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RECLAMATION OF A PARTIALLY PLUGGED
GLASS FIBER VENTILATION FILTER
AT THE HANFORD PUREX NUCLEAR FUELS
REPROCESSING PLANT

MASTER

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REVISED ABSTRACT

Canyon ventilation air from the Hanford Purex Nuclear Fuels Reprocessing plant is exhausted through a filter system before being released to the atmosphere. The filter installation consists of a bulk-particulate removing, glass fiber fore-filter, and a fine-particulate removing, high efficiency, glass fiber after-filter. The original filter installation was built in 1955 during plant construction and operated satisfactorily until early 1964 when the fore-filter indicated a loss of capacity because of particulate loading. Construction of a new filter installation was started immediately and it was put into operation in 1965.

When plugging of the original filter became evident, a program was initiated to define the cause of the problem and to develop, if possible, a way to reclaim the filter without damaging it. Samples of filter entrance air indicated a large percentage of the particulate to be crystalline ammonium nitrate. Core samples of the fore-filter glass fiber bed showed a predominantly ammonium nitrate loading confined to the inlet side of the bed. In a laboratory unit, an ammonium nitrate loading was shown to displace and disperse in the glass fiber bed, with resulting reduced pressure drop for a given air flow, by passing highly humidified air through the glass fiber bed.

Based on the laboratory studies the partially plugged original filter, which had been removed from operation, was treated for reclamation using a small flow of highly humidified air through the fore-filter. The results of the reclamation treatment were satisfactory and it was possible to return the original filter installation to service handling the entire exhaust ventilation flow. The filter was reclaimed at a cost of \$15,000; the filter replacement cost \$750,000. While the reclaimed filter again developed a high pressure drop after five months of service and is not now in service, it remains useful as an emergency back-up to the replacement filter.

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1. Introduction

The Purex Plant at the Hanford Works was designed to process low enrichment uranium fuels using the Purex Process as discussed in earlier meetings^[1]. It is a canyon type building approximately 1000 feet long, 61 feet wide, and 104 feet high. It contains fourteen individual cells of varying size to remotely contain the process equipment.

The containment of airborne radioactivity in the Purex Plant is accomplished by systematically adjusting the static pressure throughout the building. The ventilation air flows from the zones of the least contamination toward the highly contaminated process cells. Prior to discharge to the atmosphere via a tall stack, the air is filtered through an underground glass fiber filter. The original filter operated successfully from plant startup in late 1955 until March of 1964. At that time excessive differential pressures appeared across the fore filter which, in turn, reduced the total air flow of the system to marginal values. A new glass fiber filter was constructed to ensure operating continuity of the Purex Plant and a program to define and correct the high differential pressure across the original filter was initiated.

This paper describes the original glass fiber filter, the cause of the high differential pressure across it, the development work conducted in support of its reclamation, and the humidification of the ventilation air to reclaim the partially plugged filter.

2. Facility Description

The Purex Plant is used to chemically reprocess the nuclear fuels of the Hanford plutonium production complex. A solvent extraction process is used to remove radionuclide and inert impurities and isolate uranium, plutonium, and neptunium. The process is based on the extraction of uranium, plutonium, and neptunium from aqueous nitric acid solution into organic tributyl phosphate-normal paraffin hydrocarbon mixed solvent. Radionuclide and inert contaminants are relatively inextractable.

The high radioactivity associated with the nuclear fuel requires that massive concrete shielding be placed between processing equipment and operating personnel. The processes are operated remotely; i. e., the basic operations including maintenance are done from remote stations with no direct observation or contact between personnel and equipment. The processing equipment is within concrete shielded cells below concrete cover blocks. The cells are within the concrete walled Purex Building canyon. Changes in processing equipment are accomplished by removing the cell cover blocks and working within the cells with a traveling bridge crane. The crane operator sits in a shielded cab behind a concrete parapet and views the crane movements through periscope optics. Figure 1 shows the Purex canyon, the crane and the processing cells.

The Purex canyon is ventilated by drawing air into the canyon, down through the processing cells, and into the air tunnel. The supplied ventilation air is heated, filtered, and washed before going to the canyon. The exhaust air from the air tunnel is drawn through a fibrous glass filter to remove radioactive particulate material before being discharged to the atmosphere. Exhaust fans pull the air out of the canyon through the filter and discharge this air to the stack. The ventilation scheme is shown in Figure 2. The supply air normally is 80,000 cfm (cubic feet per minute) and the exhaust ventilation air 120,000 cfm. The 40,000 cfm of air not supplied comes from in-leakage via the process lines and the railroad tunnel door.

The original ventilation filter is depicted in Figure 3. It is enclosed below grade in a concrete housing 80 ft. by 50 ft. by 15 ft. and is divided into two areas with one containing a fore-filter and the other an after-filter. The fore-filter is the larger and removes most of the particulates from the incoming exhaust air. The after-filter removes additional particulate matter which passes the fore-filter. Air enters the filter through the intake duct and flows over the top, down through, and out beneath the fore-filter bed. The air then flows across beneath the inlet side of the after-filter, up through the frames of the after-filter units, and finally across the outlet duct.

The fore-filter is divided into three bays. Each of these bays contains free-packed Owens-Corning Fiberglas^{R1}, Type 115 K, retained by upper and lower stainless steel screens. With the free-packed glass fibers the desired density gradient is achieved by the weight of the media. As originally installed the bed media density varied from 0.7 lb/ft³ (pound per cubic foot) at the top to 3.3 lb/ft³ at the bottom. There is approximately 37,000 pounds of fibrous glass filter media in the fore-filter. The fore-filter collection efficiency was designed for 92.0 percent removal of all particles 0.7 micron in diameter or greater. The fore-filter has 2,500 ft² of flow area and a 50 ft/min. air velocity through the bed at 126,000 cfm designed capacity.

The after-filter is made up of 132 American Air Filter Company's Deep-Bed filter units. Each unit is packed with 1/2 in. No. 25 FG Filterdown^{R2} and 1/2 in. of No. 50 FG Filterdown. The density of the No. 25 media is 1.4 lb/ft³ and the No. 50 media is 1.2 lb/ft³. The units consist of five wedge-shaped pockets supported in a grid on frames. There is 6,600 ft² of area in the after-filter and an air velocity of 20 ft/min. through the media at designed capacity. The collection efficiency of the after-filter is 98.8 percent for all particles 0.7 micron in diameter or greater, giving an over-all filter collection efficiency of 99.9 percent.

3. Filter Plugging Problem

From 1955 to 1964, the differential pressure across the fore-filter slowly rose from a 3.0 in., w.g. (water gauge), to 4.4 in., w.g., at a nominal air flow of 125,000 cfm. The differential pressure across the after-filter remained relatively constant at 2.1 in., w.g. During a one-week period in the latter part of March, 1964, the differential pressure on the fore-filter increased to 7.0 in., w.g., with a resulting decrease in total air flow to 110,000 cfm^[2]. The rate of rise of the differential pressure across the filter increased until a rise of 1.5 in. per day was observed.

It was first thought that the increasing pressure drop and decreasing flow were caused by excessive moisture in the filter. Humidity measurements were first taken with a sling psychrometer; later, data were obtained from a Cloud-Cup apparatus. A continuous Foxboro Dew Cell^{R3} was then installed upstream and downstream of the filter. All data collected indicated that the relative humidity did not exceed 51 percent and most of the time it was below 30 percent. At these relative humidities, the accumulation of water was considered improbable.

^{R1} Registered Trade Name, Owens-Corning Glass Co., Toledo, Ohio

^{R2} Registered Trade Name, American Filter Co., Louisville, Kentucky

^{R3} Registered Trade Name, Foxboro Co., Foxboro, Massachusetts

Aliquot samples of the influent air were taken to determine particle loading and size. During the initial period of the rapid rise of the differential pressure of the fore-filter, dust loadings as high as 200 milligrams per 1000 cubic feet of air were measured^[3]. This deposition rate was ten times greater than the estimated average rate of solid deposition on the filter and one hundred times greater than the probable rate of deposition from atmospheric solids alone. A spectrographic analysis of the collected sample indicated that a large fraction of the solids was ammonium nitrate.

Several core samples, 4 in. in diameter by 24 in. long, were taken of the glass fibers in the fore-filter. The core was made up of four readily distinguishable parts^[4]. Photograph 1 shows a view of the longitudinal axis of the core with the crust lying in the foreground; photograph 2 gives a head-on view of the surface crust; and photograph 3 gives a view of the four parts with (A) being the crust; (B) the gray dirt; (C) the white powder; and (D) the glass fibers. The crust and gray dirt were 44-53 percent ammonium nitrate; 8-11 percent iron; 7-12 percent silicon dioxide; 5-9 percent water; and 3-6 percent organic material. The white powder was greater than 85 percent ammonium nitrate.

An intensive sampling program was undertaken to determine the source of the ammonia or ammonium ion. Two chemicals --- sulfamic acid, $\text{NH}_2\text{SO}_3\text{H}$, and hydrazine, N_2H_2 , used in the Purex Process --- are a possible source of ammonia. Samples of the atmospheres of all the tanks, both in acidic and basic conditions, indicated that these chemicals were not emitting ammonia to the atmosphere.

Simultaneously with the tank sampling program, the integrity of another part of the processing system containing a known source of ammonia was investigated. The removal of the aluminum jackets from the uranium fuel elements with sodium nitrate and sodium hydroxide in the dissolver vessels evolves copious quantities of ammonia. This ammonia is removed from the dissolver off-gas by a scrubber and the dilute ammonium hydroxide formed in the scrubber is routed to an underground disposal site called a crib. The hydraulics of the piping system used to route the dilute ammonium hydroxide to the crib were such that the pipe from each of the three dissolvers had to be vented via a stand pipe to the cell atmosphere. A small differential pressure was found to exist across the three cells and ammonia was being evacuated into the ventilation air via this route.

4. Development Work

Development work was initiated at the Pacific Northwest Laboratories of Battelle Memorial Institute (PNL) to establish methods for

filter reclamation. Since ammonium nitrate is known to be deliquescent, humidified air was drawn through the core sample of glass fibers in the Analytical Hot Cells at PNL. When the relative humidity remained below 65 percent, no dissolution or rearrangement of the crust was observed. When the relative humidity of the air was 75 to 90 percent, approximately 45 percent of the crust dislodged or dissolved^[5]. These data encouraged a pilot plant scale demonstration of removing the crust by adjusting the humidity of the purge air.

The demonstration equipment was a small scale model of the actual filter as shown in Figure 4^[6]. The fore-filter consisted of a 5-3/4 in. ID tower of clear plastic pipe. Two and one-half pounds of 115K Fiberglas were loaded into the pipe from the top. The bottom layers of Fiberglas were compressed more than the top layers to simulate compression of the actual filter due to its own weight. The glass fibers were held in place by packing supports that were fabricated from 1 in. mesh screen. The 11-in. free space above the packing was left open to the atmosphere to provide an air inlet to the filter. The final clean-up filter was 8-3/4 in. in diameter and consisted of two, 1/2-in. thick layers of AA Fiberglas compressed to 1/2 in. thickness between packing supports. The filter was installed in a 10-in. ID clear plastic pipe to allow visual observation of the clean-up layer. Associated pressure taps, vacuum jet, steam supply, etc., as shown in Figure 4 were supplied.

A synthetic dust was used to load the filter. As shown in Figure 4, the composition was 81.4 percent NH_4NO_3 and 18.6 percent sand. All dust was passed through a 200-mesh screen. A total of 519 grams was loaded onto the filter by dispersal of the dust in the incoming air. The differential pressure across the fore-filter increased from 2.9 in., w.g., to 5.05 in., w.g., and the differential pressure across the after-filter remained unchanged at 1.1 in., w.g. After the addition of the dust, a gray colored dust layer completely covered the filter. The layer varied between 1/4 and 3/4 in. and appeared to penetrate the filter approximately 4 in.

The relative humidity of the air entering the filter was then varied by adjusting a steam bleed. At relative humidities below 60 percent there was no apparent change in the crust of dust on top of the filter or the differential pressure. At a relative humidity of 60-65 percent at a dry bulb of 73 F the differential pressure decreased from 5.05 in., w.g., to 4.48 in., w.g., in a two-hour period. Damp areas began to appear on thin spots in the dust layer and particles of ammonium nitrate on the side walls above the Fiberglas began to dissolve. At 85-95 percent relative humidity at a dry bulb of 73 F the differential pressure decreased to 3.45 in., w.g., within three hours and remained at this level.

The remaining dust layer had the appearance of damp brown dirt and had the consistency of damp sand. The surface was very irregular and had a maximum thickness of 1/4-in. The dark brown color of the dust layer, as compared to the original gray color, indicated that a considerable portion of the ammonium nitrate had dissolved and flowed downward into the Fiberglas. No change occurred in the differential pressure across the filter below the top 6 in. No change in the differential pressure or appearance of the final clean-up layer occurred.

The remaining dust layer was dried by regulating the incoming air to 15 percent relative humidity. A light brown, dry crust appeared with several cracks and small holes. The differential pressure increased from this drying from 3.45 to 3.49 in., w.g.

Supplemental solids with the same chemical composition as those previously used were added to the fore-filter to obtain information on the effect of adding solids on top of the crust and additional information on humidity effects. Two hundred fifty seven grams of dust were loaded into the filter through a 200-mesh screen. The dust was added in four increments. After each of the first three increments, the relative humidity of the inlet air was raised and maintained for 10 to 15 minutes in the range of 90 to 100 percent. The short periods of high humidity and drying were enough to cause the dust to form a crust. Immediately after the humidity was raised, the pressure drop would decrease and the drying periods did not change the differential pressure. After addition of the final dust increment the layer of solids was approximately 3/4 in. deep and there were no visible holes or openings. The differential pressure rose from 3.49 in., w.g., to 20.15 in., w.g., during the four loadings.

The relative humidity was raised and maintained for 21 hours in the range of 80 to 90 percent. After two hours the differential pressure had decreased from 20.15 in., w.g., to 10.5 in., w.g. After eighteen hours the differential pressure was down to 8.0 in., w.g. The dust layer was damp and had the consistency of damp sand. The thickness varied from about 1/4 in. to 1/2 in. and several cracks and small holes were visible. The top of the dust layer was dark brown in color, denoting that the ammonium nitrate had migrated downward. After twenty-one hours the differential pressure had fallen to 7.8 in., w.g., with no change in the appearance of the dust layer. These observations suggest that once adequate passages are opened there is little intimate contact between the humid air and the major portion of the dust layer. As a result, further pressure drop reduction and solids removal occur very slowly or stop altogether. After the twenty-one hours at high humidity the filter was dried for two hours and the differential pressure increased to 8.1 in., w.g.

The filter was then washed with seven water washes and was dried for at least one hour after each wash. Details of the washes are shown in Table I. The differential pressure was reduced from 8.1 to 3.15 in., w.g., and 71 percent of the ammonium nitrate was flushed from the fore-filter. At the flow rates employed water addition was just a slow drip so the inlet line was moved manually to allow water to drip over the entire surface. Clear water started draining out of the bottom of the filter approximately 7 minutes after each wash began. The water always drained out along the wall in the same spot during each wash. The water formed a puddle at the bottom of the free space below the fore-filter, and then flowed through the discharge line to the chamber below the cleanup filter. No water entrainment to or wetting of the cleanup filter was observed and the differential pressure across the unit remained unchanged at 1.1 in., w.g.

The first wash rapidly dissolved several 1/4 to 1/2 in. diameter holes through the crust and removed about 30 percent of the crust. Fiberglass was visible through the holes and appeared to be free of ammonium nitrate and sand while the filter was wet. The differential pressure decreased from 8.1 in., w.g., to 3.55 in., w.g. During the one-hour drying cycle after the first wash the differential pressure across the filter increased from 3.55 in., w.g., to 7.30 in., w.g. Additional drying for 16 hours caused no further change. This was the first occurrence of a significant differential pressure increase during a drying cycle. Inspection of the filter showed that the Fiberglass at the bottom of the holes in the crust was completely covered with a thin, hard crust of white ammonium nitrate. Dissolution, possibly as a result of entrained water in the air, is required to produce a sudden differential pressure increase during a drying cycle.

After the fourth wash the entire crust was gone. No solids were visible on top of the Fiberglass but sand was visible about 1/2 inch below the top. During the drying cycles after these washes only a slight pressure drop increase occurred and no white crust was observed. These washes removed 59 percent of the ammonium nitrate and the only downward movement of the insoluble material was along the channel followed by the water.

Three additional washes were carried out with higher flow rates to further study the removal of ammonium nitrate and the movement of the insoluble materials. These washes removed an additional 12 percent of the ammonium nitrate and a very slight trace of fine insoluble material was visible at the bottom of the filter along the water channels. No changes occurred in the differential pressure across the portion of the fore-filter below the top 6 in.

No change occurred in the appearance or differential pressure of the final cleanup filter during the washing. This observation is of considerable significance since previous experience has indicated that wetting of this type of filter causes holes to appear in the filtering media.

The data from the PNL development program indicated that air with a relative humidity of 60 percent or greater would dissolve a portion of the crust which was causing the high differential pressure across the fore-filter. This dissolution would reduce the differential pressure and would allow the filter to operate for an unknown period of time. Water washing would do a more thorough job of removing the crust and if the air velocity through the fore-filter was 45 ft/min or less no water entrainment will be transported to the after-filter.

5. Filter Reclamation

Based on the laboratory development work the plugged Purex Plant ventilation filter was reclaimed by passing air with a relative humidity of 80 to 90 percent through the fore-filter bed. Previously a temporary by-pass duct leading directly to the after-filter had been placed around the fore-filter to maintain adequate air flow for plant safety during the time required for designing and constructing the second ventilation filter. This by-pass ductwork was used to return the highly humidified air to the main air stream as shown in Figure 5. Ten thousand cfm or eight percent of the total air flow was humidified by steam injection at a rate of 500 lb/hr., routed through the original filter, and returned to the main air stream [7] which was exhausted through the second ventilation filter.

The fore-filter was initially treated with humidified air for four weeks. Prior to treating the filter the differential pressure was greater than 20 in., w.g., at an air flow of 115,000 cfm and after the four-week treatment the differential pressure was 7.7 in., w.g. After satisfactory operation for two weeks, the filter was again removed from service and the humid air treatment was resumed to increase the effectiveness of ammonium nitrate crust removal. Humid air was passed through the filter for three weeks. At the conclusion of this humidifying period, full air flow was routed to the filter with a resultant differential pressure of 7.5 in., w.g.

6. Post Operation

The original canyon ventilation filter was returned to service on September 25, 1967. Initially the total air flow through the filter was measured as 115,000 cfm and the pressure drop across the fore-filter

and after-filter was 5.4 and 2.1 in., w.g., respectively. During the succeeding 12 weeks the air flow through the filter gradually decreased to 113,000 cfm at a pressure drop of 5.6 and 2.1 in., w.g., respectively. During the next 7 weeks the flow decreased and fluctuated between 106,000 cfm and 110,000 cfm. The filter differential pressures remained essentially unchanged, although some fluctuations were noted^[8].

On January 6, 1968, a significant downward trend in flow started, and continued until February 12, at which time the filter flow had reduced to 92,000 cfm. During the same period the fore-filter differential pressure increased to 6.2 in.; w.g., and the after-filter differential pressure decreased to 1.8 in., w.g. By March 9, 1968, the air flow had decreased to 83,000 cfm and the differential pressures were 6.8 and 1.6 in., w.g., respectively. Although no problems of contamination control were associated with the reduced air flow, the filter was removed from service as an operational unit. It remains available as an emergency back-up for the replacement unit.

7. Cost Analysis

The reclamation program to extend the life of the original filter was a financial success as shown below. The net savings for reclaiming the filter amounted to \$24,000 based on the beneficial use of the filter for an additional five months. Subsequent additional use will, of course, add to the savings.

Original

Replacement Cost	\$ 750,000
Useful Life	8 yr (96 mo)
Cost per Month	\$ 7,800

Reclamation

5 mo. @ \$7,800	\$ 39,000
Less Reclamation Costs	<u>\$ 15,000</u>
	\$ 24,000

8. Future Effort

The development work indicated that a more satisfactory crust removal could be obtained by contacting the deposited solids with water. Previous operating experiences with the types of glass fibers in the after-filter have indicated that these fibers must remain dry at all times to remain effective as filter media. Additional studies and a satisfactory safety analysis will have to be completed before washing of the fore-filter will be attempted.

Preliminary planning also has been done on the physical removal of the fore-filter fibers. The control of airborne contamination during the actual removal would be very difficult and additional decontamination of the structure walls with liquids would probably have to be done before a re-packing operation could commence.

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FIGURE 1
Hanford Purex Canyon

Figure 2
Process Area Ventilation System

Din Din
May '60

FIGURE 3
291-A Filter No. 1

FIGURE 4
Pilot Model of Filter

Steam
Bleed

FIGURE 5
Filter Reclamation Flowsheet

TABLE I
RESULTS OF FILTER WASHING AFTER SOLIDS LOADING

Wash Increment	Flow Rate, ml/min	Addition Time, min	Volume Added, cm ³	Volume Out, cm ³	NH ₄ NO ₃ Removed, g	% of Total	ΔP Across Fore Filter, In., w.g.	
							Filter Wet	After Drying
1	17.9	14	250	not measured	Total		3.5	7.3
2	17.9	14	250	"	from first		3.5	5.8
3	17.9	14	250	"	four washes		3.2	3.5
4	17.9	14	250	"	363	58.7	3.2	3.3
5	18.7	14	262		26.7	63.2	3.2	3.2
6	44.0	14	616		34.6	68.7	3.1	3.1
7	48.7	14	682		15.2	71.3	3.1	3.1

PHOTOGRAPH 1

Filter Core -
Longitudinal Axis

PHOTOGRAPH 2
Filter Core - Surface Crust

PHOTOGRAPH 3

Filter Core -
Four Components