORNL-CF-58-7-71 (8). We have also applied this information to an adsorber system used on a 5 MW homogeneous reactor (9). Figure 21 shows a model of one of the three adsorbers used. Each one contains 520 pounds of charcoal in pipes ranging from 1/2 to 6 inches in diameter. Two of the adsorbers are normally used in parallel and carry a total of 2500 cc/min of oxygen contaminated with fission gases. At 26°C the retention time for krypton is six days and for xenon sixty days, sufficient for virtually complete decay of all the fission gases except krypton-85 which is vented through a small stack. This system has performed satisfactorily for more than a year.

Conclusion

The physical adsorption of noble gases upon adsorbents such as activated charcoal may be used to delay the release of gaseous fission products from nuclear reactors. Two design philosophies have been used. The first employs an adsorber to provide a delay time during which the flow of the off-gas stream from a source of fission gases may be halted following an accidental release of radioactive gases. The second philosophy of design utilizes an adsorber to delay the radioactive fission gases for a time long compared to their half-lives so that they decay virtually completely in the adsorber. Only the ten year krypton-85 emerges from an adsorber of this type. This isotope may be discharged safely to the atmosphere under certain conditions in the case of low or medium power level reactors. High power reactors, especially near the heavily populated areas, will require an adsorber system of the first type for extracting krypton-85.

A theoretical analysis of the process whereby the fission gases are retained by an adsorber from a flowing gas stream provides the basis for design of practical units. Experimental data are provided which may be used under a wide variety of conditions in the design of adsorbers for the off-gas systems of nuclear reactors and of in-pile experiments.

The principal advantages of the dynamic adsorption process for controlling fission products are its simplicity and freedom from maintenance or replacement requirements. The entire fission gas processing unit is contained in metal pipes, which may be immersed in coolant water at ambient temperature, buried underground to confine the gamma radiation. The only requirement for instrumentation is thermocouples at a few representative points to verify the results of heat transfer calculations in the design of the adsorber and equipment for fontrolling or metering flow.

Acknowledgements

We acknowledge ideas contributed by C. C. Bolta, D. E. Guss, and T. W. Leland.

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Discussion

- Q. (W. B. Harris, NYOO) What does the title of the paper mean by disposal?
- A. It means that the short-lived fission gases are converted to solid materials so that they are no longer in the gas stream.
- Q. (L. Silverman, Harvard) Would like to know what the relative costs of treating this gas stream are and what volume can you handle in the system?
- A. Equipment usually just involves pipes filled with charcoal and once they are installed they last more or less indefinitely. As far as volume of gases is concerned, it would depend on the pressure drop.

Recovering radio active gases will be important - developments this morning indicate ways of recovery and minimum environmental problems - they are handling small amounts of gas.

- Q. Would like to have some idea of cost per cfm.
- A. I don't believe I had better name a figure for the cost, I believe it is available.

THE RECOVERY OF FISSION PRODUCT XENON AND KRYPTON BY ABSORPTION PROCESSES

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NOTE: This paper is not included in this publication because it has already been published as BNL-542 (T-140) and is available from the Office of Technical Services, U. S. Department of Commerce, Washington, D. C., for \$0.75 per copy.

However, the discussion which followed the presentation of this paper is included here.

Discussion

- Q. (F. T. Selleck, Fluor) Very much interested in the work on means of separating fission gases especially in its use for power reactors.
 whether flow sheets shown by Steinbergh are process designs based on laboratory scale in pilot plants or at actual operating plants
- A. Based on estimates from laboratory data and small pilot plants.
- Q. Also interested in type of compressors used and whether or not you have any good history of leakage or was it a straight problem of higher pressure operation?
- A. We don't have any real good operating data on compressors engineering problem that will have to be looked into.
- Q. Cost of CO₂ quoted was that per liter of fission gases produced?
- A. This is per liter of fission gases produced. I might add that you can recover the CO_2 in another process but you have to pay for this in some sort of way such as a hot carbonate process for recycling from CO_2 but you have to pay for a plant to do this.
- C. (Silverman) I would like to give a comment here because I think this question of recovery of radioactive gases which is going to be a problem in the power economy program in the question of Nuclear Power. It's going to be very important.

I think these developments we heard this morning both Ackley's paper and Steinberg's paper indicate ways of recovering and possibly minimizing the environmental problem. In one case recovery for economic purposes and the other case for preventing an environmental problem. I have some knowledge of the system at Oak Ridge and it seems to me that they are handling a very small volume of gas with a very large detention system so that what I would like to have had was some figure of the cost per CFM. I know that the original freeze-out system here at Idaho cost about \$500 per CFM but that was in the early days of trying to freeze out all of the rare gases.

A VENTURI SCRUBBER INSTALLATION FOR THE REMOVAL OF FISSION PRODUCTS FROM AIR

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ABSTRACT

A local exhaust collection system and a venturi scrubber installation for the cleaning of exhaust air contaminated with acid mists and mixed fission products are described in detail. The features of the collection system that are designed to offset the hazard of perchloric acid condensing in the collection system are stressed, and the feasibility of a venturi scrubber with a caustic solution for the removal of iodine vapors as a scrubbing medium is demonstrated. The efficiency of the scrubbing unit for removing acid mist, total fission products, and iodine vapor tested 90% or better. The performance of the units met design expectations and no major modifications were necessary.

INTRODUCTION

The construction of a new radiochemistry building at the Los Alamos Scientific Laboratory made it possible to design an entirely new control and air-cleaning system for the off-gas from the process of dissolving large filter papers containing fission products. In this process, a cellulose-type filter paper is dissolved in a hot mixture of nitric and perchloric acids. The off-gas is composed of: (1) perchloric acid mist, (2) nitric acid mist, (3) radioactive iodine vapor, (4) mixed fission products, and (5) oxides of nitrogen.

Two problems are associated with the off-gas: Condensation of the perchloric acid in the collecting system and the evolution of radioactive materials. Under certain imperfectly understood conditions, the accumulation of perchloric acid in an exhaust system gives rise to a serious explosion hazard. For this reason, special hoods with water sprays are frequently specified for operations in which perchloric acid fumes are evolved. The release of the radioactive material, mostly in the form of iodine vapors, to the atmosphere is undesirable because of health considerations and because of the possible increase in the airborne activity of the building intake air. A slight increase in the activity of this air creates serious difficulties by raising the background count of the elaborate electronic equipment in the radiochemistry building. It was a basic decision, therefore, that the new building be provided with facilities for cleaning the exhaust air from the dissolving process.

After considerable study, the Radiochemistry Group decided that dry boxes and the small laboratory fume hoods used in the old building were undesirable from an operational standpoint. It was determined that a laboratory fume hood, 8 feet long to permit the installation of two dissolving stations, would provide the most convenient setup for the operators. The required eight hoods of this type would exhaust approximately 16,000 cubic feet of air per minute (cfm), but cleaning this flow of air to the desired level of decontamination would require a large, expensive installation. Consideration was given, therefore, to small, local exhaust facilities located in close proximity to the source of contaminants and served by a separate exhaust system of approximately 350 cfm capacity. A typical hood installation is shown in Figure 1.



Fig. 1—Over-all view of hood and two dissolving stations.

The estimate of the air flow to be cleaned was a basic factor in determining the most suitable type of air cleaning. Other considerations in the cleaning of this particular exhaust air, however, posed a number of problems. The perchloric acid mist cannot be allowed to condense in the duct work and must, therefore, be controlled or removed at the hood. The oxides of nitrogen and iodine exist as gases or vapors. In addition, since the dissolving process is performed at irregular intervals, it should be possible to activate and deactivate the air-cleaning system without adversely affecting its performance.

Initial consideration was given to the use of wet filters for the removal of acid mists and a scrubbing tower using silver salts for the removal of the iodine vapors. The experience at the Los Alamos Scientific Laboratory with wet filters,¹ however, has not been entirely satisfactory, and estimates of the initial and maintenance cost for the complete system including a scrubbing tower were rather high. Also, the ability of this equipment to perform adequately under conditions of intermittent use was in serious question.

The use of a venturi scrubber as a means of removing acid mists and small particulate matter appeared attractive. The main question in connection with this type of scrubber was its ability to remove iodine vapors. A series of tests were, therefore, conducted on an existing venturi scrubber installation, and it was determined that the unit would not remove iodine vapors from the air stream if water was used as the scrubbing medium. A caustic solution was tried and removal efficiencies of about 95% were obtained for a variety of iodine vapor loadings. The normality of the caustic solution did not appear to be an important factor and 1.0 N sodium hydroxide was used in most experiments.

The use of the venturi scrubber with a caustic solution for this particular air-cleaning problem appeared to offer the following advantages:

1. A single unit would remove all contaminants of interest with good efficiency.

2. It had the ability to collect the contaminants in such a manner as to permit storage for radioactive decay and for ultimate disposal without exposing maintenance personnel.

3. The air collection and air-cleaning systems could be thoroughly decontaminated at the end of an operating period, and minimum maintenance would ensure maximum efficiency for the next operation.

4. Because of the simplicity of the system, it could be anticipated that maintenance would be minimal. The ease of decontaminating the unit would, in any case, simplify and reduce the cost of necessary repairs.

DESIGN CONSIDERATIONS

Local Exhaust Hood. The configuration of the local exhaust hood was determined by operating requirements, and the final design specified by the Radiochemistry Group is shown in Figure 2. Exhaust air requirements were determined empirically by varying the rate at which air was exhausted while the dissolving operation was actually being performed. For the initial studies, the effectiveness of the air flow pattern was determined by observing the capture of the fog generated when dry ice was dropped in beakers of boiling water. The studies indicated that a flow rate of 20 cfm would give satisfactory control under conditions in which the evolution of the fumes was at an anticipated maximum.

Venturi Scrubber. The design of the venturi scrubber was based on sixteen local exhaust hoods, exhausting 20 cfm each for a total flow rate of 320 cfm. At this flow rate, the anticipated loadings of interest are shown in Table I. Anticipated loadings are based on data obtained at the old dissolving installation and on the assumption that all dissolving stations would be operating.

The extremely low loadings and the existence of I^{131} in the vapor state were the major concerns of the Chemical Construction Corporation, fabricator of the venturi scrubber, in regard to the desired iodine removal efficiency of 95%.



Fig. 2—Details of local exhaust hood and dissolving station.



Fig. 3—Flow diagram-collection and air-cleaning systems.

Material	Loading
Acid Mist (nitric and perchloric)	500 to 3000 mg/m^3
Total Radioactivity	$l to 6 mc/m^3$
Expressed as I ¹³¹	0.008 to 0.05 µg/m
Expressed as Sr ⁹⁰	5 to 30 $\mu g/m^3$
Solid Particulate Matter	$< 10 \text{ mg/m}^3$
Range of Mass Median Size	0.58 to 11.0 μ

TABLE I Design Loading Factors

FINAL DESIGN

The final design of the entire system is shown in Figure 3 and discussed in detail below.

Local Exhaust Hoods and Exhaust Piping. The local exhaust hoods and piping were constructed of welded stainless steel. Horizontal runs of the piping were sloped, and regulating valves were placed only on vertical sections to facilitate drainage and washdown. A valvecontrolled bypass was utilized to enable the operator to control the air flow through the hood and still maintain a constant flow through the venturi scrubber (see Figure 2).



Fig. 4—Venturi scrubber and cyclone separator as installed.

Venturi Scrubber and Cyclone Separator. The venturi scrubber, approximately 48 inches in over-all length and with a throat diameter of 2-3/8 inches, was also constructed of stainless steel (Figure 4). To

provide for possible future needs, the venturi scrubber was actually designed by the Chemical Construction Corporation to handle 475 cfm of exhaust air with an expected pressure drop of 36 inches of water, but capable of operating at 320 cfm and 25 inches of water pressure drop with good air-cleaning efficiency. The throat velocities would be 15,420 and 10,390 feet per minute (fpm), while exhausting 475 and 320 cfm, respectively. It has been reported that throat velocities over 12,000 fpm are generally used in venturi scrubbers with pressure drops between 10 and 15 inches of water.² The design feed rate for the scrubbing solution, 1.0 N sodium hydroxide, was established at 6 gallons per minute (gpm), at 15 pounds per square inch pressure (psi), with the system exhausting 320 cfm. This rate of approximately 19 gallons per 1000 cfm is higher than the reported rates of 2 to 9 gallons per 1000 and accounts for the higher than usual pressure drop of 25 inches cfm, of water across the venturi scrubber.

The cyclone separator was not used as a cyclonic scrubber, although at one time such an arrangement was considered. A cyclonic scrubber in this system would have had a retention time of approximately one second, and it was thought that this factor would be important in absorbing the iodine vapors and oxides of nitrogen. The scrubbing solution, however, would have had to be fed to the cyclonic scrubber at a pressure of 100 psi. This would necessitate a separate pump or a high pressure system for both the venturi scrubber and cyclonic scrubber. It was decided, therefore, to convert the cyclone separator to a scrubber only if operating experience indicated it would be necessary.



Fig. 5—Caustic solution recycling and mixing tank installation.

Caustic Solution Recycling and Mixing Tanks. Since the installation is used on an intermittent basis, the capacity of the recycling tank (600 gallons) was designed to provide storage for sufficient caustic solution for the maximum anticipated run. The mixing and recycling facilities are essentially a standard installation for this type of equipment and are shown in Figure 5. A few special details, however, are the result of operating experience at the Los Alamos Scientific Laboratory. Leakage of radioactive liquids creates serious difficulties and consequently the pumps were mounted in trays equipped with proper drains, and a bypass was provided for the rotameter.

Fans. Two exhaust fans (U. S. Hoffman Machinery Corporation, Model 4202, TYPE EBA), each with a rated capacity of 350 cfm at 50 inches of water, were specified for the installation. The extra fan was installed as a safety measure, and the electrical system so arranged that an outage in either fan automatically causes the other fan to operate.

EVALUATION OF AIR-CLEANING SYSTEM

The effectiveness of the air-cleaning system was determined by sampling upstream and downstream from the scrubbing units for total fission products, acid mists, and iodine vapors.

<u>Acid Mists</u>. The concentration of acid mists was determined by back titration of the caustic solution used as the collecting medium in two large impingers in series. A limited number of tests indicated that the air-cleaning efficiency of the unit for the combination of nitric acid mist and oxides of nitrogen was apparently 90% with peak loadings of $2 \times 10^3 \text{ mg/m}^3$. In the case of perchloric acid, with peak loadings of $3 \times 10^3 \text{ mg/m}^3$, removal efficiencies of 95% were obtained. There is some mixing of the acids, but in the main the acid mists come off in two separate fractions. The efficiency for total acid mist removal for a complete run was approximately 92% with an apparent average loading of $1 \times 10^3 \text{ mg/m}^3$.

TABLE	II
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Run	Dissolving Stations Operating	Loadings mc/m ³	Air-Cleaning Efficiency %
l	6	0.3	94
2	5	0.1	95
3	4	0.05	94
4	4	0.01	91
5	6	0.4	96
6	4	0.15	92
7	3	0.05	93
		Ave	rage 94

Air-Cleaning Efficiency for Total Fission Products

<u>Total Fission Products</u>. Fission products were sampled from the air stream by means of a sampling train consisting of two large impingers with a caustic collecting solution in series and a high efficiency glass fiber filter paper. Aliquots of the collecting solution were evaporated to dryness on metal planchets. The radioactivity on the planchets and filter paper was determined by means of a gas flow proportional counter. The effectiveness of the system for total fission product removal is indicated by the results shown in Table II. The system was exhausting 320 cfm and the caustic solution was fed to the venturi scrubber at the rate of 6 gpm for all runs. <u>Radio-Iodine Vapors</u>. Sampling for radioactive iodine was accomplished by a modification of a sampling train developed by Claude W. Sill, A.E.C., Idaho Falls, Idaho.³ The train used in this study consisted of two high efficiency glass fiber filters, a Millipore membrane filter, and two Willson organic vapor respirator cartridges. Provisions for extremely high filtration efficiency are necessary to prevent trapping particulate matter in the cartridges. The second glass fiber filter was provided as a safety feature in the event the first filter, which is damaged by the acid mists, ruptured completely. Tests indicated that 99% of the iodine was collected in the first cartridge and no detectable iodine escaped through the second cartridge. Activity in the cartridges was determined by counting on a 2-1/2 inch NaI crystal counter, and identified as I^{131} activity by determining the radiological half life.

The efficiency of the scrubbing unit for removing iodine is indicated by the results in Table III.

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Run	Dissolving Stations Operating	Loadings		Removal Efficiency
		mc/m^3	µg/m ³	# #
1	4	0.03	2.4 x 10 ⁻⁴	97
2	4.	0.7	5.7 x 10 ⁻³	98
3	4	0.9	7.3 x 10 ⁻³	97
4	4	0.3	2.4×10^{-3}	96
5	6	0.05	4.0×10^{-4}	85
6	4	0.1	8.0×10^{-4}	93
7	3	0.03	2.4×10^{-4}	96
				Average 95

TABLE III

SUMMARY

The local exhaust collection system and the venturi scrubber installation was designed for the cleaning of exhaust air contaminated with acid mists and mixed fission products. It was determined that 20 cfm exhausted by a local slot exhaust hood would control the maximum evolution of gases from a 1500 ml beaker. Features of the exhaust system that were designed to offset the hazard of perchloric acid condensing in the system included welded stainless steel construction, sloping horizontal runs, installation of regulating valves only on vertical sections, and the ability to wash down the system after use.

The feasibility of a venturi scrubber with a caustic solution as the scrubbing medium for low loadings of iodine vapors (2.4 x 10^{-4} to 7.3 x $10^{-3} \mu g/m^3$) was indicated by an average removal efficiency of 95%.

Air-cleaning efficiencies for acid mists were dependent on the type of acid suspended in the air stream. Removal efficiencies of 90% were obtained with nitric acid and oxides of nitrogen loadings of 2×10^3 mg/m³, and 95% with perchloric acid loadings of 3×10^3 mg/m³. Total fission product loadings ranging from 0.01 to 0.4 mc/m³ were removed from the contaminated air with an average efficiency of 94%.

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- 3. Sill, Claude W.: Personal Communication (1957).

Discussion

C. (H. F. Johnstone, Univ. of Ill.) Passing suggestion out in facing difficult problems in the separation of gases which are unusual, rare and exotic in extremely small concentrations, don't overlook the possibility of using something that isn't in the text book. When you are dealing with very large quantities of gases or using something that isn't in the textbooks - that's a fluidized bed process. To our surprise we have found actual absorption of CO₂ in a fluidized bed and you can actually accomplish some gas chromatography in such a bed.

VENTILATION SYSTEMS AT ATOMICS INTERNATIONAL

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Abstract

A brief summary of ventilation systems employed on reactors, hot cells and critical facilities designed and/or operated by Atomics International is presented. The two power reactors discussed are contrasted by use of a rather loose building containment system in one (Sodium Reactor Experiment) and a very tight vapor container in the other (Piqua Organic Moderated Reactor). Similarly, of the two hot cells described, one operates with a comparatively large volume air flow, the other with a very low ventilation rate. Of the two remaining facilities considered, the Organic Moderated Reactor Critical Facility employs a somewhat unique dual ventilation system to avoid filter plugging by non-radioactive organic condensate. The Kinetic Experiment Water Boiler (KEWB) uses a sealed, static atmosphere during reactor operation with subsequent high volume flushing before re-entry of the test building.

Sodium Reactor Experiment (SRE)

Description

The SRE is a 20 Mwt sodium cooled graphite moderated reactor. The reactor is located in the Santa Susana mountains on the west end of the San Fernando Valley, approximately 900 feet above the valley floor. The reactor has been in operation approximately 2 years and has supplied power to valley communities through an Edison Company power plant.

The reactor and its heavily shielded heat transfer system vaults are located below grade and are housed by a 10,000 sq. ft. superstructure. The superstructure is a conventional type building of reinforced steel, 5.5" thick concrete tilt-up panels and has a poured gypsum roof. The building is not a pressure container, but is designed to be diffusion tight as long as no internal pressure exists. There is no stack associated with the main reactor floor area. These design features are permissible at the SRE since the reactor operates as a completely closed system, hence no radioactive effluents or radioactivity are released to the environs during normal operation except under controlled conditions (from the decay tanks through a stack). A helium atmosphere is maintained over the sodium in the reactor vessel and a nitrogen atmosphere is maintained in the vaults containing the heat transfer system. Approximately 15,000 cfm of fresh air is supplied to the high bay area through Farr Air Cleanable Filters and unit heaters. An estimated 24,000 cfm of air is exhausted directly through two power roof ventilators designed to maintain a 1/8" H₂O negative pressure in the high bay area and to provide approximately 2 fresh air changes per hour. Standby MSA Ultra Aire glass filters have been added on the downstream side of each exhauster and may be cut in during an emergency, if needed. These were added to filter potential releases of activity from fuel handling operations or other maintenance operations involving contaminated sodium. These filters are by-passed during normal operation. The service and administration area is attached to the reactor building. This area is maintained at a positive pressure by fresh air supply fans and is exhausted directly through wall and roof louvers. The positive pressure insures against potential leakage of radioactivity from adjacent reactor building areas. The offices and control room are air conditioned to insure reliable operation of instrumentation and for personal comfort in hot weather.

The reactor and primary sodium system are operated at a low pressure and are contained in a closed system. The reactor top shield, which can be rotated to remove large core components, employs a cerrobend seal to prevent leakage of reactor cover gas. Fuel access plugs, located in the top shield, use double o-ring seals to prevent gas leakage to the reactor room. Welded stainless steel construction is used in the remaining parts of the system. The helium blanket gas of the reactor is normally maintained at a pressure of about 3 psig and no gases are released during normal operation. Four shielded decay tanks are provided to store fission product or other radioactive gases in the event the system must be emptied or vented. The four tanks have a capacity of 10,800 standard cubic feet at 100 psig. Activity may be bled from the tanks, under controlled conditions, to a stack which is equipped with a 25,000 cfm dilution fan. This stack discharges above the SRE roof. Cambridge absolute type filters (8" x 8") are provided before and after the decay tanks to remove particulates. Because of possible sodium hazards the primary heat transfer and piping system is contained in concrete vaults containing an inert atmosphere (nitrogen). The system is designed to operate at a slightly positive pressure to exclude all oxygen. In the event of airborne activity, the vaults can be vented to the gas hold up system through absolute filters.

Experience

Sodium exposed to moist air ignites and burns. Contact of the smoke or hydroxide mist with the throat and lungs is very irritating, however, protection is easily afforded by a number of different types of breathing equipment.

In the two and one-half years of a vigorous experimental program at the SRE, no lost-time injuries have occurred due to sodium handling. Small sodium and NaK fires have taken place, but these have all been controlled successfully and no significant activity has been released to the environment. Leaking valves tend to plug themselves by the sodium freezing and clean-up of spilled sodium has been accomplished using direct methods (e.g., shovels and buckets).

To date, about 2,400 Mwd of operation have been accumulated, with the production of approximately 15 million kwh of electricity. The primary sodium is usually maintained at about 950°F. Higher temperatures have been attained and steam has been produced at 1000° F on occasion.

SRE Hot Cell

Description

The SRE building contains a hot cell area consisting of two cells, an operating area in front and a personnel air lock leading to a service area in the rear.

The cells are equipped with three windows and six manipulators. These cells are used for inspection and disassembly of SRE fuel elements and components, and for non-destructive testing of fuel. Potential radiological hazards in the cell may arise from sodium, NaK and fission products.

Since the hot cells are not designed to be gas tight, a conventional high volume air system is used to ventilate the cells. Raw outside air is drawn into the cells through normally open isolation dampers and a pre-filter. If the exhaust fan is denergized, the dampers close automatically to isolate the cell. The cell atmosphere is exhausted through American pre-filters (hood-type) located in the cell and three parallel Cambridge absolute filters, which are located outside of the cell. The absolute filters are followed by two 10 hp blowers (one standby) which discharge the air to a dilution stack, exhausting above the SRE roof level. Each blower is rated for 2400 cfm at 15" of water. During normal operation a negative pressure of approximately 0.25" of H₂O is maintained in the cell. When the service door is open, a conventional air flow of 100 fpm is maintained across the opening to prevent contamination of the service area. A 25,000 cfm fan located at the base of the stack dilutes the normal cell exhaust and discharges it above the SRE roof level. Service and operating area ventilation is discharged to the stack through Cambridge absolute filters (3500 cfm). Any leakage from the cell is therefore filtered before being discharged to the environment.

One of the cells is provided with a vent line leading to the SRE gas decay tanks in the event experiments are planned which could release hazardous quantities of fission product gases. Fire fighting equipment is installed in the cell.

Experience

The cell has been operated successfully for approximately 2 years. Two fires involving NaK, alcohol and Na^{2l_1} have occurred in the cell during handling of irradiated fuel. Interior fission product contamination of the hot cell occurred on these occasions, but no significant contamination was discharged to occupied areas or the environment.

Component Development Hot Cell (CDHC)

The CDHC is a structure, owned, designed and operated by Atomics International in the Santa Susana mountains in California. It contains four large hot cells which have a combined floor area of 820 sq. ft. (metallurgical, physical test, fuel disassembly and reactor component disassembly cells). Each cell leads to a separate shielded decontamination cell in the rear, which in turn leads to a common service area behind the cells. This design was chosen so as to provide improved control over contamination. A common operating area in front contains nine windows in the cell block, and provisions for 18 manipulators, periscopes and other related viewing equipment.

The ventilation system which is employed is unique for hot cells. The unusual design was necessary because of the hazardous nature of the operations planned for the facility (handling Na, NaK and disassembly of spent fuel). Basically, it is a low volume system employing sealed cells with an exhaust gas hold-up and decay system. The interior is surfaced with welded sheet steel and openings are sealed with gaskets and/or inflatable seals. Normal operation is at 5 cfm per cell, the cells being maintained at a negative pressure of $\frac{1}{4}$ " water. The cells can be filled and operated with an inert gas to accommodate special experiments requiring inert atmospheres. The cell exhaust is routed through Cambridge pre- and absolute filters (8" x 8") to a dilution stack or to a gas holdup system storing gases at 150 psig, if gaseous activity is present. The gas storage tank system is sized to keep one cell in operation for 3 days (5 cfm for 72 hours).

When the cell service doors are open, a conventional high volume (7200 cfm) system is employed to discharge the exhaust to the stack via pre- and absolute Cambridge filters (24" x 24" filters). The system is designed to provide a conventional air flow of 100 fpm through the opening with two of the service doors fully open. A 25,000 cfm dilution fan is provided at the base of a 65 foot stack to provide safe discharge of gases from the decay tanks.

Inflatable seals are used to hold the absolute filters for both the high and low volume exhaust system in place. New filters are installed by (1) placing a new filter in a horizontal duct on one side of the unit, (2) releasing the pressure on the inflated seal, (3) applying manual pressure to the new filter to force the old unit into a duct and plastic bag located on the opposite side and (4) inflating the seal once the new filter is in its proper location. This design provides a convenient and safe means of removing, handling and disposing of contaminated filters.

Organic Moderated Reactor Critical Facility

This facility is located in the Santa Susana mountains in California and is used to obtain nuclear parameters for organic moderated reactors. Basically, it consists of a large carbon steel tank containing fuel elements in a heated organic pool which is maintained at atmospheric pressure. The tank lid consists of loosely fitting pieces which can be readily removed to make necessary experimental changes. The reactor is contained in a section of the building which is isolated from the control room and administration area by concrete shield walls. Access to the reactor room is through a hallway and opening at one end of the shield wall. Sealed doors at each end of the hallway are kept locked when the reactor is on. Two ventilation systems are provided for the reactor room - a "reactor off" system and a "reactor on" system.

When the reactor is off, raw outside air is drawn through wall louvers and is exhausted directly through four roof mounted power exhausters equipped with motorized dampers. Exhaust filters are not used during this period since there is no danger of airborne radioactivity with the reactor off. If filters were continuously in use, the organic fume and condensation would require filter replacement at least daily. During shutdown periods a high room air change rate (8000 cfm) is employed for removing radiated heat and terphenyl fumes which escape from the organic system. The room may be occupied only when the reactor is off.

Interlocks require that the roof exhausters be turned off and the exhaust dampers closed prior to reactor start up. The intake louvers are spring loaded, equipped with fusible links and are tied into the scram network with a solenoid latch. These intake louvers close in the event of fire or a scram to reduce the air flow and pressure in the reactor room.

When the reactor is on, 3500 cfm of exhaust air is routed through a separate system. Components of this system consist of a local exhaust duct leading from the reactor atmosphere, an organic trap, a room exhaust grill mounted above the critical assembly, a filter system and a bypass system, a 7.5 hp exhaust blower and a stack which extends 10 feet above the roof level. The exhaust duct leading from the reactor atmosphere is routed through an air-cooled organic vapor trap. The terphenyl mixture melting point is 293° F. A 3 scfm CO₂ purge within the organic system expels fumes from the core tank through this exhaust duct and trap and is finally exhausted immediately in front of the room exhaust grill located above the critical assembly. This duct leads to two banks of fireproof pre- and absolute filters located just upstream from the blower (four 24" x 24" absolute filters in each bank). A water sprinkling system is located in the duct work to extinguish possible organic fires.

Normally, the above filters are bypassed to prevent their becoming clogged with organic fumes. A stack monitor continuously monitors the exhaust for radioactivity content. In the event of high activity, the reactor is automatically scrammed and fail-safe motorized dampers divert the exhaust through one of the two filter banks and automatically close the building intake shutters. The second filter bank is maintained as a reserve to be used in the event of clogging of the first bank. Manual operation of the dampers from the control room is also possible.

Kinetic Experimental Water Boiler (KEWB)

The KEWB is located in the Santa Susana mountains west of the San Fernando Valley. This facility is used for investigating the safety of solution type reactors by employing planned power excursions. The test building containing the reactor is buried in the ground and is controlled from a remote control station. The first test core has a nominal power rating of 50 kw.

The test building is unoccupied and sealed during all reactor runs. A continuous ventilation system was not provided since planned excursions are relatively infrequent and hazardous levels of air activity are not expected during normal tests. The interior atmosphere is continuously sampled by a re-circulating gas monitor. Substantial A⁴¹ concentrations are present in the test building following large power transients. Release of some Xe and Kr activity has occurred in the test building upon occasion. Before re-entry to the building, the intake vents are opened and a blower rated at 2000 cfm is turned on to discharge the A^{41} ; the exhaust air leaves through two 24" x 24" absolute filters and a 60 foot stack. If excessive airborne contamination is detected by the air monitor, the room remains sealed to allow decay and/or is vented at a controlled rate through the absolute filter system. Control is obtained by a manually operated butterfly valve in the exhaust system which allows the admittance of dilution air to the stack during this venting. When this valve is opened, the rate of air discharged from the facility is considerably reduced. This procedure continues until access is possible.

Piqua Organic Moderated Reactor

Reactor Description

The Piqua reactor is a 45.4 Mwt organic cooled and moderated reactor presently under construction at Piqua, Ohio. The plant will be integrated with the existing municipal city power plant, in order that the present plant may continue to function as a conventional plant.

The reactor plant consists of a reactor building and an auxiliary building. The reactor building consists of a 73 foot diameter steel vapor containment shell half buried in the ground; the above grade portion is shielded by 18 inches of ordinary concrete. The shell houses the reactor, heat transfer rooms, fuel storage pool and reactor room. The reactor floor is at grade level whereas the heat transfer equipment is located in shielded rooms below the floor level. Normal organic operating temperature and pressure are 575°F and 120 psig, respectively. The auxiliary building contains reactor services, coolant purification and waste systems, a control room and an administration area. The superstructure is of conventional industrial design with a concrete shielded basement containing the process rooms. For convenience the ventilation systems of interest can be divided into the following categories:

- (1) reactor building ventilation

- (2) auxiliary building ventilation
 (3) process off-gas system
 (4) waste fired burner exhaust system
- (5) reactor fume hood

Reactor Building Ventilation

During normal operation fresh air is supplied to the reactor building through fiber glass media filters (Rollamatic) by a 40 hpblower (\checkmark 33,000 cfm) and is exhausted through fiber glass pre- and absolute filters, to a 90 foot stack using two 25 hpfans. The stack discharges at a height which is 20 feet above the top of the reactor building. The supply fans are located in the containment shell and are used for cooling the reactor building atmosphere in the event the normal ventilation system is interrupted. The exhaust fans are located outside the containment shell to save space. The reactor room is ventilated at the rate of two air changes per hour (approximately 10,000 cfm). Because of heat loads and a higher contamination potential, the basement rooms and heat transfer rooms located below the reactor floor are ventilated with a minimum of ten air changes per hour.

For basement rooms the pre-filters are installed at the outlet of each room to trap potential organic particulates and prevent contamination of the exhaust duct system.

The containment shell is equipped with isolating valves in the ventilation ducts which close if hazardous radioactivity levels are present. When the isolation valves close, the reactor atmosphere is re-circulated through the supply fans and a cooling system to prevent over-pressure on the containment shell. A vacuum breaker valve is provided to protect the shell against excessive underpressure. The containment shell is designed for 5 psig positive and 0.5 psig negative pressures, and for a maximum leakage rate of 0.2% of the total volume per psi per 24 hours.

Auxiliary Building

The auxiliary building ventilation system consists of a 10 hp supply fan delivering $\sim 23,000$ cfm of air to the building through pre-filters and an air heating coil. Pre-filters are provided on the exhaust of all process rooms (filter located in the room) to prevent accumulation of organic material and contamination in exhaust ducts. One 20 hp exhaust fan draws air through absolute filters and discharges 22,000 cfm of air to the main stack. The flow pattern is always from areas of no contamination to areas of higher contaminated areas. The total volume normally exhausted from the facility stack from both the reactor building and auxiliary building is $\sim 55,000$ cfm.

Process off-gas System

During normal operation a stream of organic coolant is withdrawn from the reactor vessel, filtered, degassed and returned to the reactor with pressurizing pumps. This permits continuous removal of the small volume of gas formed by organic decomposition and also allows continued operation with minor water leaks and fuel cladding failures.

The decomposition gas flow rate from the degassifier is approximately 2 sofh and contains hydrogen, methane, and other light hydrocarbons. Water vapor and volatile fission products may also be present. This stream is routed through two activated charcoal absorbers to remove organic material, and a condenser to remove steam. The gas stream then flows through a waste gas decay system consisting of a series of tanks which is capable of delaying release of gases for a period of up to 48 hours. This amount of decay permits reactor operation to continue with minor fuel cladding failures. Continuous discharge of these gases to the stack through absolute filters is employed. In the event of a serious fuel rupture, wherein 48 hour holdup is not sufficient, the reactor will be shut down.

Waste Fired Burner

The coolant purification system continuously removes high molecular weight hydrocarbon components from the coolant by a vacuum distillation process. After sufficient radioactive decay, this organic waste is injected into a waste fired boiler and burned. In the present system, storage tanks are provided to hold the waste coolant for about 5 months before it is burned. If the radioactivity concentration in the organic wastes, after the 5 month decay period, is too high to burn safely the wastes can be packed in drums and stored or shipped off site for disposal. The development of methods for further improving the removal of particulate and gaseous activity from the combustion gases in order to permit burning the organic wastes with shorter decay times is currently underway at Atomics International. Present plans include a bag type pre filter and an absolute filter for removal of particulates from these combustion gases. Approximately 150 cfm of room air is mixed with the 250 cfm of air from the burner for cooling purposes. A fan discharges this air stream to the stack where it is diluted with the building ventilation.

Reactor Fume Hood

When the reactor top lid is removed, local ventilation is provided by a fume hood which prevents the escape of organic fumes to the reactor room. The fume hood uses lateral exhaust and is designed into the fixed shielding at the reactor top. Approximately 10,000 cfm of reactor room air is drawn into the opening at the top lid and is exhausted through a horizontal slot around the periphery of the opening. To remove organic material the air is filtered by an automatic filter unit consisting of a renewable roll type filter media with a leno-weave backing (similar to Farr Company Roll-Clean Model). The cleaned air is routed to the reactor building ventilation exhaust system which leads to the absolute filters and the stack.

Discussion

- Q. (J. F. Newell, AEC Washington) If they have any operating experience on the quantities of fission products that are -well, what is your experience with leakage from fuel into the sodium coolant and how is it retained in the coolant?
- A. They have had experience in Idaho with fairly serious ruptures. Don't know exactly where all these fission products are at the moment.
- Q. You mentioned burning of the residues from your organic were these contaminated residues?
- A. I think I can answer your question now. I had mentioned that we could run this reactor with a one centimeter area of uranium exposed. Normally the concentration in the organic primary loop is less than we might care to receive. With a one centimeter exposed fuel area the concentrations from fission products may be a factor much less than that - this gives no problem. The xenon, krypton and iodine would come off immediately in the gas system and go the stack with out hold-up. Whereas, the organic that is burned can be held up to a period of 6 months - these activities have been based on cooling rates, etc...
- Q. In the reactor they would be able to run for 48 hours after exposure - with the 10⁻⁹ mc to the top of the stack. Do you
really believe they would shut the reactor down if it reached 10⁻⁹ microcuries - or would they use some dissolution factor to the ground plus a long term average of concentration at the ground as is done everywhere else?

- A. A 48-hour period gives an opportunity to search for the ruptured fuel element - the trend is toward shorter and shorter stacks -AI is limited due to proximity to the city.
- Q. (F. T. Sellick, FLuor) Relative pressure on gas decay system or tanks - is this a pressurized gas or is it induced into a sphere by a vacuum?
- A. It is pumped in by pressurized pumps.
- Q. (W. Culkowski, Oak Ridge Weather Bureau) This pressure for shorter and shorter stacks, where is this pressure coming from?
- A. The committees that reviews the hazard summaries reports brings up items like this. First the question of criteria - what was our criteria on the stack height. I think the first stack was 125 feet. It's written that they are going for shorter stacks. I understand that from talking to someone the other day it was mentioned that the English consider one person dies for every hundred foot of stack length that is built.
- C. (A. Wolman) I might say that as one interested at least in some detail on AEC policy, I am a little astonished at Mr. Piccot's comments that AEC now, among other policies, decisions are moving toward no stacks at all. I have some doubts about that.

SUMMARY OF AIR CLEANING ACTIVITIES AT CHALK RIVER

J. NEAL

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I have been asked to say a few words about our experience in air cleaning at the Chalk River Plant of the Atomic Energy of Canada Limited. I do not have a prepared paper with me, however, I will try to tell you as much as I can in the few minutes at my disposal.

On December 12, 1952, our N.R.X. reactor went out of control and the reactor was badly damaged. At that time the only filters on the exhaust air system from the reactor building were of the deep bed roughing type. Fortunately the wind direction was such that the air-borne radio-activity released was spread over a large uninhabited area. The other buildings in the plant were not seriously contaminated.

Following the successful removal of the damaged reactor vessel and the installation of a new unit the reactor was ready to go back into operation. Before start-up it was decided to install absolute filters in addition to the existing deep bed roughing filters. Thirty-two 1000 CFM filters were installed. These were installed in four plenum chambers, each chamber having a bank of eight absolute filters installed against a vertical frame.

After start-up it was noticed that the amount of radioactivity passing through the filters was greater than would be expected if the filters were working at their rated efficiency of 99.97% retention of 0.3 micron size particles. At the next shut down of the reactor an inspection was made of the absolute filters. It was found that some had been damaged in installation. There were obvious bruise marks on the filter media. Also the filter retaining frames were so rough, particularly at the welds, that there was considerable edge leakage past the filters.

It was decided that all installations of filters of this type should be inspected and approved by a small group. This group is made up as follows:

- (a) One man from the building requesting the filter installation.
- (b) One man from the Maintenance Branch responsible for the installation work.
- (c) One man from the Radiation Hazard Control Branch.

This system has been in use for some years and has increased the overall efficiency of this type of filter installation.

Following the 1952 incident to N.R.X. reactor a complete survey was made of all air exhaust outlets in the plant. It was decided to install absolute filters on all outlets where either radio-activity was being released or where there was this possibility. It was also decided to form a Ventilation Specification Panel which would be responsible to examine all proposed ventilation installations and either approve or reject them. This panel is made up of senior representatives from the Design Branch, Maintenance Branch, Research Division, Operations Division and Radiation Hazards Control Branch. They submit their recommendations directly to senior management.

This Panel has made some recommendations which are now company policy. For example, because of the difficulty of ensuing a tight installation when installing a number of absolute filters in a plenum frame, all new installations are made with filters which are enclosed in a box to which are fitted an inlet and outlet nipple. These filters are of fire-proof media. The containing boxes are made fire-resistant by chemical treatment. The D.O.P. test is made at the supply company's plant before shipping, and the efficiency across the filter from nipple to nipple is marked on the On arrival at the Chalk River plant they are checked by a box. Radiation Hazard Control Branch inspector for any obvious damage or flaws in the filter media. They are stored with the spacers in the vertical position to avoid any possibility of sagging of these spacers occuring whilst in storage. When a multiple filter installation is made the main ducts before and after the filters are fitted with the required number of nipples to accommodate the filters. All filters are visually checked before installation using strong lights and are approved by the inspecting group previously mentioned. After the unit goes into operation air samples are taken before and after each filter and checked for radio-activity. Any filter not working to its rated efficiency is removed and replaced with a new filter. This is easily done as the inlet and outlet nipples are connected to the ductwork by rubber sleeves.

Some previous speakers have mentioned receiving a considerable number of defective filters. This to date has not been the case at Chalk River. However, on my return to the plant I hope to install a stricter inspection of all absolute filters received. The Defence Research Chemical Laboratories are located in Ottawa. They have a D.O.P. testing unit and as they have always been most cooperative with A.E.C.L. I feel that arrangements could be made with them to inspect some of our incoming absolute filters to ensure that they are being manufactured to specification.

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HIGH-EFFICIENCY, HIGH-VELOCITY ELECTROSTATIC PRECIPITATORS

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The protection of personnel both ashore and at sea against objectionable or toxic aerosols is important today. A part of our work in this field at the Naval Research Laboratory has been an investigation of electrostatic precipitation.

Laboratory studies of electrostatic precipitation were initiated at NRL in 1949. The objective of this work was to determine if the aerosol removal efficiencies of standard, commercial, ventilation-type precipitators could be improved. It was hoped that efficiency-wise, they could be made competitive with paper filters while still retaining their great advantage of low air resistance.

At that time (and it is still generally true today) commercial, ventilation precipitators operated at linear air flows of about 300 feet per minute and had DOP aerosol penetrations of about 15%. The units used for these early studies were two-stage precipitators. They consisted of an ionizer section of high voltage wires separated by ground tubes while the collector section was a set of parallel plates with alternate plates at ground potential. We refer to this design as the two-stage plate type.

It was shown that dramatic improvement in performance of the commercial two-stage plate precipitator could be achieved by rather simple modifications. The first alteration included the liberal use of gaskets and seals to insure that no air by-passed the active zones of the precipitator. The effectiveness of the sealing was determined by the effect on the overall penetration of DOP aerosol. These penetrations were measured, using the NRL E-3 Light-Scattering Meter. The location of a leakage source was detected by the use of a sampling probe connected to the same meter.

The second alteration which improved the performance was the use of pre-filters of crimped screen. These filters provided a more uniform air flow through the precipitator which insured that the total cross section would operate with equal effectiveness. In addition, these smoothing filters assisted in the removal of large particulates (over 10μ) from the air stream. Representative data from these studies are contained in figure 1. We have listed here the average DOP aerosol penetration obtained from the commercial units at air flow of 200 feet per minute. Included also is the DOP penetration at the same flow rate for the modified unit. It is readily apparent that there is more than a two order improvement due to the gasketing and smoothing of the air flow in the modified unit.

Туре	Linear Flow Rate (F PM)	DOP Penetration (%)
Commercial	200	15
Modified	200	0.1

Fig. 1—DOP penetration of commercial precipitators.

Encouraged by these results, NRL initiated a program to investigate electrostatic precipitation more extensively. The program was sponsored by the Bureau of Ships.

The first phase of this program was the feasibility studies of precipitator performance aboard ship. At that time it was yet to be proven that precipitators could be used in a naval environment. For this purpose sixteen commercial units modified to NRL specifications were procured. These units after alteration were found to have a DOP penetration of 0.5% at 300 feet per minute. The units were installed aboard the light cruiser, USS ROANOKE, in auxiliary plenum chambers attached to the superstructure of the ship. Eight units were contained in an 8000 cfm vent system while the other eight were used in a 10,000 cfm system.

Hours Service	Aerosol_Penetration_==_%		
	Fwd. System	After System	
0	0.4	0.5	
4 000	0.8	0.2	
8000	0.7	0.1	

Fig. 2—DOP aerosol penetration at 300 fpm of precipitators aboard USS Roanoke.

After 8000 hours operation the units were removed as it was apparent that adequate information had been obtained. Figure 2 contains data taken at the time of installation, after 4000 and after 8000 hours of operation. The differences are not considered significant. It is to be noted that during this time there were no major component failures. Shock, vibration, salt air and corrosion did not limit their usefulness. These results are perhaps more impressive in that they were obtained with precipitator units obviously not designed for naval service. The conclusion was thus reached that it would be practical to use precipitators aboard naval ships.

The second phase of the precipitator program consisted of R and D contracts for basic and developmental studies. The ultimate goal of these contracts was to obtain prototype models designed for naval service. Since laboratory studies had shown that increased linear flow rates were possible, the models were to operate at maximum linear flow rate and



Fig. 3—Prototype model high-velocity, high-efficiency precipitator, 1500 cfm.

provide aerosol penetration less than 0.1%. Pressure drop was not to exceed one inch of water. A limit of 0.1 part per million of ozone was set as maximum allowable.

The design which came closest to meeting all these requirements was that of Research-Cottrell of Bound Brook, New Jersey. It is a two-stage tube-type unit. Two-stage plate-type units were found to be inferior for high-velocity use. A photograph of the tube-type model is shown in figure 3. Figure 4 is a schematic showing the main components. The ionizer which operates at voltages up to 40 KV



Fig. 4—Prototype model high-velocity, high-efficiency precipitator, 1500 cfm schematic.

is of wire and plate design. The collector contains nested ground tubes with high-voltage, centered rods. It operates at 20 KV maximum. The unit is complete with vaned turns at top and bottom. It also includes a set of spray nozzles for remote-controlled washing with hot water or steam. A drain is provided in the base. The requirement for remote washing was included to provide cleaning of the unit without physical contact in the event the collected material was hazardous.

An interesting and important result of the analysis of all the performance data reveals that the limiting factor in achieving optimum performance is the desired ozone level. The production of ozone (or total oxidants) is directly related to the coulombs of high voltage current generated. But as this also affects the charging of aerosols, setting a maximum for ozone and simultaneously setting a value of 0.1% penetration for aerosol in effect sets the values for all other variables.

Thus, although this particular unit can achieve an efficiency of 99.9% at 2000 feet per minute with pressure drop of 0.9 inches of water, the ozone produced is more than 0.1 part per million. It has thus been necessary to rate this model at 1500 feet per minute.



Fig. 5-High-velocity, high-efficiency precipitator, 9500 cfm.

A large unit of 9500 cfm capacity and based on the same design was also procured from the manufacture. A photograph of the unit is shown in figure 5.

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Both of these units have received extensive evaluation; the smaller unit was run for 18 months ashore before installation in the ship which also contains the larger unit. There has been no difficulty with either unit and both have given completely satisfactory service.

Unit	Air Flow		DOP Penetration	P	
	FPM	CFM	(%)	(in H ₂ 0)	
Small	1500	1500	0.07	0.5	
Large	1500	9500	0.02	0.6	

Figure 6 contains the basic performance data as obtained from these units. We have listed here the aerosol penetrations at rated flow of 1500 feet per minute.

Fig. 6 — Performance of high-velocity precipitators.

Figure 7 is a summary which illustrates the great improvement which has been achieved during the past ten years in the performance of ventilation type electrostatic precipitators. Air flow rates have been increased by a factor of eight while DOP penetrations have been reduced by a factor of 300.

Туре	Linear Air Flow (FPM)	Average DOP Penetration (%)
Commercial	200	15
Modified	200	0.1
High Velocity	1500	0.05

Fig. 7 — Summary of electrostatic precipitators performance.

In conclusion, it has been shown that compact, highvelocity, high-efficiency precipitators can be manufactured for naval service. It has also been proven that they can be used aboard ship with full confidence of satisfactory performance.

THE USE OF IODINE AS AN INDICATOR FOR REACTOR MONITORING

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ABSTRACT

Radioactive iodine is a positive indicator of reactor fission products and distinguishes between these and neutron activation isotopes which are of far less importance biologically.

A comparatively simple method for collecting iodines is the use of filter paper or filter tape treated with silver nitrate to give Ag^O and Ag⁺. A treated tape lends itself to a continuous fission product monitor; the use of activity read-out and gamma energy discrimination can easily be employed.

Laboratory evaluation of silver nitrate treated paper gave a 95% collection efficiency for gaseous iodine. Iodine collection efficiencies varying from 40-95% were characteristic of treated paper when the Chemical Processing Flant was used as a source. These erratic results were thought to be due to part of the iodine entering into a complex chemical form less susceptible to collection by the silver iodide treated paper.

Using an air cooled reactor as a source the treated paper collected over 55% of the gaseous iodine. The collection efficiency for total iodine - gaseous and iodine acting as particulate - was over 95%.

Introduction

The presence of radioactive iodine in a reactor effluent is positive evidence of fission products in the effluent. Since iodine is one of the earliest fission products to escape the cladding, its use as a fission product indicator has merit¹. Activation products such as Argon-41, erosion products that have been neutron activated, and others may give radioactivity in the effluent but these are of far less biological importance and do not forewarn of increased future activity.

The use of a treated tape for the collection of gaseous fission product iodine would have considerable merit since the tape could be moved at regular intervals. The tape could also be read for activity with an energy discriminator if required. From previous experience and discussions with others² it was expected that a portion of the iodine would plate out on dust particles and act as a particulate. The amount of this plating would depend on the dust concentration and contact time.

<u>Method</u>

The evaluation of the removal efficiency of treated paper against iodine was checked using the following methods:

- 1. Laboratory generated I-131 gas
- 2. Chemical Processing Plant used fuel
- element dissolution vapors
- 3. Effluents from an air cooled reactor

It was assumed that activated carbon had one hundred per cent removal efficiency against iodine³; therefore, the treated paper - AgNO3 treated to give Ag^{O} and Ag^{+} - were placed ahead of activated carbon to give a quanitative test.

Equipment

Two arrangements of sampling equipment were used. The first arrangement used a Hi-Vol, sampling at 12 c.f.m. or 182 f.p.m. face velocity (see Photo I). The filtering media were arranged in series as follows:

> Untreated CC-6, labelled #1 Treated CC-6, labelled #2 Carbon chamber filled with activated carbon labelled #3 and #4

The second piece of equipment (see Photo II) sampled at 2 c.f.m. or 30 f.p.m. face velocity. In this equipment the following filter media are in series (see Photo III):

Untreated CC-6, labelled #1 Untreated CC-6, labelled #2 Treated CC-6, labelled #3 Treated CC-6, labelled #4 Treated CC-6, labelled #5 Activated Carbon cartridge, unlabelled and in background

In evaluating the efficiency of treated paper for iodine removal in the laboratory, Oak Ridge I-131 was titrated into warm concentrated nitric acid. A pre-filter was used for moisture removal.

In the use of the air cooled reactor as a source, a mobile unit (see Photo IV) was used sampling downwind at not less than one mile distant from the reactor effluent stack.

Test Results

The removal efficiency of the treated paper under laboratory conditions is over 95% as shown in Table I:



Photo I



Photo II

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Photo III

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Photo IV

Sample <u>Number</u>	Sample Description	Disintegrations Per Minute	Distribution of Activity
1	lst Pre-filter	92,418 ± 485	
2	2nd Pre-filter	33,748 ± 300	
3	lst AgNO3 treated filter	372,035 ± 968	96.22%
4	2nd AgNO3 treated filter	2,445 ± 108	.62%
5	3rd AgNO3 treated filter	1,438 ± 95	•37%
6	lst Carbon Sample	10,678 ± 170	2.76%
7	2nd Carbon Sample	108 ± 52	•03%

Table I

The removal efficiency of the treated paper using the Chemical Processing Plant fuel element dissolutions as a source was found to be unsatisfactory. The major percentage of the iodine released under this condition was either in the form where it acted as particulate or combined chemically in a form which inhibited collection on silver treated paper. Collection efficiencies varied between 40 to 95% for iodine, gas or vapors.

Using the air cooled reactor as a source, and sampling at over one mile from the stack allowed all the meteorological factors to come into play4. In all cases the first fission products to be identified were the iodine isotopes. In most cases the major portion of iodine was suspected to be in the particulate form or attached to particulates⁵. This particulate was removed by the untreated CC-6 paper. The gaseous iodine was then collected by the treated paper or by the activated carbon. Table II gives the results from two field tests. Data gathered after these dates gave similar collection efficiency for iodine but is not included here due to time limitations.

Table II

Test May 15

	<u>Hi-Vol W</u>	Hi-Vol Y	Low Vol. Sampler
Untreated CC-6	48,158 cpm	48,825 cpm	5,838 cpm
Untreated CC-6	not used	not used	0 cpm
Treated CC-6	7,143 cpm	5,540 cpm	443 cpm
Treated CC-6	not used	not used	683 cpm
Treated CC-6	not used	not used	391 cpm
Activated Carbon	790 cpm	1,952 cpm	0 cpm

Test May 19

	<u>Hi-Vol Y</u>	Hi-Vol S	
Untreated CC-6	9,957 cpm	7,247 cpm	
Treated CC-6	1,467 cpm	846 cpm	
Activated Carbon	296 cpm	278 cpm	

<u>Notes:</u>

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All the above counts are gross gamma and are to be used for comparative purposes. Statistical variations are not given since they serve no useful purpose in this instance.

In the sample Hi-Vol Y from May 15 it is suspected that the treated paper may not have been seated properly and allowed activity to filter through to the activated charcoal.

Conclusion

The use of iodine as an indicator for reactor monitoring is feasible and can be accomplished by employing a material which has a high collection efficiency for this element.

A continuous treated tape has merit since the background activity can be reduced to a low level and energy discrimination can be employed in the read-out.

Collection efficiency of the silver nitrate treated filters is high using an actual operating reactor as a source.

Further evaluation of materials and equipment is in process to supply the necessary empirical data in sufficient quantity for confidence.

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