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Waste Disposal

AEC RESEARCH AND DEVELOPMENT REPORT

DECONTAMINATION OF DISSOLVER VENT GASES AT HANFORD

By

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February 16, 1951

HANFORD WORKS
RICHLAND, WASHINGTON

CLASSIFICATION CANCELLED

DATE 7/6/59

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ABSTRACT

The dissolver vent gases constitute the most highly contaminated gas stream in the present Hanford Separations Plants. From the health hazard viewpoint there are two principal contaminants, radio-iodine and an aerosol composed of other fission products. An extensive study has been made at Hanford of methods and equipment to remove effectively these materials. This investigation has led to the development and adoption of the silver reactor and Fiberglas filter for iodine and particulate matter removal, respectively. Plant scale equipment has been designed and installed in the dissolver cells. The first silver reactor - Fiberglas filter assembly was placed in operation on October 26, 1950. The efficiencies of the two components have been evaluated. Caustic scrubbing monitors were used to determine the radio-iodine content of the dissolver vent gases upstream and downstream from the silver reactor, and CWS Type 6 monitoring filters were used to determine the particulate decontamination of the Fiberglas filter. The monitoring data have established the I^{131} removal efficiency of the silver reactor to be 99.9% and the filtration efficiency of the Fiberglas filter bed to be greater than 99.9% and probably near 99.99%. A conservative estimate of the minimum useful lives of the two components (under Hanford conditions) is at least one year.

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DECONTAMINATION OF DISSOLVER VENT GASES AT HANFORD

INTRODUCTION

In the separations process the irradiated metal, after a suitable "cooling" period, is charged to the dissolver, or the first processing vessel. Here the aluminum jackets are removed and the uranium is dissolved. The dissolver vent gases during actual metal dissolution and the agitation and transferring of the prepared solution constitute a very highly contaminated gas stream in the present Separations Plants. The radioactive contaminants considered in this report are of two types, radio-iodine, and an aerosol composed of other fission products. One of the primary problems of the Stack Gas Group at Hanford Works has been the development of methods and equipment to remove effectively, the radioactive iodine and particulate contamination from the dissolver off-gas streams. The criteria guiding the study of possible removal processes have been: high efficiency, low maintenance requirements, avoidance of secondary disposal problems, and simplicity of operation. The investigation has led to the development and adoption of the silver reactor and Fiberglass filter for iodine and particulate matter removal, respectively. The functions of the two apparatus are independent and they will be discussed individually.

SILVER REACTOR

(A) Background

The investigation of the silver reactor as a method of radio-iodine removal was initiated at Hanford late in 1947. The basis of operation is the use of a silver-bearing, reacting bed at an elevated temperature. The iodine reacts chemically to form silver iodide and is retained within the bed. For efficient removal of iodine it is necessary to maintain the silver bed above the condensation temperature (230°F) of the nitric acid vapors evolved during metal dissolution. The optimum temperature appears to be between 300 and 425°F. The equilibrium of the reaction

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favors Ag I formation even at low temperatures. The low vapor pressure of iodine above silver iodide at the operating temperatures makes possible the rapid and essentially complete removal of iodine from the gas stream.

In the first series of tests an 8 inch depth of solid silver pellets (4 to 10 mesh) was contained in a 2 inch stainless steel reactor pipe heated by a resistance tube furnace. An aliquot of the dissolver off-gas stream, 0.3 cfm. was passed thru the reactor. A scrubbing tower containing trichlorobenzene was used in series downstream from the reactor to determine the quantity of iodine which was not stopped by the silver bed. In subsequent runs, a 6000 watt preheater was installed prior to the reactor to permit the treating of larger quantities of the gas stream (10 to 12 cfm.), and a corundum base silver catalyst, which contained 2% silver by weight, was used in the interest of economy of silver. The catalyst was obtained from the Air Reduction Sales Company. These early Hanford tests clearly indicated that removal efficiencies in the order of 99.9% could be obtained.

Due to the pressure of other Plant problems, the assistance of the Air Reduction Sales Company was enlisted in the silver reactor development program. Their investigation was conducted over the period from March 1948 to August 1949. Efficiency determinations under various operating conditions were made by Air Reduction with cold iodine and with an iodine stream containing tracer quantities of I^{131} . This work has been described in their Reports ARSC - 10, 18, 20, 25, 28, and 29.

A series of confirmatory pilot plant runs was made at Hanford from October to December, 1949. All the experimental results indicated that the method could provide an iodine removal efficiency of at least 99.9%. Accordingly, the decision was made to install plant-scale silver reactors.

(B) Plant Silver Reactor

The first silver reactor - Fiberglass filter assembly was placed in operation on October 26, 1950. A schematic representation of the equipment is given in Figure I. Dimensional details of the heater and reactor column are presented in Figure II. The heater and reactor are designed for an off-gas flow of 150 scfm. (35% air and 65% oxides of nitrogen) and a gas temperature of

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435°F. entering the reactor. The heater section, 1 foot 9 1/2 inches x 10 1/4 inches in cross-section and 6 feet 11 1/4 inches long, has 9 stainless steel encased calrod heating elements arranged in 3 parallel circuits connected to a 3 phase 440 volt power supply. Each element is rated at 12 KW. The silver reactor is 2 feet in diameter with an overall height of 9 1/4 feet. The packed section is 8 feet in height and composed of 1/2 inch unglazed Berl saddles coated with silver nitrate. The packing was prepared by dipping the saddles in 20 molal silver nitrate solution, drying and charging to the reactor. The temperature of the gas stream leaving the heater is controlled at 475°F; a 40°F drop in temperature occurs between the heater and reactor. A platinum thermohm is located in the gas stream prior to its entering the reactor column. The temperature reading is recorded on a Leeds and Northrup Micromax and is transmitted to a Reactrol and bank of 3 saturable reactors. The reactors proportionate the power input to the heater elements to maintain a constant temperature at the control point. A second thermohm is located in the plenum chamber above the reactor bed. This is used to establish the fact that the entire reactor is at temperature before metal dissolution is begun. Approximately 8 hours are required to bring the entire column from room temperature to the operating temperature. Because of the frequency of dissolving, the reactor unit is continuously maintained at the operating temperature. The melting point of silver nitrate is 413°F and the film becomes quite fluid at temperatures above 500°F. To prevent damage to the silver nitrate surface from overheating the bed an audible alarm has been incorporated into the temperature recorder. This will warn the operator if the temperature of the gas stream to the reactor reaches 500°F. The current to the heating elements can then be cut off until the difficulty has been corrected. From the operational viewpoint, the performance of the equipment has been satisfactory.

(C) Performance Data

The first silver reactor - Fiberglass filter assembly was provided with complete sampling facilities. Monitoring data, based on sampling by caustic scrubbing of aliquots of the dissolver off-gases upstream and downstream from the silver reactor have established the I^{131} removal efficiency to

be 99.9%. It is believed that the precision of the present sampling and analytical techniques prevent the determination of a fourth significant figure. The data are presented in Table I.

TABLE ISILVER REACTOR MONITORING DATA

<u>Date</u>	<u>Upstream Monitor</u> <u>μc. I¹³¹</u>	<u>Downstream Monitor</u> <u>μc. I¹³¹</u>	<u>Efficiency</u> <u>%</u>
11/10/50	1260	0.6	99.95
11/10/50	1420	0.4	99.97
11/14/50	20780	63.8	99.69
11/14/50	11120	9.8	99.91
11/14/50	12640	5.0	99.96
11/16/50	1640	5.6	99.66
11/16/50	2740	10.4	99.62
11/21/50	10620	14.0	99.87
11/21/50	11880	14.0	99.88
11/21/50	6340	12.0	99.81
11/21/50	15080	12.0	99.92
11/21/50	30080	12.0	99.96
12/14/50	2060	0.56	99.97
1/23/51	3430	2.09	99.94
		<u>Average</u>	<u>99.9</u>

The data for each set of upstream and downstream monitoring scrubbers have been normalized to equal volumes of gas delivered to the monitors. The same volume of gas was not necessarily used in all the monitoring runs.

The maximum life of the silver reactor can be estimated from the quantity of silver available for reaction. The average silver nitrate content of the dissolver cell reactors is 50 pounds. On this basis the available silver is 31.7 pounds or 14,400 grams, and the theoretical iodine capacity would be 16,900 grams. This calculation is based upon a complete utilization of the silver and would require penetration of iodine through the silver iodide film initially formed. Data on this point are not available. An estimate of the life of the

reactors for the Hanford dissolvers is very difficult at the moment because of operating changes being made. Consideration of these two factors has led to a conservative estimate of one year's operation on the Hanford dissolver off-gas stream. *

FIBERGLAS FILTER

(A) Background

The success of the sand filter installations at Hanford led to the investigation of employing the same filtration mechanism, but with filter media of more desirable properties. Fine glass fibers were selected as a material offering much promise. An extensive study has been made of the parameters affecting filter performance of the various grades of Fiberglas produced by the Owens Corning Fiberglas Corp. This has resulted in comprehensive data that make it possible to design a Fiberglas filter to meet any specific operating conditions and requirements. A detailed account of this development work and the bases for the selection of the filter bed composition used in the dissolver off-gas filter will be presented in a separate report now in preparation.

(B) Description of Plant Equipment

The dimensional details of the dissolver cell Fiberglas filter are presented in Figure III. The unit is 2.5 feet by 5.5 feet with an overall height of 4.3 feet. The cross-sectional area, 12.5 ft.^2 , results in a superficial velocity of approximately 20 ft./min. under conditions of maximum gas flow (170 scfm. during the charging operation) and at the temperature of the gas stream in the filter (approximately 300°F). The filter has been designed to provide a decontamination of the gas stream of 10^4 and a pressure drop of 4.0 inches of water at this flow rate. The filter bed composition is tabulated below:

* Details of this calculation are not included in order to avoid a production index.

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<u>Filter Bed Strata</u>	<u>Type Fiberglas</u>	<u>Bed Depth</u>	<u>Packing Density</u>
Clean-up Layer	"AA"	1 inch	1.2 lbs. /ft. ³
Third Layer	115-K	20 inches	6.0 lbs. /ft. ³
Second Layer	115-K	10 inches	3.0 lbs. /ft. ³
Bottom or First Layer	115-K	12 inches	1.5 lbs. /ft. ³

It is essential to prevent an appreciable condensation of liquid in the Fiberglas bed. This is due to the following factors: the greatly increased resistance to air flow associated with liquid in the packing, the probability of the formation of channels with consequent loss in filtration efficiency, and decreased life due to the deterioration of the glass fibers. This latter effect is caused by a leaching action of the water or acidic condensate. The alkali present in the glass is removed and results in a severe embrittlement of the glass fibers. In the dissolver cell installation the formation of condensate within the filter has been prevented by having the filter downstream from the reactor. The temperature of the gas stream passing through the filter is approximately 300°F.

During the pilot plant filtration study of the dissolver off-gas stream indications were obtained that residual nitric acid remained in the filter following metal dissolution and that the subsequent evolution of ammonia during the aluminum jacket removal resulted in the deposition of ammonium nitrate. As this salt formation would appreciably shorten the useful life of the filter and possibly constituted a potential explosive hazard a by-pass line around the filter has been provided. The gas stream is directed through the by-pass line during the coating removal process. The experimental data indicating this ammonium nitrate formation are open to some question. The only available access point for sampling the off-gas stream involved an extremely long length of piping (approximately 300 feet) with possible trap points for acidic condensate. Consequently, it could not be definitely established whether the salt formation was occurring in the dissolver cell system or the sampling line itself. As a safeguard, by-pass lines were provided in the first units. Recently, the lines used to obtain the pressure drop across the filter have been used to draw samples of the gas stream into the Canyon Building pipe gallery. The possibility

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of ammonium nitrate formation within the filter is now being studied with the use of these short, condensate free lines. In this way the need for by-pass provisions in future dissolver off-gas installations will be determined.

(C) Performance Data

To obtain accurate efficiency data for the Fiberglas filter, monitoring equipment was installed in the Canyon Building pipe gallery. The upstream and downstream gas samples were drawn through the pressure lead lines (mentioned above) and passed through CWS Type 6 monitoring filters. This system is schematically represented in Figure IV. The data have established the particle removal efficiency of the Fiberglas media to be greater than 99.9% and probably near 99.99%. The data are given in Table II.

TABLE II
FIBERGLAS FILTER MONITORING DATA

Date	Upstream Monitor*		Downstream Monitor*		Efficiency (%)
	Flow (cfm)	Mrep/hr.	Flow (cfm)	Mrep/hr.	
12/19	1.0	6000	1.0	<5	>99.92
12/19	1.0	1500	1.0	<5	>99.67
12/21	1.0	4250	1.0	<5	>99.88
12/21	1.0	33000	1.0	8	99.97
12/21	0.5	8750	1.5	<5	>99.98
12/22	0.5	6000	3.0	<5	>99.98
12/22	0.5	135	3.0	<5	>99.39

* All activity measurements are Cutie Pie readings. This instrument does not give quantitatively reliable readings below about 5 mrep/hr. This leads to the "greater than" efficiencies listed above and discussed in the paragraph below.

In this efficiency range, with only approximately 1 part in 10,000 passing the filter it is quite difficult to obtain definitive efficiency values. Even with the expedient of using different flow ratio through the two monitors it is difficult to obtain a set of monitoring filters for which the upstream sample is not too "hot", or the downstream sample too "cold" for accurate activity readings.

Consequently, the majority of the efficiency determinations can be expressed only as "greater than" values. It should be noted that these efficiencies represent the actual removal of contamination by the Fiberglas bed. The efficiency values obtained agree very well with the calculated design figures. If there is a slight leakage through the seat of the by-pass valve, the leakage would not be recognizable with the use of the pressure taps as sampling positions for efficiency determinations. The effective efficiency of the unit, as constructed, could, therefore, be less than indicated by the efficiency determinations. Leakage tests performed on the by-pass valves before installation on the filter assemblies in the operating cells indicated less than 0.05% leak through the valves. Even this small flow through the closed by-pass valve could represent several times the amount of activity passing through the Fiberglas.

The extremely high contamination level of the dissolver vent gases has prevented a determination of the actual loading of the gas stream. The effective loading of the main ventilation air stream however, has been measured at several points throughout the ventilation system. These data have been obtained by determining the pressure drop increase across test CWS filters at the following locations: the building supply air prior to the air conditioning treatment, after conditioning, at the Canyon deck, and downstream from the process cells. These measurements have established that only a small fraction of the filter burden of the effluent building air stream is contributed by the conditioned air supplied to the Canyon deck. The majority of the suspended material in the effluent air is due to the aerosol generated in the process operations and discharged from the vessel vent lines. When the effluent air stream from the process cells and the conditioned air entering the cells were used as test streams, the pressure drop across a CWS test filter was doubled twenty five times as fast for the effluent air as for the conditioned air.

The activity level of the dissolver vent gases, per unit volume, is in the order of 15 times that of the main ventilation air stream (downstream from the process cells). Therefore an effective loading 15 times that of the ventilation air can be used for calculation purposes. It should be noted that this approach will yield a fictitiously high loading for the dissolver off-gas stream due to the fact

that its specific activity is much greater than that of the aerosol generated in the later stages of the Canyon separations process. Consequently, a figure obtained with this loading represents a minimum filter life.

A corollary study has been made of the relative life expectancies of Fiberglas and sand filters. The procedure and results will be presented in detail in the Fiberglas filtration report now in preparation. The tests were conducted by passing an air stream laden with methylene blue smoke through parallel filter units containing various grades of Fiberglas media and a model of the present Plant sand filters. The methylene blue smoke had a median particle size comparable to the radioactive ventilation air aerosol (0.6 microns). The quantity of methylene blue passed to the various units was determined by spectrophotometric analyses of solutions obtained from CWS Type 6 monitors. The useful life of the filters was obtained by the pressure drop increase in relation to the quantity of dust deposited. The data have made it possible to estimate a sand filter life in the order of 6-8 years. The study established that the useful life of a Fiberglas filter with the composition of the dissolver off-gas filter would be approximately twice that of the sand filter for a methylene blue aerosol. On the basis of the loading of the dissolver vent gases being 15 times that of the ventilation air, the minimum life of the dissolver cell filter should be at least one year. The first dissolver off-gas filter was placed in operation Oct. 26, 1950. There has been no detectable increase in pressure drop, to date.

SUMMARY

The silver reactor and Fiberglas filter have been developed and selected at Hanford Works as the methods best fulfilling the requirements for the removal of radio-iodine and particulate contamination from the dissolver vent gases. Plant scale equipment has been designed and installed in the dissolver cells and the efficiencies of the units have been evaluated. The I^{131} removal efficiency of the silver reactor is 99.9% and the filtration efficiency of the Fiberglas filter bed is greater than 99.9% and probably near 99.99%. A conservative estimate of the minimum useful lives of the two components (under Hanford conditions) is one year. The equipment has been in operation for over three months with no change in the performance characteristics.

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E. F. Kutz	W. C. Schmidt	B. Weidenbaum	

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Figure I

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SCHEMATIC DIAGRAM
OF SILVER REACTOR-
FIBERGLAS FILTER
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Figure III

SECTION - AA

SECTION-BB

PLAN

Scale: 1" = 1'-0"

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DISSOLVER CELL
FIBERGLAS FILTER

Figure IV
FIBERGLAS FILTER MONITORING EQUIPMENT
221-B PIPE GALLERY

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