UNCLASSIFIED

OAK RIDGE NATIONAL LABORATORY

Operated By

UNION CARBIDE NUCLEAR COMPANY

UCFPOST OFFICE BOX P
OAK RIDGE, TENNESSEE

External Distribution

Authorized

ORNL
CENTRAL FILES NUMBER

CF-58-5-59

DATE: May 21, 1958

COPY NO. 66

SUBJECT: Proposed Method for Removal of Radio-Iodine Vapor from Experiment
Off-Gas System of the ORR

TO: Listed Distribution

FROM: R. E. Adams and W. E. Browning

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Abstract

Various methods of removing radio-iodine from air streams have been surveyed for possible use in removing radio-iodine accidentally released from reactor experiments into the experiment off-gas system of the ORR. Solid adsorber materials appear to be best suited for this purpose. An iodine decontamination factor of 2000 was experimentally determined for activated charcoal and Linde Molecular Sieve Material (13X) using I^{131} to simulate mixed radioisotopes of iodine. A conceptual design for an iodine trap containing activated charcoal is presented. The iodine removal efficiency of this hypothetical trap is discussed under conditions resulting from discharge of 3.0×10^4 curies of mixed radioisotopes of iodine into the ORR experiment off-gas system.

NOTICE

This document contains information of a preliminary nature and was prepared primarily for internal use at the Oak Ridge National Laboratory. It is subject to revision or correction and therefore does not represent a final report.

UNCLASSIFIED

520 001

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

- A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
- B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission to the extent that such employee or contractor prepares, handles or distributes, or provides access to, any information pursuant to his employment or contract with the Commission.

Don't
Start

I. Introduction

In-pile experiments containing fissionable material in the ORR accumulate very large amounts of radioactive fission products. Such experiments are designed so that these radioactive fission products are contained completely, or, in case of accidental rupture of the experiment container, released through a special off-gas system to a gas disposal stack to prevent contamination of the interior of the reactor building. Preliminary estimates have indicated that if the radio-iodine from all the various experiments in the ORR were suddenly released into the atmosphere through the off-gas system by accident, the radiation dose levels received by personnel over a large area of the Laboratory would exceed acceptable limits. The non-volatile fission products may be removed from the off-gas stream by an ordinary air filter but volatile fission products would be expected to pass through such a filter. At the request of the ORNL Operations Division, this study was undertaken to evaluate the possible methods for removing radio-iodine from the experiment off-gas system of the ORR.

This off-gas system will serve to dispose of cooling air from the experiments and to remove any fission products accidentally released. Air flow rates up to 1000 cfm are anticipated. To guard against possible release of large amounts of fission products into the gas disposal stack this off-gas stream must be filtered and cleaned at all times during operation. Therefore, an effective off-gas decontamination system must be capable of handling large volumes of uncontaminated air for long periods of time and still remain in readiness to remove radio-iodine and non-volatile fission products accidentally released into the air stream.

Various methods for removing iodine activity from an air stream have been reported. The methods may be grouped into these general categories: (1) gas scrubbers using liquid adsorbants, (2) high temperature silver or silver salt reactors, and (3) adsorption systems utilizing various solid adsorber materials. In situations where the gas flow is moderate and iodine contamination is continuous, the scrubbers and silver reactors perform quite well. Efficiencies for iodine removal of 99.9% have been reported (3, 4, 9). The operating expense for either the scrubber or the silver reactor will be high because of the operating temperature (200°C or higher) required for the silver reactor and the necessity for replacing and maintaining the absorbing liquid removed from the scrubbers by the air stream. This expense may not be prohibitive for batch operations, but may very well be too expensive for continuous operation as in the case of the ORR experiment off-gas line. Solid adsorber materials such as activated charcoal or Linde Molecular Sieve material (a synthetic zeolite) have been used successfully for removing iodine activity from air streams. Efficiencies greater than 98% have been reported for these materials (4, 7, 11). An iodine adsorption system using these materials would not require close supervision during reactor operation and would be in readiness at all times. Copper metal in the form of machine turnings has been suggested as a possible adsorber for iodine; however, no experimental work has been reported. Adsorption of iodine on copper oxide, however, is relatively inefficient even at high temperatures and pressures (10). Copper oxide would be formed on copper surfaces exposed to an air stream for long periods of time.

II. Experimental Study of Iodine Adsorption

An iodine adsorption system utilizing solid adsorber materials was selected for this study as the most promising system for use in the experiment off-gas line. Both activated charcoal and Linde Molecular Sieve material were tested. Since the investigations reported in the literature were conducted at much lower gas flow rates, it was deemed necessary to perform iodine adsorption tests under conditions similar to those anticipated in the ORR experiment off-gas line. Before designing a laboratory iodine adsorption system it was first necessary to design a hypothetical large scale system which could then be scaled down for experimental study. The experiment off-gas system from the ORR consists of an 8 inch diameter pipe carrying an air stream up to 1000 cfm with an available pressure drop of 30 inches of water. It was assumed that a pressure drop of 3 inches of water through the iodine trap could be tolerated. On this basis a hypothetical iodine trap containing 2-4 mesh activated charcoal was designed. An iodine trap 2.75 feet in diameter and 1 foot in length would give a pressure drop of 2.8 inches of water. The linear velocity of the gas stream through this trap would be 168 ft/min. The scaled-down laboratory trap consisted of a 0.5 inch diameter x 12 inch glass pipe containing either activated charcoal or Linde Molecular Sieve material. The air flow through these small traps was 7 liters/min which gave a calculated linear velocity of 180 ft/min. Palladium iodide synthesized from I^{131} was thermally decomposed to liberate the I^{131} used in the experiments. The first iodine trap was filled with 18.8 grams of Columbia G activated charcoal, 8-14 mesh. The I^{131} was generated and swept into the trap by the 7 liter/min air stream. This flow of air was maintained for 20 hours. Two cold traps,

immersed in an acetone-dry ice mixture and connected in series, were employed to trap any I^{131} that passed through the charcoal mass. At the end of the 20 hour period the charcoal was removed and examined for I^{131} activity. It was determined that 99.95% of the I^{131} was retained by the charcoal. A second iodine trap was filled with Columbia G charcoal, 8 mesh, and studied under identical conditions. Again 99.95% of the I^{131} remained on the charcoal. A third iodine trap was filled with 20 grams of Linde Molecular Sieve material (13X, 1/16 in. pellets) and an identical experiment performed. This material also retained 99.95% of the iodine but the longitudinal activity distribution along the bed was different. When using charcoal more than 90% of the I^{131} remained in the first 3/8 in. of the charcoal mass; however, the Molecular Sieve material contained only 55% of the activity in the first 3/8 in. of the mass. Longitudinal I^{131} activity distributions for both charcoal and Molecular Sieve material are given in the attached graph.

The iodine seemed to be "fixed" on the surface of the charcoal, suggesting chemisorption rather than physical adsorption. No movement of activity down the length of the charcoal mass could be detected during the 20 hour duration of air sweep. Other workers have noted this effect (4). The sieve material did not "hold" the iodine as strongly as the charcoal and a very slow downstream transport of iodine activity was noted over the duration of the test.

The effect of moisture adsorbed on the charcoal from the air stream on iodine adsorption is not considered to be serious. In the laboratory study the charcoal studied contained approximately 5% water by weight and no effort was made to dry the air used to sweep the trap. It has been

suggested by a manufacturer of activated charcoal that iodine is selectively adsorbed in preference to water. If this is the case, the iodine will replace adsorbed moisture on the charcoal surface.

The maximum capacity of charcoal for iodine was not determined; however, it is reported that charcoal is capable of adsorbing up to 12 curies of iodine per gram of charcoal (7).

On the basis of the experimental data reported in the literature, it appears that either activated charcoal or Linde Molecular Sieve material would perform adequately for the removal of iodine activity from the ORR experiment off-gas system.

III. Conditions Resulting from Accidental Release of Iodine Activity

In order to formulate a conceptual design of an iodine adsorption trap it was first necessary to determine the worst possible conditions that could result from experiment failures in the ORR. It is estimated that the maximum fission power output from experimental assemblies would total 120 KW (8). If all the experiments, after having operated for 39 days, failed at the same instant and released all the contained radioisotopes of iodine in a gaseous form, the total activity would be 3.0×10^4 curies. The distribution of the various radioisotopes of iodine, derived from data given in a report by Binford and Burnett (2), are given in the following table.

<u>Nuclide</u>	<u>Decay Constant (hr⁻¹)</u>	<u>Initial Activity (Curies x 10⁻⁴)</u>
I ¹³¹	0.00355	0.30
I ¹³²	0.289	0.48
I ¹³³	0.033	0.71
I ¹³⁴	0.792	0.83
I ¹³⁵	0.104	<u>0.64</u>
		3.0

It is assumed that suitable air filters are contained either in the individual experiment assemblies or in the ORR experiment off-gas system so that all radioactive particulate matter will be removed upstream from the iodine adsorption trap.

IV. Conceptual Design of Iodine Adsorption System

The hypothetical iodine trap, described previously, can now be evaluated based on experimental iodine adsorption data under conditions of iodine discharge from experiment assemblies described above. The trap would consist of a container 2.75 feet in diameter and 1 foot in depth, containing approximately 178 pounds of activated charcoal, 2-4 mesh size. At an air flow of 1000 cfm, with a pressure of 30 in. of water, a pressure drop of 2.8 in. of water through the trap will result. The calculated linear air velocity through the trap will be 168 ft/min.

Based on laboratory data, this trap will remove 99.95% of the iodine vapor from the air stream of this nature. Assuming that the 3.0×10^4 curies of mixed radioisotopes of iodine are released at the same instant into the off-gas system and that all radioactive particulate matter is

removed upstream from the iodine trap, then only 15 curies of mixed iodine isotopes will pass through the trap, over a period of 1-2 minutes. A continuous permissible emission rate of 20 mc/min I^{131} for the entire Laboratory area has been reported by Burnett (5). Of the 15 curies of iodine passing through the proposed iodine trap, only 1.5 curies will be I^{131} . This is the same as releasing the maximum permissible amount of I^{131} for the entire area for a period of 75 minutes. Making the pessimistic assumption that the biological hazard of each of the different radioisotopes of iodine is equal to that of I^{131} , then the 15 curies released will represent the maximum permissible iodine discharge for a period of 12.5 hours. In other words, the radiation dose received by Laboratory personnel from this sudden release of 15 curies of radio-iodine would be equal to the dose received from continuous emission of 20 mc/min of I^{131} over a period of 12.5 hours.

It has been assumed that all the gaseous iodine isotopes released will reach the charcoal trap. In practice, some of the iodine will deposit on the walls of the off-gas lines and at other points where turbulent conditions exist. Deposition of I^{131} vapor from air streams moving at velocities up to 900 ft/min have been reported. The iodine vapor does not deposit readily from laminar air streams, however any surface imperfections will create turbulent conditions downstream from it and iodine vapor will deposit in this area (6). This effect will relieve the charcoal trap of some of the iodine load, but in addition will introduce contamination of the lines of the off-gas system.

V. Comments on Iodine Adsorption and Iodine Trap Design

While the proposed iodine trap design gives indications that it will reduce the amount of iodine emission from the ORR experiment off-gas systems in case of an accident, it is felt that additional experimental study is needed to insure successful performance of a finalized design.

The iodine removal efficiency decreases very rapidly down the length of the small laboratory traps. At the inlet of the charcoal trap the iodine concentration was reduced by a factor of 10 in 0.17 inches, while at the outlet the efficiency dropped to a factor of 10 in 19 inches. The removal efficiency should be constant if all the iodine exists in the same form and the condition of the adsorber material is uniform down the length of the trap. Several explanations of this phenomenon may be offered. A small part of the iodine may be in the form of an "exotic" unadsorbable species (4) or it may be iodine adsorbed on either small particulate matter in the air stream or charcoal dust from the trap. In either case this iodine activity would pass through the charcoal trap. Another explanation is that the adsorber material at the inlet end of the trap was different in such a way that adsorption of iodine was enhanced. Additional laboratory study is needed to determine the correct explanation.

The iodine trap must be capable of withstanding long service under high air flow rates and still be in readiness for iodine adsorption. The expected life of a charcoal trap should be determined. In addition to possible physical damage to the charcoal by extended air flow one must consider the effect of adsorbed water and oil from the air stream on iodine adsorption efficiency of charcoal. Also the lengths of time charcoal should be expected to retain adsorbed iodine under continued air flow merits investigation.

The maximum capacity of charcoal for iodine has not been determined under conditions peculiar to the ORR operation. The proposed trap will have a capacity of approximately 9.7×10^5 curies of iodine, based on a reported capacity of 12 curies per gram of charcoal, assuming the reported capacity is applicable to conditions present in the ORR off-gas system.

While charcoal is effective for adsorption of iodine it suffers from the disadvantage of being combustible. The mass of charcoal at the entrance of the trap will be heated by decay of the adsorbed iodine isotopes. While the overall heat generation is not large, the possibility exists that a high concentration of radioiodine in one small area of the charcoal bed would produce a large temperature rise with possible ignition of the charcoal. One method of reducing this possibility is to place a layer of molecular sieve material upstream from the charcoal mass. The sieve material would adsorb the iodine vapor and slowly release it to the charcoal mass. Short-lived isotopes would decay in this zone and reduce the heat load on the charcoal mass. Thermocouples placed in both the sieve material and charcoal masses could be used to monitor the heat generation.

If ignition of the charcoal should occur the combustion rate can be controlled by reducing the air flow, thus reducing the available oxygen. Experimental tests have shown that complete stoppage of the O_2 supply will extinguish the fire (1). This may be accomplished by installing two parallel iodine adsorption traps. If the trap in use should happen to ignite from the heat generation of the decaying iodine, then the air stream would be diverted to the spare trap and the burning bed sealed off.

V. Conclusions and Recommendations

On the basis of these experimental data and data reported in the literature, it is concluded that either activated charcoal or Linde Molecular Sieve material would perform adequately for removal of gaseous iodine activity from the ORR experiment off-gas system. It is recommended that additional laboratory study be conducted before a final design of the iodine adsorption system is made.

IODINE-131 DISTRIBUTION IN EXPERIMENTAL IODINE TRAPS

ORNL-IR-Dwg.-29948
UNCLASSIFIED

References

1. R. E. Adams and W. E. Browning, "Estimate of the Probability and Consequences of Ignition of the HRT charcoal Beds", CF Memo (In preparation).
2. P. T. Binford and T. H. J. Burnett, "A Method for the Disposal of Volatile Fission Products from an Accident in the Oak Ridge Research Reactor", ORNL-2086, August 2, 1956.
3. A. G. Blasewitz, et al., "Decontamination of Dissolver Vent Gases at Hanford", HW-20332, February 16, 1951.
4. J. R. Bower and G. K. Cederberg, IDO-14419, September 25, 1957 (Classified); IDO-14422, December 31, 1957 (Classified).
5. T. H. J. Burnett, personal communication, unpublished report.
6. A. C. Chamberlain, "Experiments on the Deposition of Iodine-131 Vapour onto Surfaces from an Airstream", AERE-HP/R-1082, January 1953.
7. J. W. Finnigan, et al., "Removal of Iodine Vapor from Gas Streams by Sorption on Charcoal", HW-26113, November 4, 1952.
8. H. Klaus, ORNL Operations Division, personal communication.
9. R. McNabney and A. N. Lyon, "The Removal of Iodine from Gas Streams by Reaction with Silver in Packed Towers", ARSC-28, August 26, 1949.
10. H. F. Reichard, et al., Homogeneous Reactor Fuel Reprocessing Quarterly Progress Report for July 1 - September 30, 1956, KLX-10045.
11. M. A. Wahlgren and W. W. Meinke, "Molecular Sieves Adsorb Iodine-131 from Air", Nucleonics 15, 156 (1957).

Distribution

1.	R. D. Ackley	43.	J. A. Lenhard (AEC)
2-12	R. E. Adams	44.	R. A. Lorenz
13.	T. A. Arehart	45.	J. P. McBride
14.	R. G. Berggren	46.	H. F. McDuffie
15.	D. S. Billington	47.	R. A. McNees
16.	W. A. Brooksbank	48.	J. R. McWherter
17-24	W. E. Browning	49.	F. C. Maienshein
25.	T. H. J. Burnett	50.	J. G. Morgan
26.	T. A. Butler	51.	K. Z. Morgan
27.	R. L. Clark	52.	G. Parker
28-33	T. E. Cole	53.	F. Pleasanton
34.	E. Collins	54.	P. E. Reagan
35.	J. A. Cox	55.	A. F. Rupp
36.	J. N. Crawford	56.	R. P. Shields
37.	E. P. Eppler	57.	O. Sisman
38.	D. E. Ferguson	58.	W. H. Tabor
39.	J. C. Hart	59.	D. B. Trauger
40.	D. T. Jones	60.	J. C. Wilson
41.	G. W. Keilholtz	61.	J. C. Zukas
42.	J. E. Lee, Jr.	62-65	Laboratory Records
		66-80	TISE
		81-82	Central Research Library
		83	M. J. Skinner
		84	Document Reference Section

Dent
Sty