

# RADIOACTIVE PARTICULATE FALLOUT PROBLEMS AT THE ICPP

**GEORGE WEHMANN**

*Health and Safety Division, U. S. Atomic Energy Commission,  
Idaho Falls, Idaho*

## A B S T R A C T

The Site Survey Branch of the Health and Safety Division routinely conducts a monitoring program for determining levels of airborne radioactivity at the National Reactor Testing Station and its environs. Among methods utilized for the detection of particulates are horizontal collection plates of gummed-paper which are located outside plant areas. During the summer of 1958 routine autoradiographic analysis of these samples indicated a particulate problem connected with the Idaho Chemical Processing Plant. The follow-up program is discussed, including particle distribution, size, isotopic content, and health hazard calculations. A special attachment to the microscope to facilitate location and identification of radioactive particles is also discussed.

\* \* \* \* \*

The use of gummed-paper collectors for monitoring fallout from nuclear operations was started in 1951 by the Atomic Energy Commission (Eisenbud and Harley, 1953, 1955, 1956 and Holland, 1959). The extreme simplicity of placing, collecting, and of measuring the gummed paper for radioactivity was a strong inducement to employ this technique at a number of locations at the National Reactor Testing Station (NRTS) and its environs. As a result, the Site Survey Branch of the Health and Safety Division adopted this method as a routine means of monitoring radioactive particulate fallout. Stations have been located outside several of the plant areas in the prevailing down-wind direction (southwesterly during the day and northeasterly at night).

At each station a horizontal plate has been attached to the top of a steel fence post approximately four feet above the ground. The gummed-paper is mounted on a thin aluminum frame and the frame attached to the plate with binder type clips. The effective surface area of the gummed-paper presently in use is 12 square inches. The samples are collected bi-weekly from all stations. Each sample is scanned with a G. M. instrument to determine if a detectable amount of activity has

been collected, following which the samples are covered with a thin polyester film and exposed to X-ray film for seven days and then developed in the normal manner. The presence of particulate radioactivity on the sample is indicated by dark images on the film. In a study undertaken to determine the efficiency of this monitoring procedure, Rosinski (1957) found that it does not provide a precise or quantitative means of determining radioactivity. However, when carried out routinely, it serves as an economical means for detecting changes in the rate of fallout.

As a result of this monitoring program an increase in the amount of radioactive particulate fallout was noted at stations surrounding the Idaho Chemical Processing Station (ICPP) during the summer of 1958. (The location of stations relevant to the MTR-ETR and ICPP are shown in Figure 1.) The increase started about the last week in July and reached a peak during the first week in September. At station "E" which is located approximately 100 feet east of the 250 foot stack, only three small particles were detected during the months of May and June. For the period of July 25th through September 5th an average of one particle per day was detected at this station. If it is assumed that the size of the image on the X-ray film is proportional to the amount of activity the particle contains, then there was an indication that the amount of activity per particle had also increased. However, this was not borne out statistically and particle size could be the controlling factor.

A check of gummed-paper samples from other areas on the NRTS did not reveal any increase in particle deposition. Therefore, it was assumed that the most logical place to check was the ICPP. As a follow-up of the gummed-paper program, a field survey of the area was conducted. Four plots, each approximately three-feet square, chosen at random within a 50-foot radius of the gummed-paper stations were surveyed with a G.M. instrument and a record maintained of the number of particles detected. (Figure 2 shows the results of this survey.)

Because of the size of the particles the position of maximum deposition is believed to have been predominately controlled by the terminal settling velocity of the particle and not from vertical atmospheric turbulence. The terminal settling velocity of a 300 micron size particle with a specific gravity of 1.0 settling in air at 70°F is approximately 3 feet per second. Then, neglecting the effect of vertical turbulence, this particle falling from a height of 250 feet in a 10 mph wind would likely be deposited approximately 1,200 feet downwind from its release point. This agrees with the distance of maximum deposition found from the survey. During the field survey, samples of soil containing particles were collected for laboratory analysis. At the laboratory the soil samples were divided and sub-divided and a thin end-window probe used to locate the radioactive particle. To finally isolate the radioactive particle from the nonradioactive particles considerable time was required until the microscope was fitted with a special attachment. This attachment was modeled after one discussed by Mr. F. E. Adley in the April 1958 issue of the Industrial Hygiene Journal. Primarily it consists of a "collimating attachment," mounting plate, and a thin end-window probe which have replaced the microscope sub-stage condenser. (Figure 3 shows the special attachment.) Two "collimating attachments" were made; one with a hole .125 inch in diameter and the other with a hole .024 inch in diameter. The probe, which is usually flush with the bottom of the "collimating attachment", is connected through an amplifier to provide an audible signal. The specimen slide is moved over the hole until the signal indicated that the radioactive particle is over the hole. Usually the attachment with the .125 inch diameter hole is used. Once the radio-

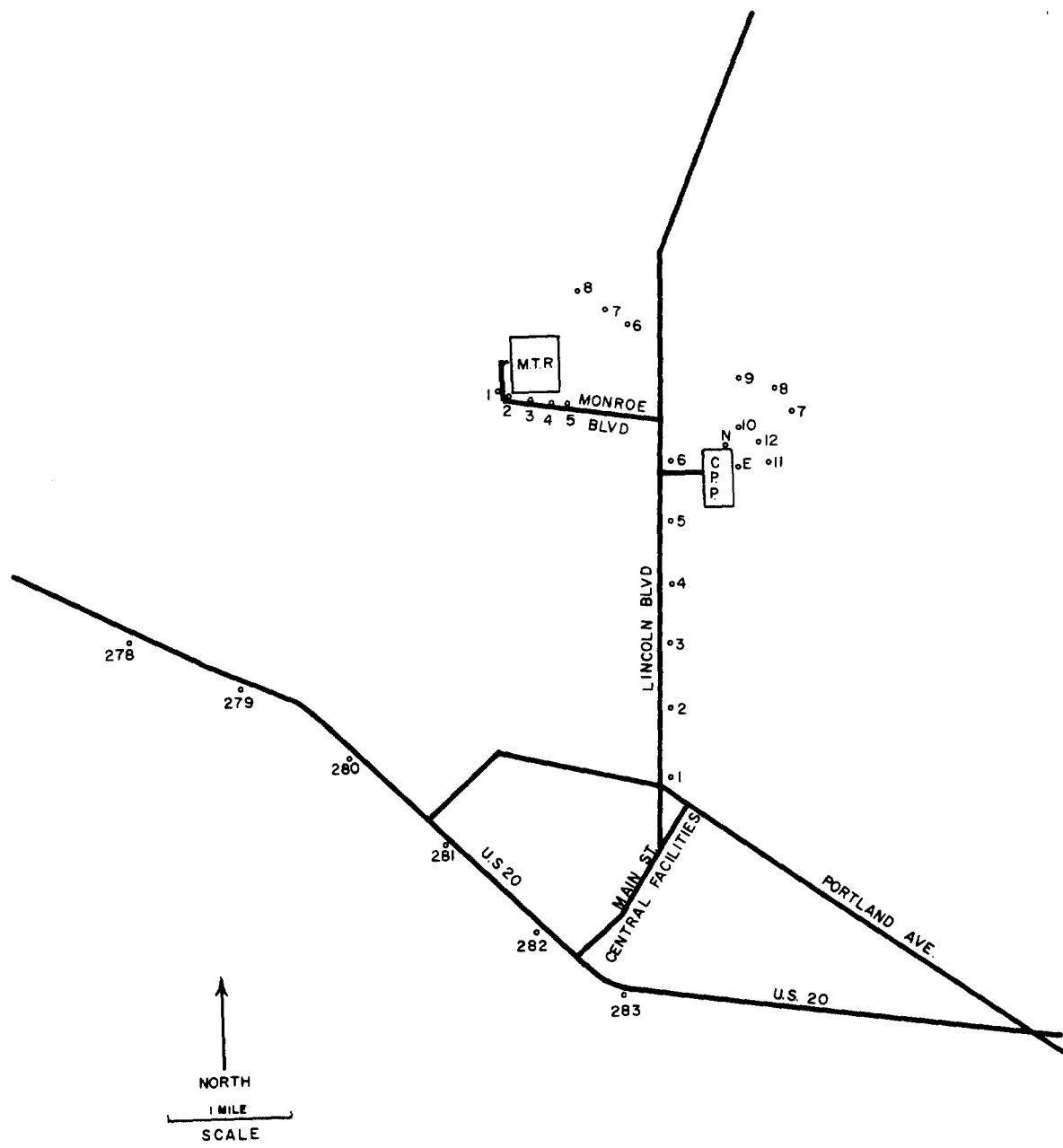


Fig. 1 — Fallout stations around MTR, ETR, and ICPP.

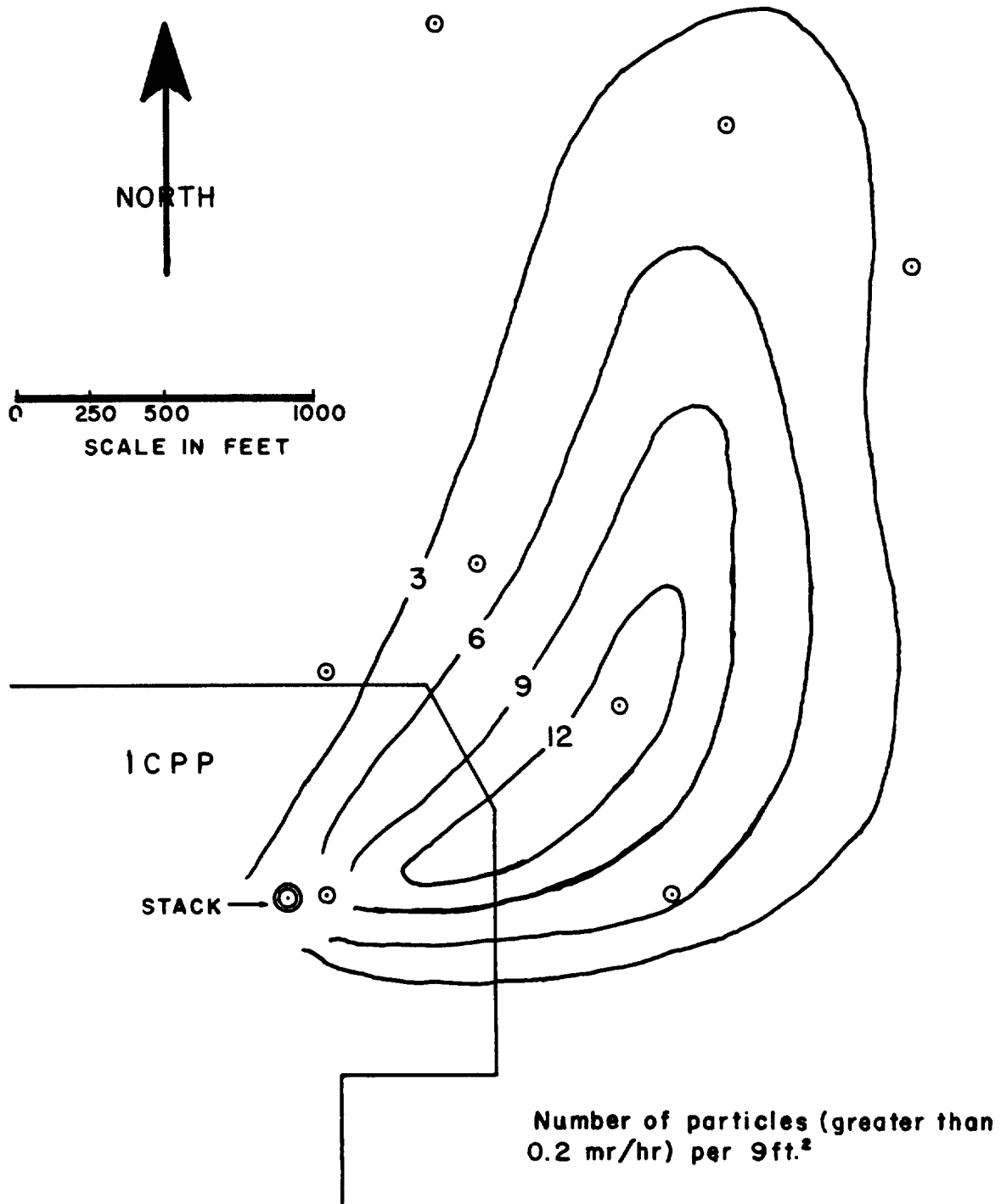


Fig. 2—Concentration of particles on the ground downwind of the ICPP stack.

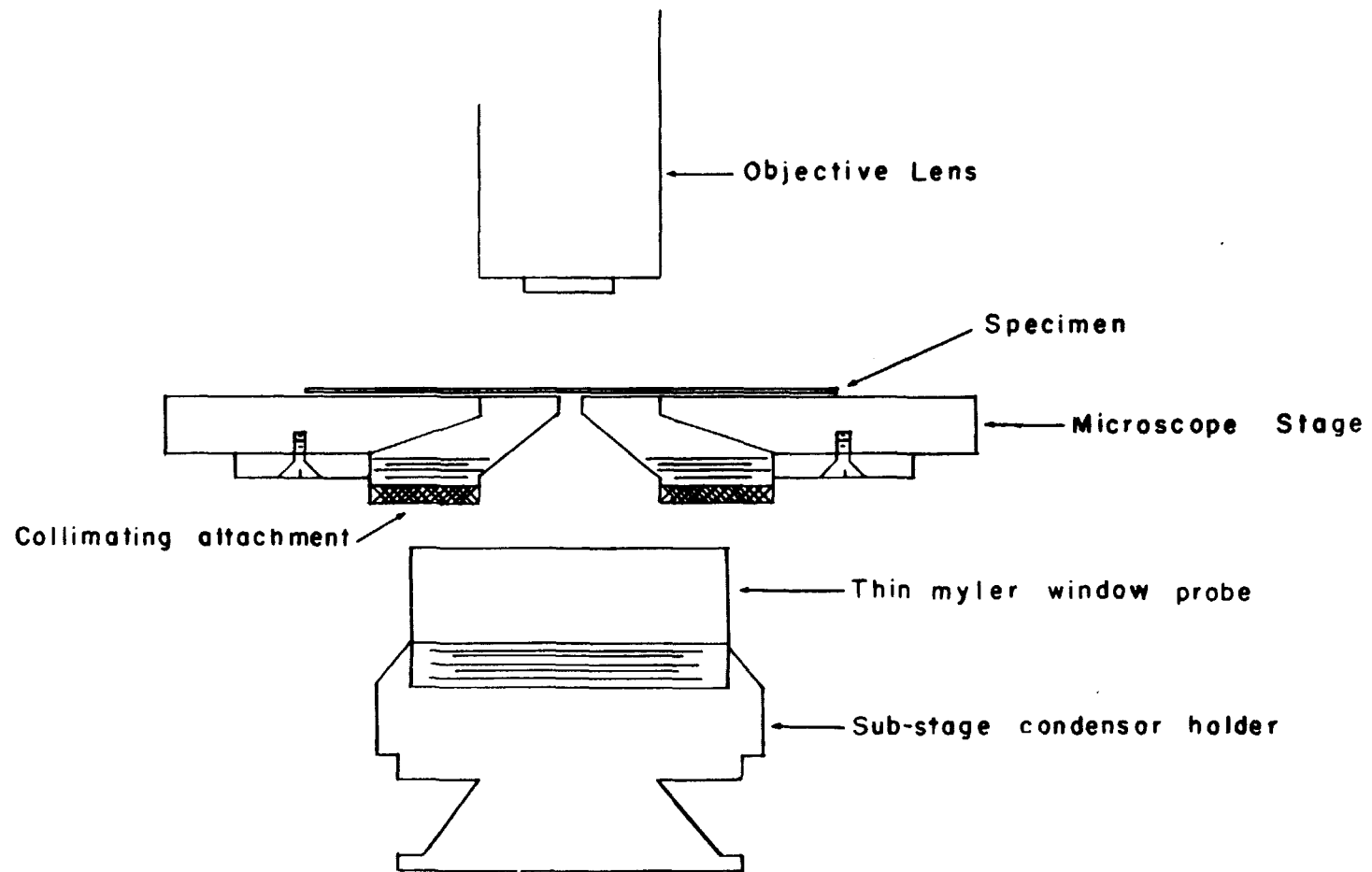


Fig. 3—Special microscope attachment used to facilitate isolation of radioactive particles.

active particle is located the attachment with the smaller holes is used to finally locate the particle. By successively moving the specimen slide in several directions it is possible to locate the radioactive particle. With this attachment considerable time is saved in isolating particles. After isolation the size of the particles is determined. The particle sized varied over a wide range, from 70 to 350 microns. However, this does not exclude the existance of smaller particles. The activity emanating from the smaller particles is not detectable with the G.M. survey instruments. Tiny images on the X-ray film definately indicate that small particles were involved, but the small amounts of activity present did not permit any evaluation of these particles.

Calculations of absorbed dose for two critical organs were made from gamma ray spectrographic analysis of particles collected downwind from the 250' stack at the Chemical Processing Plant. These were the "hottest" particles noted on fallout plates in the area. It was found that the particle indicating the most cerium-144 contained 0.1 microcurie of this isotope. The particle containing the highest amount of zirconium-95, niobium-95 contained 0.01 microcurie of these isotopes.

For dose calculations the zirconium-95, niobium-95 activity was assumed to be all niobium-95, since this isotope presents a greater biological hazard than does zirconium-95. Evaluations were made to determine what dose to the bone could be expected if such a particle were inhaled and were completely solublized even though the latter is remote. For calculations of dose from soluble constituents, 100% solubility was assumed. In addition to the above, the following assumptions were made:

1. For the case where the bone is the critical organ, the isotope is eliminated from the body exponentially.
2. All NBS Handbook 52 values apply.

Table I presents the maximum dose in millirads from a single particle calculated as Cerium-144 and Niobium-95, with both the bone and the G.I. tract as being the critical organs.

TABLE I

DOSE DUE TO INHALATION

Isotope	Critical Organ	Soluble	Insoluble
Cerium-144	Bone	25. mrad	--
Niobium-95	Bone	0.05 mrad	--

DOSE DUE TO INGESTION

Isotope	Critical Organ	Soluble	Insoluble
Cerium-144	Bone	0.05 mrad	--
Cerium-144	G.I. Tract	17. "	17. mrad
Niobium-95	Bone	0.05 "	--
Niobium-95	G.I. Tract	0.02 "	0.02 mrad

It must be emphasized that the values given in the above table represent the maximum doses and may make the problem appear greater than it is in actuality. All assumptions were necessarily made to maximize any hazard. The fallout is not considered to be an inhalation hazard since all of the particles isolated have been far above the respirable range of 0.1

to 5 microns. The hazard due to the smaller particles was not considered since these particles contained only a small fraction of the total radioactivity.

Since construction workers were often within the fallout area, it was necessary to consider the probability of transferring a particle from the ground to the face area where inhalation or ingestion might take place. Schwendiman (1956) has estimated that in an area where surveys show one particle per square foot, a man may walk on the average of 2,000 hours before a particle will be deposited in the face area. In Schwendiman's study the mean particle diameter was 2 microns and the probability of inhaling larger particles should be even smaller. Therefore, no restrictions were placed on the area surrounding the ICPP and no restrictions were imposed upon construction workers in the area. Surveillance of the area continued while ICPP Operations sought a solution by locating and correcting the leak.

In conclusion, it should be pointed out that the use of a network of gummed-paper stations does provide an economical means of monitoring for fallout in the vicinity of a plant utilizing radioactive material. Once fallout is detected the assistance of plant operations is needed to pinpoint the source of the activity. Throughout the entire time that this problem existed, liason was maintained with ICPP Operations. The results of their findings are reported in paper by Cederberg and Bower (1959).

#### REFERENCES

- Eisenbud, M., and J. H. Harley, Radioactive dust from nuclear detonations, Science, 117, 141-147, 1953.
- Eisenbud, M., and J. H. Harley, Radioactive fallout in the United States, Science, 121, 677-680, 1955.
- Eisenbud, M., and J. H. Harley, Radioactive fallout through September 1955, Science, 124, 251-255, 1956.
- Holland, J. Z., AEC Atmospheric Radioactive Studies, WASH-1016, April 1959.
- Rosinski, Jan, Some Studies on the Evaluation of Gummed-Paper Collectors Used in Determining Radioactive Fallout, Transactions, American Geophysical Union, Vol. 38, No. 6, December 1957.
- Adeley, F. E., Instrument Developements in Health Physics, Industrial Hygiene Journal, Vol. 19., No. 2, April 1958.
- Schwendiman, L., Probability of Human Contact and Inhalation of Particles Health Physics, Vol. 1, No. 3, December 1958.
- Cederberg, G., and J. Bower, Minimizing Gaseous and Particulate Activity and Particulate Activity in Idaho Chemical Processing Plant Off-gas, (To be published.)

## Discussion \*

Q. Any α activity measured in this matter?

A. None

Q. It wasn't measured or wasn't found?

A. (?)

Q. Could you not get a small particulate added by turbulence bringing it down to 1200 feet?

A. Yes, etc.

Q. Did you actually measure the size of the particulate?

A. Were measured.

Q. Haven't you found that it did not -- however, when carried out routinely it does serve as a means of detecting - in cleaning the amount of radioactive particulates was noted in the - while in some instances it might have indicated the dose quite high, - minimize any hazard.

A. No restrictions were placed on the area surrounding the CPP. The monitor program continued through the problem and we maintained consistent contact with the CPP operations group to assist them in determining the source of the problem - the results of their findings will be reported in the next paper by Mr. Cederberg and Mr. Bower.

### \* NOTE:

All the questions, answers and comments were recorded on tapes. Later, when the tapes were transcribed it was noted that some of the material was incomplete and in several cases it may have been improperly interpreted. Due to the time involved, no attempt has been made to change this material.



# MINIMIZING GASEOUS AND PARTICULATE ACTIVITY IN IDAHO CHEMICAL PROCESSING PLANT OFF-GAS

G. K. CEDERBERG and J. R. BOWER  
*Atomic Energy Division, Phillips Petroleum Company,  
Idaho Falls, Idaho*

## A B S T R A C T

Bench-scale tests on iodine removal from RaLa process off-gas, reported at the last Air Cleaning Conference, were continued and evaluation tests of the Plant charcoal beds have been completed. About 97 percent iodine-removal efficiency was demonstrated. Iodine-containing particles possibly formed by the gas-liquid contactor preceding the charcoal beds, limit the efficiency of the bed in removing iodine.

Radioactive particles are formed in the Fuel Element Cutting Facility by cutting, carrying, and charging techniques. Cyclones and filters are used for collection.

Studies of particles originating in the vessel off-gas system and the waste solvent incinerator are directed towards minimizing their frequency and activity level.

\* \* \* \* \*

This discussion of Idaho Chemical Processing Plant off-gas cleaning problems involves the removal of iodine from off-gas formed by the RaLa process and the diminution of particulate activity from other ICPP equipment off-gas.

### Iodine Removal

The RaLa process produces barium-140 from a 2-day cooled MTR element by digesting the aluminum in the element in sodium hydroxide and by chemically separating the barium from the uranium-gross fission product residue. Because of the two days cooling, compared to 120 days cooling normally given to other fuel elements, this process contributes large quantities of fission product iodine to the process off-gas. The majority of the iodine dissolves in the caustic-aluminate and gaseous iodine is removed in a packed scrubber.

The objective of the iodine studies was to lower the iodine concentration in the off-gas by preventing it from volatilizing from solutions and by more effectively scrubbing it from the off-gas. The RaLa caustic scrubber, containing 1M NaOH and 0.1M Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub> as the scrub solution, gave a decontamination factor of 100 to 500 on the gaseous iodine but this was not consistent and not completely adequate. Numerous tests scoped and evaluated a variety of materials for removing more of the gaseous iodine which passed through the RaLa scrubber. Some of these tests were reported at the last air cleaning conference.<sup>(1)</sup> Activated charcoal was concluded to be the most efficient and practical of the materials tested because of its high efficiency at room temperature and ease of handling. Development tests showed a 1-inch diameter by 12-inch long bed of 8 to 14 mesh activated charcoal gave a decontamination factor of 33 at a superficial flow velocity of about 0.2 feet per second.

Based on the development work, two 7-cubic foot beds of activated charcoal, containing extruded pellets of 2.8 mm diameter, were installed to treat from 15 to 20 cfm of RaLa off-gas. The RaLa equipment is illustrated by Figure 1. Normally this off-gas is treated separately from other plant off-gas streams although it can be valved to go directly to the plant vessel off-gas system. The charcoal beds can be operated individually or in parallel with the exit gas from the charcoal bed passing through a sintered stainless steel filter to prevent charcoal fines from passing to the stack.

Evaluation tests of the plant charcoal beds showed they give decontamination factors between 10 and 30. This was determined by the use of small filter-adsorber cartridges constructed of a filter medium and activated charcoal in a polyethylene tube. Identical cartridges were used to sample equal flows of the inlet and outlet gas of the charcoal bed.

Although the literature<sup>(2)</sup> cites activated charcoal as giving a decontamination factor of 1000 or better for elemental iodine, I<sub>2</sub>, the lower efficiency observed in the tests is attributed to small particles entrained from the scrubber. Apparently small droplets containing NaOH-Na<sub>2</sub>CO<sub>3</sub>-Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>-NaI are formed at the surface of the scrubber and entrained, dried, and carried through the charcoal bed. Disassembly of the outlet sample cartridge showed that the filter contained a considerable proportion of the total activity on the cartridge with no indication that the retained particles were radioactive carbon fines.

Comparison of the parts in the inlet and outlet cartridge after one test showed that the filter medium preceding the charcoal cartridge was 8.5 times more radioactive than the filter in the outlet cartridge. The charcoal in the inlet sample cartridge was 2800 times more radioactive than the charcoal in the outlet sample cartridge. Cartridges used in other tests showed smaller ratios of the total activity on the inlet and outlet cartridges, but the activity on the filters of the cartridge was still indicative of particles.

Victoreen radiation monitor heads on the sides of the beds indicated that the inlet surface of the bed was from 500 to 5000 times more radioactive than the outlet of the bed. Decay rates of the activity on the inlet surface indicated it was predominantly iodine-132.

The peak radiation at the inlet surface of the bed varied from 60 Roentgens per hour to greater than 1000 Roentgens per hour.

At the last conference no applicable chemical additive had been found which was compatible with the caustic-aluminate waste solution,

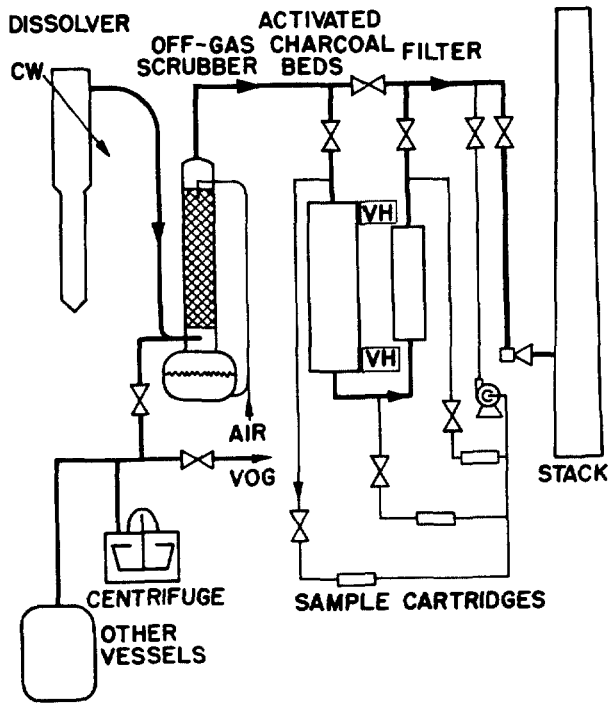


Fig. 1—RaLa iodine removal system.

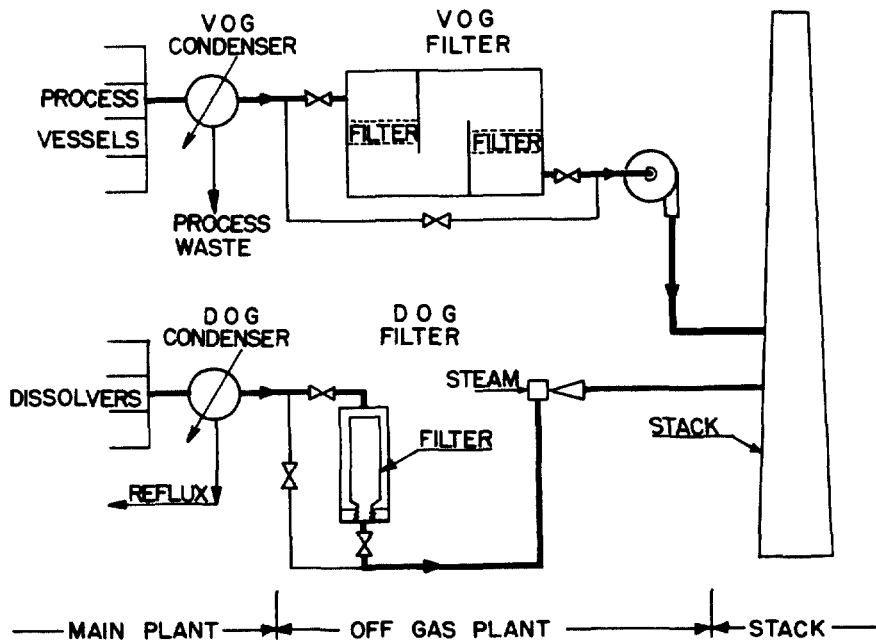


Fig. 2—Plant off-gas systems.

containing about 90 percent of the iodine, and which could more effectively hold the iodine in solution during acidification. Mercuric nitrate in the caustic would be precipitated and the use of mercuric nitrate in the uranium-fission product mixture prior to the barium separation was ruled out because of potential product contamination. However, mercuric nitrate has recently been added to the 60 percent nitric acid used to neutralize and acidify the waste caustic-aluminate solution and to the water used to dilute the uranyl nitrate-fission product nitrate solution remaining after the barium separation. In these solutions from 0.001M to 0.005M mercuric nitrate has effectively limited the volatilization of iodine during later transfers and samplings.

In another recent process change, suggested by work done at HAP0,(3) five percent nitric acid containing 0.001M mercuric nitrate and 0.001M mercurous nitrate has replaced the 1M NaOH-0.1M Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub> previously used as the RaLa off-gas scrubber solution. Although the NaOH-Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub> effectively scrubbed the iodine (I<sub>2</sub>) from the off-gas, it contributed entrained solids, predominantly NaOH and Na<sub>2</sub>CO<sub>3</sub>, into the outlet pipe. This plugged the rotameter and partially plugged the scrubber column. Subsequent runs showed that the HNO<sub>3</sub>-Hg(NO<sub>3</sub>)<sub>2</sub>-HgNO<sub>3</sub> scrub solution eliminated the plugging and provided adequate scrubbing.

After mercuric and mercurous ions were added to the waste solutions and to the acid scrub solution, the peak activity at the inlet surface of the carbon bed during several runs was between 10 and 20 Roentgens per hour compared to the 60 to 1000 Roentgens per hour observed previously. This demonstrated that a smaller amount of iodine was volatilized from the process solutions.

Results from the stack gas monitor have confirmed the effectiveness of mercuric and mercurous ion in suppressing iodine release. When the waste streams are transferred from the RaLa cell to other cells the RaLa scrubber can no longer be used and any iodine released into the off-gas passes directly to the stack. Since the addition of mercury to the waste streams, the amount of iodine released from the stack to the atmosphere has been significantly reduced.

#### Particles in Regular Plant Off-Gas

The problem of radioactive particles in other ICPP equipment off-gas came into focus when hot particles were observed on gummed-paper fall-out plates stationed around the ICPP. The Site Survey Branch of the IDO Health and Safety Division routinely monitors the NRTS area with gummed-paper fall-out plates. Beginning about July, 1958, plates from the stations on the northeast perimeter of the CPP collected an unusually large number of particles.

Part of our investigation involved taking samples from all of the possible sources. The samples were used to determine if the material taken from any potential source was similar in physical and radiochemical properties to the particles on the fall-out plates. The data from these samples provided a basis for remedial action to minimize the dispersion of particles into the effluent off-gas from the CPP process buildings.

Some of the observed particles were about 70 microns (barely visible) although many were much smaller. The activity varied from 16 milli-roentgens per hour to barely detectable. Some particles which had a very low activity and which were not detectable with an open window beta counter were detected by the small dark spot they made on X-ray film during autoradiographing. The fall-out plates were autoradiographed by a week-long exposure to an X-ray film.

In all possible cases the particles were examined, either with or without magnification, to determine the color and physical characteristics of the material. Gamma pulse height analyses of particles and samples by CPP and AEC analysts helped identify some of the particles as having come from specific sources. Some of the particles were yellowish-colored crystals showing a high percentage of cerium-144 in the gamma spectrum as well as other long-lived fission products. This indicated this was very old material which had probably been formed with essentially no selectivity.

Solid material found around the discharge of the vessel off-gas (VOG) system blower was very similar in color and gamma spectrum to the observed particles. It was concluded that the vessel off-gas and the dissolver off-gas (DOG) system could be emitting particles as a result of corrosion in the filters. These systems are shown schematically in Figure 2.

Although no substantial increase in the number of particles had been detected by Site Survey before July, 1958, a study of the DOG and VOG systems showed no positive reason that particles were not emitted prior to this. The high levels of radiation near the VOG and DOG systems leaves little doubt that these systems have accumulated radioactive contamination from entrained liquids and solids from plant vessels since the plant began operation in 1953. Although both systems have filters to remove this material, observation of the outside of the VOG filter showed it to be corroding.

According to the VOG filter drawing details the filter medium is Fiberglas but the containment box is carbon steel. The carbon steel box has been repaired in times past by welding metal patches over some of the corroded holes and by putting patches of masking tape over other holes. Even though the corroded condition could allow particles to pass unfiltered to the stack, the important aspect is that corrosion products, which have collected in the plenum chamber on the outlet side of the filter medium, could be stirred by air turbulence and carried into the off-gas.

The vessel off-gas has traces of oxides of nitrogen and small concentrations of water vapor. Although the off-gas condenser cools the gas to between 15 and 25°C, small changes of temperature in the VOG system could allow water to condense and dissolve some of the corrosion products. Migration of the solution with the off-gas and later increases in temperature could redeposit the solids at a different location in the system. The solid crust forming on the outside of the outlet flange of the VOG blower, possibly the crystallization of the corrosion products, indicates there are liquids and solids in the VOG system between the filter and the stack. The gamma ray spectrum, showing a predominance of cerium-144, and the yellow color of a sample of the crust were similar to the spectrum and color of some of the particles found on the fall-out plates and on the ground around the CPP area. This proves that the VOG system can be a source of particles.

The gas flow through the VOG filter has been continuous, but sometime in the history of the VOG system the VOG filter bypass was opened. About July of 1958, several attempts were made to decrease the absolute pressure in the VOG headers in the main process building for the purpose of eliminating intermittent pressurizations which occurred in dissolving cell. These changes were made by adjustment of the air bleed rate in the VOG blower room and adjustments of the filter bypass valve. The adjustments could have stirred up some of the particles of corrosion products in the filter and some of the fission product-corrosion product particles lying dormant in the bypass line.

The DOG filter vessel is fabricated of stainless steel but the filter insert is a carbon steel shell containing the filter medium. Since the dissolver off-gas normally contains some nitrogen dioxide and waste vapor, it is almost certain that the filter cartridge is highly corroded. Samples of debris from the DOG system were unavailable because of its all-welded construction.

Coincident with the appearance of particles the ICPP was being operated at a high rate. Off-gas rates varied considerably and very likely could have stirred up solids existing in the lines and related equipment.

In order to minimize the dispersion of particles, the VOG filter was completely bypassed and it was recommended that a drum should be installed to provide de-entrainment of liquid droplets and particles. It was also recommended that the DOG filter element should be replaced with a new all-stainless steel element as soon as operating conditions would permit. These recommendations have not yet been carried out.

During the recent operating interval from November, 1958, to May, 1959, the frequency of occurrence of radioactive particles was much lower than during the July to September interval in 1958. Although the plant was at times operating at a somewhat reduced rate, this should have been a minor factor in the number of VOG particles which were released. Apparently, the action taken to bypass the VOG filter helped reduce the number of dispersable particles.

#### Solvent Burner Particles

The possibilities of particles being formed by the waste solvent burner were also considered in connection with the above problem. Waste Amsco, a kerosene-type hydrocarbon, and waste Amsco containing tributyl phosphate (TBP) are accumulated from the first cycle extraction columns and burned at the stack in a specially designed burner, illustrated in Figure 3. Although the operation of the solvent burner and the appearance of the particles suggested a relationship of the burner to the particles, the dissimilarity of the gamma spectrum of the larger particles found on the field and of the sample particles taken from the burner duct indicates that the solvent burner is not the major contributor. A lack of correlation between the appearance of particles on the Site Survey plates from week to week and the intervals of operation of the burner also indicates the burner is not the major contributor.

Since Amsco containing traces of TBP absorbs zirconium better than it absorbs ruthenium and other fission product, zirconium and niobium and only traces of other fission products will appear in the burner effluents. Some of the activity eventually deposits on the walls of the fire box and exit ducts with the soot. Apparently, during normal operation this residue of soot and fission products is continuously eroded from the surfaces and carried into the effluent gases. The majority of the gas, carrying these eroded particles is mixed with all the plant ventilation air and is discharged from the 250-foot stack to the atmosphere. It is possible that these particles were too small to be detected individually and only caused a pin-point of exposure on the radioautograph. However, some of the burner effluent gas puffs out the bottom openings of the duct venturi and, possibly, some larger particles are scattered in the immediate vicinity of the burner building and the base of the stack. Larger particles, appearing black and, sometimes glassy and having a high zirconium-95 concentration, have been found in the vicinity of the stack although they were not observed on the fall-out plates. These particles resembled the sample taken from the burner duct.

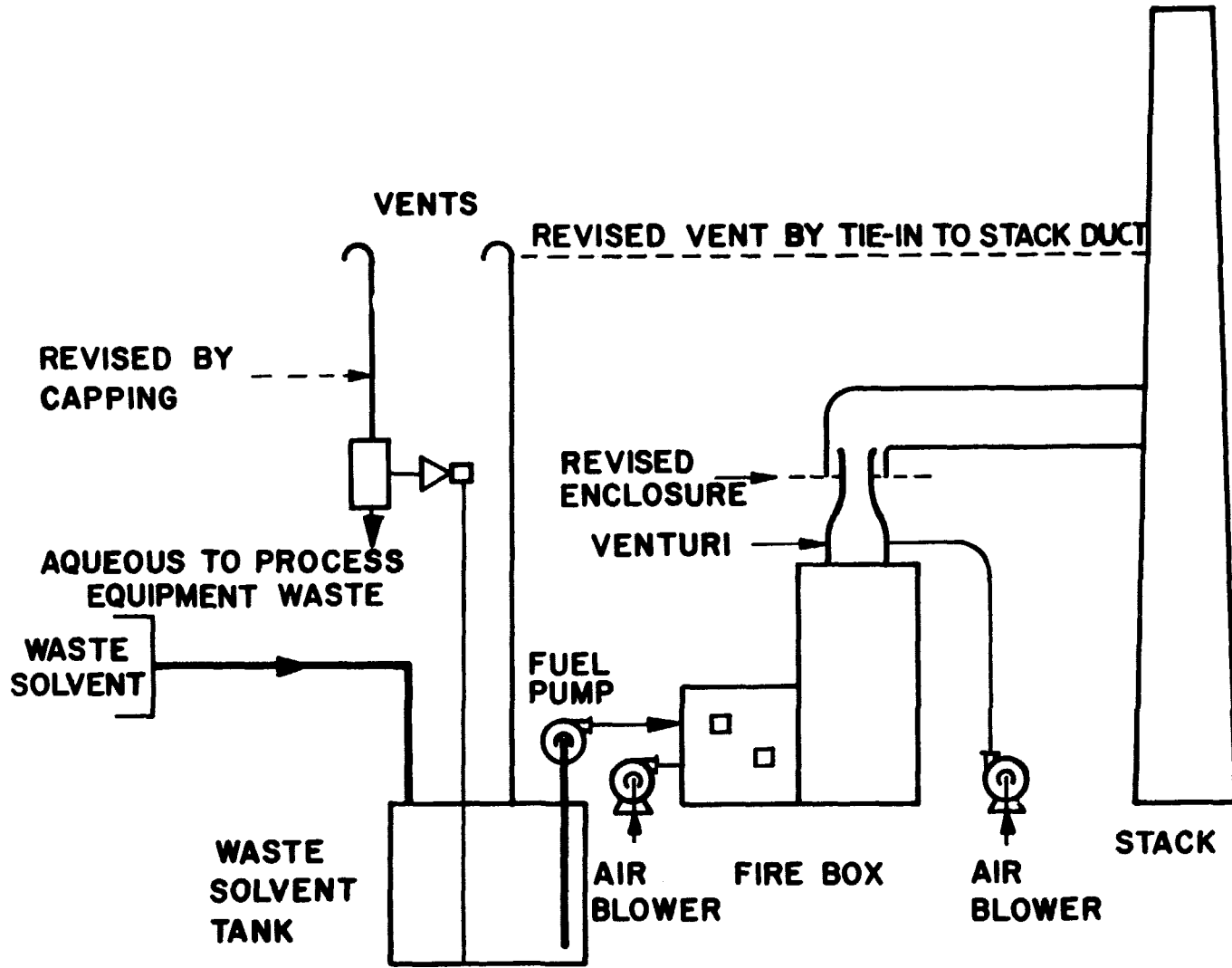


Fig. 3—Waste solvent burner system.

Although the organic waste generally has a low activity level, it is possible for small volumes of aqueous-organic mixtures to be sent to the waste solvent collection tanks. Recently, in an attempt to minimize the activity, the solvent has been washed with an aqueous sodium carbonate solution which is decanted to the aqueous waste facilities. Prior to the introduction of the washing procedure some highly radioactive column interface crud, probably an aqueous-organic emulsion, was sent to the burners. This caused the radiation levels of the burners and ducts to increase from 0.1 Roentgen per hour to 1.5 Roentgen per hour with maximum radiation levels up to 4 Roentgen per hour.

The decanting jet on the solvent collection tank was also a source of particles. After decanting the aqueous layer from the tank, the Amsco in the organic-aqueous interface vaporized in the jet. This vapor rose in the vent to the roof of the solvent burner building and came out of the vent as a mist. Some of this mist coalesced on the top of the building and some settled randomly down-wind from the burner building. This was remedied by repiping the decanter equipment directly to the process equipment waste system and piping the vessel vent to the plant vent duct. The interim remedy was to control the jetting and, if possible, stop the jetting before the organic-aqueous interface reached the jet intake.

Radioactive carbon particles which have escaped from the openings in the burner equipment have been detected on the flat surfaces around the burner facility. These have been minimized by sealing the opening of the exit duct venturi. The exit duct venturi was designed to admit ambient air to help cool the burner exhaust gases. The burner has been operated continuously at lower rates to prevent gas from puffing out of the burner during stop and starts. This lower rate has helped keep the exit duct from over heating although some other cool dilution air is provided by a compressor.

The flat equipment surfaces and the ground around the burner building have been cleaned to prevent the wind from carrying previously deposited loose particles from the solvent burner area.

#### Other Particle Sources

Other particles are formed in the Fuel Element Cutting Facility (FECF) during the cutting of aluminum-uranium fuels, but these particles were not similar to the other hot particles seen on the plates. Some of these particles, though adhering to the cut pieces, sift from the transfer cask but are collected in a pan under the cask. Other particles in the FECF are air-borne and are collected in cyclones and AEC-type air filters built into the FECF. One of these air filters ruptured during a cell decontamination procedure and caused some contamination of the environs. Although some of the contamination was scattered by the winds, the majority of it was isolated into the drains by water flushing. Other areas which were contaminated by this activity were isolated as limited access areas.

Other possible sources were also investigated but were found to be insignificant.

Although the RaLa off-gas system contains some particles of activated charcoal as well as the previously mentioned residual from the scrubber, this system was not deemed to be a significant source of particles because short half-lived isotopes such as iodine-131 were not detected on the fall-out plates. The activated charcoal is closely confined by perforated support plates and filters.



## References

1. Paige, D. M. et al, "Two Gas Cleaning Problems at the Idaho Chemical Processing Plant Site," Fifth Atomic Energy Commission Air Cleaning Conference held at the Harvard Air Cleaning Laboratory, June, 1957, TID-7551, USAEC Technical Information Service, Oak Ridge, Tennessee.
2. Finnigan, J. W. et al, "Removal of Iodine Vapor from Gas Streams by Sorption on Charcoal," HW-26113, 1952, Unclassified.
3. Bupp, L. P., "Monthly Report January 1958," Chemical Research and Development Operation, HW-54760 C, February 10, 1958, Secret.

## Discussion

- Q. At this stage of the game the Rala system does not require meteorological control? Is this correct?
- a. Still provide forecast but not under control it used to be. Primarily for site survey monitoring system. Forecast for site survey for Rala system.
- Q. Is one to get the impression from that, that the site survey problems associated with it are less than they have been in the past?
- A. Yes, I believe it is considerably reduced. It has now been reduced - we do routine monitoring, but not nearly as extensively as previously.
- Q. (John Hall, United Kingdom) What sort of filter do you have in your rala - in the charcoal beds?
- A. Stainless steel filter.
- Q. (C. E. Lapple - Stanford Research Lab.) Do you have any data on how long they did last or how often you need to replace them?
- A. No, we do not. We feel it will have a very long life - equipment - and material can be removed later in the life.
- Q. (R. C. Walker, GE - HAPO) I would like to know two things - what is the velocity through the charcoal beds, is this in df in the two stages or in each stage?
- A. They are not in series - they are either individual or parallel. Velocity through the bed 7 cu. ft. Bed can handle 7 cu. ft. of material off-gas.

## A METHOD FOR THE COLLECTION AND IDENTIFICATION OF RADIOACTIVE XENON AND KRYPTON

J. KENNETH FLYGARE, JR., GEORGE WEHMANN,  
ALAN R. HARBERTSON, and CLAUDE W. SILL  
*Health and Safety Division, U. S. Atomic Energy Commission,  
Idaho Falls, Idaho*

### Abstract

The Health and Safety Division is responsible for radioactive monitoring of out-plant areas of the National Reactor Testing Station. In order to determine if a leak existed in the RALA off-gas system a sampler capable of collecting radioactive xenon and krypton was required so that identification could be made. A sampler consisting of a "U"-shaped copper tube filled with activated carbon and cooled in liquid nitrogen has proved very effective. Two sizes have been made using 3/4" and 1-1/2" tubing. The collection efficiency for xenon and krypton was essentially 100% at flow rates of 1 cfm even with the smaller sampler. Several such samplers were used at strategic locations and disclosed some leaks in the process system. Retention of xenon and krypton in activated carbon at room temperature is also discussed.

\* \* \* \* \*

The Health and Safety Division is responsible for radioactive monitoring of out-plant areas of the National Reactor Testing Station. During a RALA operation for the recovery of barium-140 from short-cooled fuel elements (1), activity was detected in the field at a time when all off-gas was supposed to have gone to storage. The activity could be detected with portable survey instruments but could not be collected on either filter paper or activated carbon. Shortly before the activity was discovered, multi-curie quantities of radioactive xenon and krypton had been released into the off-gas line from the caustic dissolution of the aluminum cladding of the fuel element. Rare-gas fission products were suspected, apparently from a leak in the RALA off-gas system. Such a leak would be difficult to detect because of the high general radiation background in the plant. A sampler capable of collecting the inert gases was required so that the activity could be identified.

Since the melting points of xenon and krypton are  $-112^{\circ}$  C. and  $-156.5^{\circ}$  C., respectively, liquid nitrogen with a boiling point of

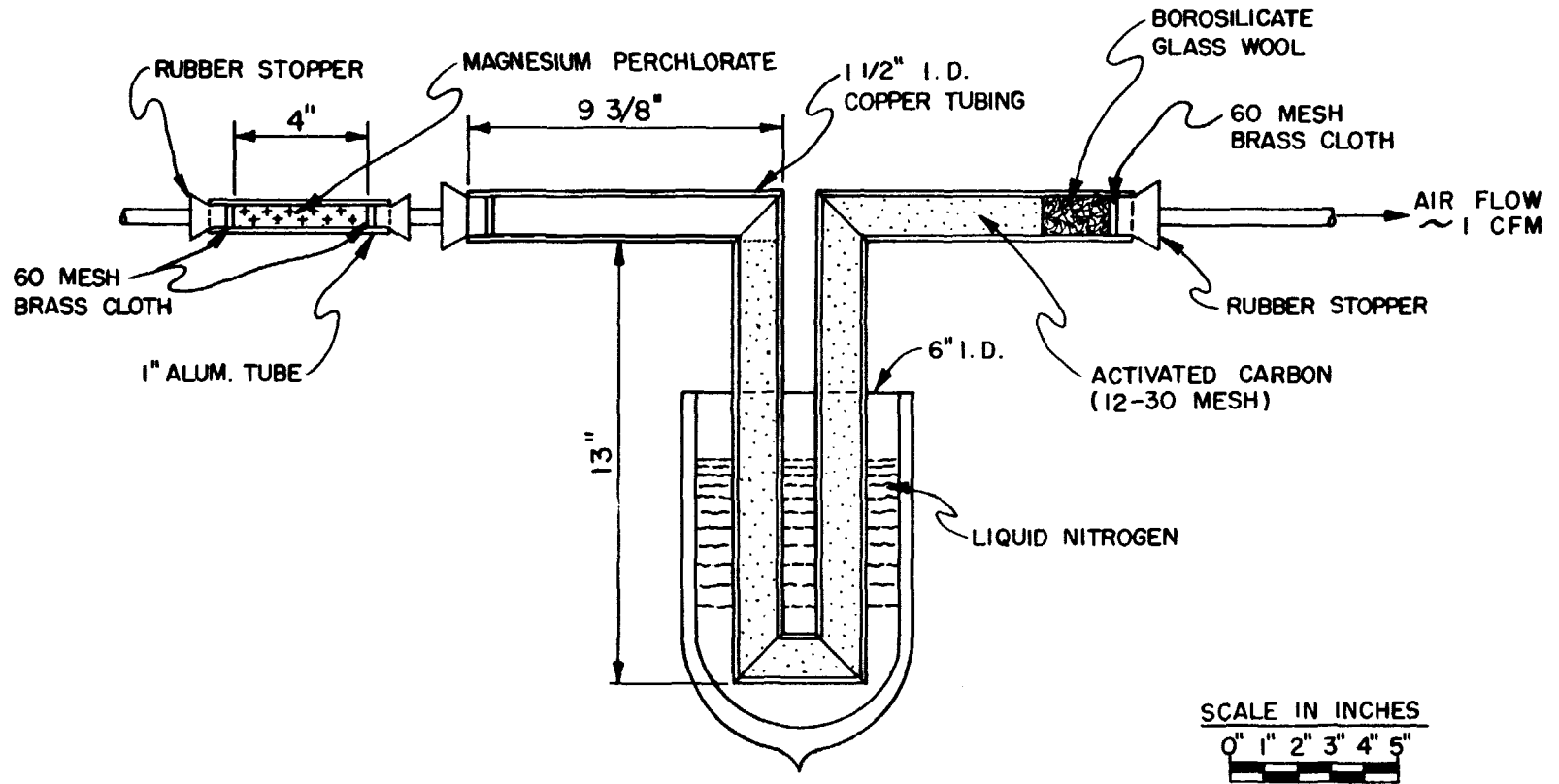


Fig. 1 — Large sampler system.

-195.8° C. (2) should be adequate to freeze xenon and krypton out of the air stream efficiently. Browning and Bolta (3) determined the holdup time of krypton on charcoal down to -110° C. and reported that retention increases with decreasing temperature. Activated carbon was used to obtain a large surface for adsorption and to provide large heat capacity for cooling the gas stream.

The sampler is shown in Figure 1 and contained 450 grams of 12 to 30 mesh activated carbon when filled as shown. The borosilicate glass wool was used to prevent the carbon from packing against the brass screen. The air stream was passed through a column of 10 to 20 mesh anhydrous magnesium perchlorate to remove moisture to prevent the cold trap from freezing shut. The samplers were operated for periods up to one hour without the drier becoming saturated. The rate of flow of air was dependent upon the drier and not the carbon trap. A 3" piece of 1/2" copper tubing connected the drying unit to the sampler. The drier also served as a filter for any particulate matter that might be in the air.

The copper tube filled with carbon was set 9" deep in a 4300 ml. Dewar flask and clamped to the steel arms of the frame holding the flask. During operation the level of liquid nitrogen was maintained at 2" to 4" below the top of the flask. The rate of evaporation was a function of the depth of the liquid nitrogen in the Dewar flask. A sampler in operation lost one liter in about twenty minutes with a total loss of approximately 2600 ml. in one hour. The 1600 ml. left in the flask was about the minimum amount required for satisfactory operation.

Six sampling units were constructed and placed at strategic locations both in the field and inside the Idaho Chemical Processing Plant during a RALA operation. The average rate of flow of the air through the samplers was 1 cfm. Radioactive xenon and krypton were found in quantities up to 3 r/hr at contact in all but one of the samplers with the intake side containing essentially all of the

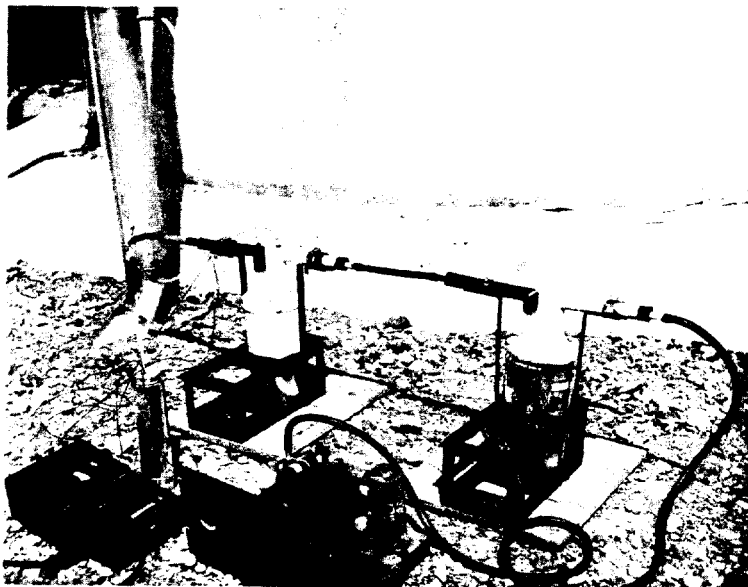


Fig. 2—Two large samplers in series.

activity. Two samplers were placed in series as shown in Figure 2 to determine the collection efficiency. The collection efficiency of

the sampler was very nearly 100% and dropped as low as 99% in only one instance. Xenon-133 and krypton-85 were the only isotopes found in concentrations of any significance. Traces of xenon-135 and krypton-85m were detected. Identification was made by gamma ray spectroscopy using a multichannel analyzer with a 3" x 3" sodium iodide (thallium-activated) crystal as the detector.

Because the efficiency of the sampler was so high, it was apparent that its size was too large for the sampling flow rate used and that a smaller one could be used without impairment of the collection efficiency. The smaller sampler was made from a "U"-shaped piece of 3/4" I.D. copper tubing. The overall dimensions were 1-3/4" wide and 10-3/4" long. A total of 75 gms. of 12 to 30 mesh activated carbon was held in the sampler with 60 mesh brass screens and two #3 rubber stoppers. Connection to the suction line was provided by 1/2" copper tubing inserted into pre-cut holes in the rubber stoppers. A laboratory clamp on a ring stand held the sampler about 7" deep in a 665 ml. Dewar flask. The liquid nitrogen level was maintained at 1" to 3" below the top of the flask during operation.

To determine the distribution of the xenon and krypton through the small sampler, the carbon was divided into 5 equal sections. Each section was separated with a sixty-mesh brass screen. The small sampler was placed in the liquid nitrogen and connected between two large samplers at liquid nitrogen temperature. The air stream was drawn consecutively through the drying unit, the first large sampler containing radioactive xenon and krypton from RALA, the smaller sampler, the second large sampler with no activity, a flow rate meter, and the suction pump. The flow rate was adjusted to 0.5 cfm. A shielded sodium iodide crystal was directed at the inlet of the small sampler. The output from the detector was connected in parallel to the multichannel analyzer and a scaler. The scaler was set to run continuously throughout the experiment.

The first large sampler was removed from the liquid nitrogen and allowed to warm up to room temperature. A few minutes after the temperature of the air coming out of the first sampler reached zero degrees centigrade the scaler showed an increase in activity. A gamma spectrum showed that only radiokrypton was coming off. A few minutes later the radioxenon started coming over and continued for about 20 minutes. The small sampler was removed from the liquid nitrogen, and the sections were quickly and carefully poured out into 5 separate bottles. The bottles were then tightly capped to prevent the escape of xenon and krypton. Gamma spectra of the five sections were made. The first and second sections contained all of the radioxenon with no radiokrypton showing on the spectra. The second section had slightly more activity than the first section. The third and fourth sections contained only radiokrypton with the fourth section containing less than 0.1% of what was in the third section. The fifth section contained a trace of radiokrypton. A gamma spectrum of the second large sampler showed a trace of both radioxenon and radiokrypton. The concentration of radioactive xenon and krypton found in the fifth section and the second large sampler was insignificant. The collection efficiency of the small sampler was essentially 100% for both radioactive xenon and krypton.

One of the large samplers was modified for temperature studies. Copper-constantan thermocouples extending 5" through OO stoppers were inserted into the activated carbon through two 1/2" holes drilled

over the center of the vertical tubes of the sampler. The other ends of the thermocouples were kept in a water-ice equilibrium mixture. A potentiometer bridge was connected to the thermocouples to measure the emf. Figure 2 shows this modified sampler and the other equipment while a sample was being taken. The temperature of the air coming out of the first sampler was  $-150^{\circ}$  C. when the first activity reached the sampler. Seven minutes later when sampling was stopped the temperature had dropped to  $-170^{\circ}$  C. The flow rate was approximately 0.7 cfm. The first sampler measured about 500 mr/hr at contact. There was no activity in the second sampler in the series. The carbon in the outlet arm of the first sampler was carefully poured out and examined. No activity was found, indicating that when the samplers are cooled to about  $-160^{\circ}$  C. radioactive xenon and krypton are collected completely.

To determine the length of time activated carbon retains xenon and krypton at room temperature a 30" piece of 25 mm pyrex glass tubing was filled with 12 to 30 mesh activated carbon. Two sixty mesh brass screens and two rubber stoppers were used to hold the carbon in place. The air stream was drawn consecutively through the drying unit, the modified sampler containing radioactive xenon and krypton from the previous run, a continuous carbon cartridge (4) in a shielded sodium iodide (thallium-activated) well crystal, the glass tube filled with carbon, a second continuous carbon cartridge in a second shielded well crystal, a flow rate meter, and the suction pump. The multichannel analyzer and a scaler were connected in parallel to the first well crystal. A scaler was connected to the second well crystal and both scalers were set to run continuously during the experiment. Any increase in activity could be readily observed. The pump was turned on while the sampler was still immersed in the liquid nitrogen. The flow rate was maintained at approximately 0.2 cfm.

Figure 3 shows the temperature vs. time curve with time zero being the time the sampler was removed from the liquid nitrogen. The temperature continued to drop for a few minutes because the sampler had not run long enough to completely cool the carbon around the thermocouples. Curve #1 is the temperature of the inlet side and curve #2 is the outlet side.

As shown by the line marked Kr in Figure 3, the radiokrypton was detected 30 minutes later in the first carbon cartridge. The carbon cartridge is a  $5/8"$  x  $2\frac{1}{2}"$  plastic vial filled with 3 gms. of 12 to 30 mesh activated carbon. At room temperature the holdup time of radioxenon and radiokrypton on the carbon cartridge is sufficient to allow detection of the activity but is short enough not to interfere with the experiment. At the end of 32 minutes a gamma spectrum of the radiokrypton was made and is shown in Figure 4. The radiokrypton was detected in the second carbon cartridge two minutes after the isotope appeared in the first carbon cartridge. In approximately 36 minutes the radiokrypton had disappeared and both counters returned to background, represented by the small circles in Figure 4. As shown in Figure 3 the radioxenon started coming off 52 minutes from time zero. Figure 4 shows the gamma spectrum of the radioxenon at the end of 55 minutes. The radioxenon reached the second carbon cartridge in about 14 minutes. At  $18^{\circ}$  C. the 30" glass tube filled with carbon retained the radiokrypton for 2 minutes and the radioxenon for 14 minutes. A survey of the distribution of the activity along the large sampler was made and a peak identified as radioxenon was found. The peak of the xenon activity moved 7" through the carbon in about 12 minutes. The increased holdup time on the carbon in the sampler over

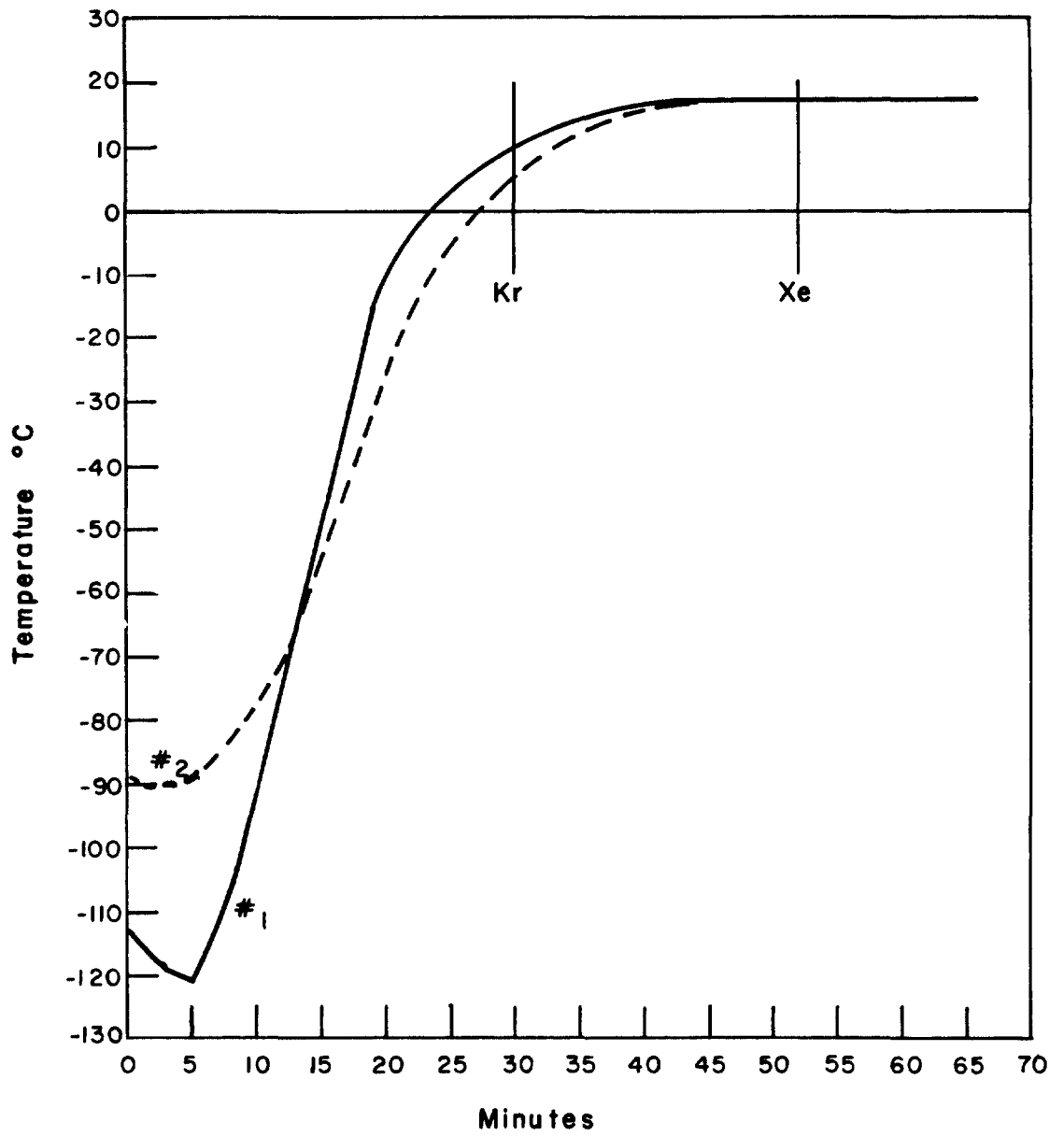


Fig. 3—Evolution of xenon and krypton.

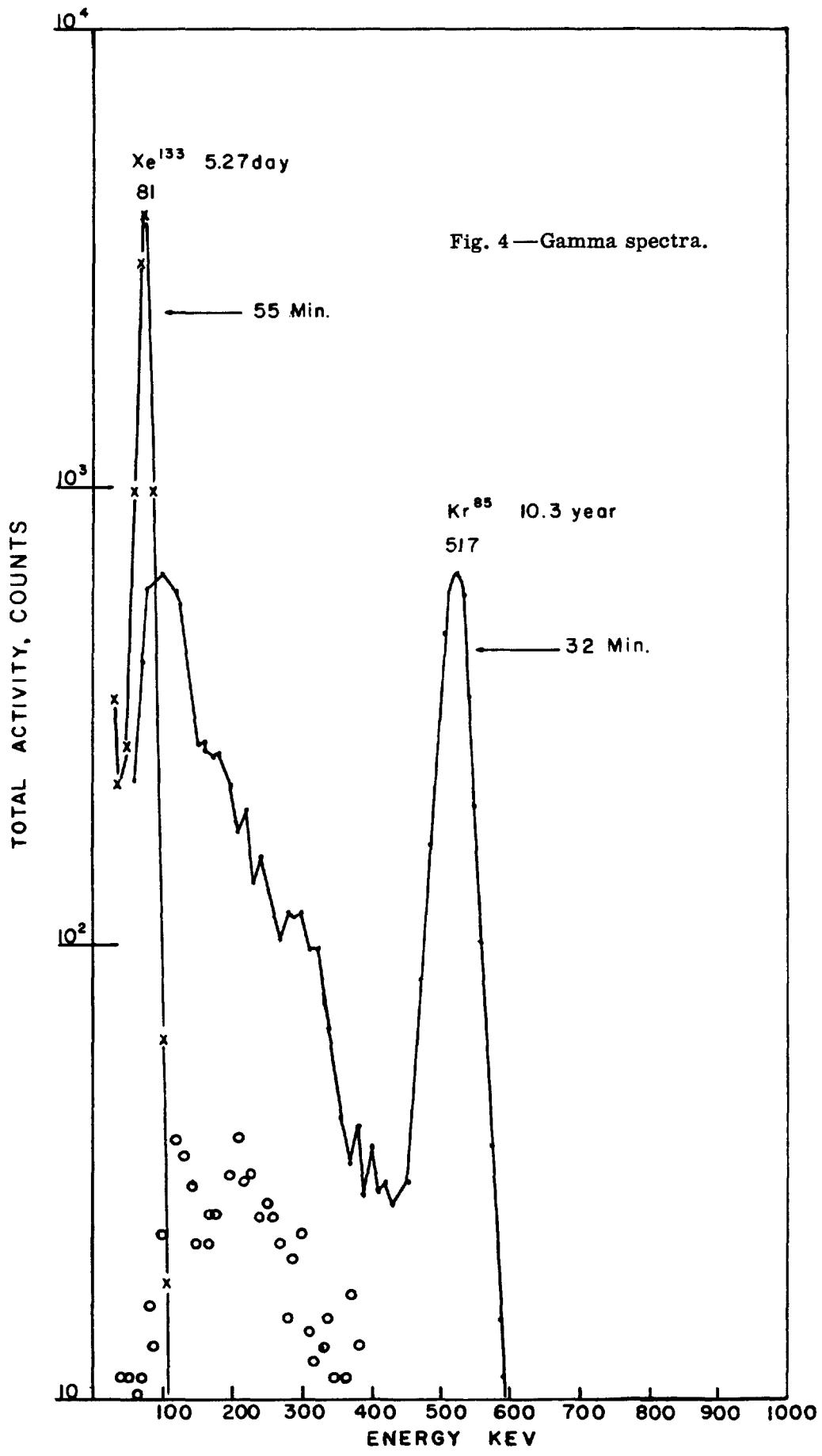


Fig. 4—Gamma spectra.



the carbon in the glass tube was probably due to the larger diameter of the sampler tube. It was evident from the position and movement of the xenon activity in the large sampler that the radioactive xenon and krypton evaporated at approximately the same time. The 22 minute delay in the appearance of radioxenon after the radiokrypton as shown in Figure 3 was caused by the longer holdup time of the radioxenon on activated carbon.

#### CONCLUSIONS

Krypton as well as xenon can be collected completely if the temperature is below  $-160^{\circ}$  C. and the surface area for heat transfer is large. The large sampler had more than enough capacity and with the proper drying unit the flow rate of air could be increased many times above 1 cfm. without any loss in efficiency. The small sampler contained sufficient activated carbon to do an adequate job of collection at 1 cfm. At room temperature the holdup time of radioxenon on activated carbon is longer than that of radiokrypton. Previously unsuspected leaks in the RALA off-gas system were found which would have been almost impossible to detect using conventional methods.

#### REFERENCES

1. Legler, B. M., Fairbourne, S. F., Kelley, P. N., and Robinson, R. A., IDO-14414, 1957.
2. "Handbook of Chemistry and Physics", Fortieth Edition, Chemical Rubber Publishing Co., Cleveland, Ohio, 1958.
3. Browning, W. E., and Bolta, C. D., ORNL-2116, 1956.
4. Sill, C. W., and Flygare, J. K., Jr., Health Physics, in press.
5. Strominger, D., Hollander, J. M., and Seaborg, G. T., Reviews of Modern Physics, 30, No. 2, 1958.

## WASTE CALCINATION OFF-GAS STUDIES

**B. R. WHEELER**

*Atomic Energy Division, Phillips Petroleum Company,  
Idaho Falls, Idaho*

### A B S T R A C T

Development studies concerned with treatment of off-gas from the waste calcination facilities currently being installed at the Idaho Chemical Processing Plant are described. Fission products contained in dust from the calciner make an essentially 100 per cent gas-solid separation mandatory.

Calcination conditions determine the size and concentration of solids in the off-gas. Data from the evaluation of venturi scrubbers on a pilot plant calciner show that all but  $1.6 \times 10^{-3}$  grains of solids per cubic foot of gas can be removed. Tests with a venturi scrubber and electrostatic precipitator independent of the cold calciner indicate that a gas containing only  $1.5 \times 10^{-6}$  grains of solids per cubic foot of gas may be attained at the expense of 10 to 15 inches Hg pressure drop across the venturi scrubber.

### INTRODUCTION:

The current construction of a demonstrational fluidized bed calciner is the Idaho Chemical Processing Plant's (ICPP) attempt to dispose of radioactive waste in a safe economical manner and evaluate the method as an ultimate means of disposal. A fluidized bed calciner is, of course, an excellent aerosol generator and presents some complex air cleaning problems. Unlike most industries which are satisfied with an off-gas cleaning efficiency of 95 to 98 per cent, the ICPP can only be satisfied with essentially 100 per cent gas-solids separations because of the radioactive nature of the solids. The ICPP is limited to the maximum permissible concentration of strontium 90, the controlling isotope, at the perimeter fence as given by NBS Handbook 52. Absolute quantities are dependent upon the prevailing atmospheric conditions and the physical dimensions and operating characteristics of the ICPP stack and are beyond the scope of this paper.

The fluidized calciner will convert the corrosive radioactive liquid aluminum nitrate, currently stored in stainless steel tanks into

a solid aluminum oxide product. Conversion to the aluminum oxide form eliminates the corrosive nature of the radioactive waste and reduces the volume nominally by eightfold. The process is very simple: liquid aluminum nitrate is sprayed into a bed of fluidized aluminum oxide which is at a temperature of 400°C - 500°C. Upon contact nitrates and water vapor are driven off leaving behind the alumina and radioactive oxides. NaK circulated through a bundle of heat exchange tubes transfers the heat from an oil fired furnace to the calciner bed. The fluidizing media is air, and of course, it entrains solids which must be separated before the air plus other waste gases are discharged to the atmosphere.

Calcination studies have shown that product size and solids concentration in the off-gas are primarily dependent upon the quantity of air used to atomize the feed in the spray nozzle. Space velocity of the fluidizing air in the calciner also affects the quantity of solids entrained in the off-gas. Control of calciner variables plus the off-gas cleansing apparatus will permit sufficient gas-solids, separations. Off-gas cleansing apparatus includes (1) a primary cyclone, (2) venturi scrubber plus knockout cyclone, (3) ruthenium adsorbers, and (4) AEC filters. Our pilot plant studies also encompass evaluation of an electrostatic precipitator. The balance of this paper will be primarily concerned with off-gas studies and equipment used only in the pilot plant calciner.

#### EQUIPMENT:

Figure 1 shows the calciner pilot units on which the variables affecting calcination are studied. Fluidizing air under pressure is introduced to a jet which in turn feeds the fluidizing gas to bubble cap air distributors in the bottom of the calciner. The fluidizing air entrains solids as it passes through the aluminum oxide bed. The solids laden off-gas enters the primary cyclone after leaving the calciner where about 95 per cent of the solids are removed. The separated solids in the cyclone plus about 30 per cent of the off-gas are returned via the jet to the bottom of the calciner along with make-up fluidizing air.

TABLE I

Calciner Sizes	Off-Gas Rates SCFM	Diameter Venturi Scrubber Throat	Type Cyclone
6" Diameter	12 - 15	5/16"	US Hoffman
2 ft. sq.	130 - 150	1"	Ter Linden

The off-gas leaving the primary cyclone is then introduced into a venturi scrubber where it is contacted by water. Scrub solution on the pilot calciners can be either fresh water or recycle solution. Conditions permitting recycle are used to simulate proposed plant operation. A cyclone downstream of the scrubber then separates the liquid-solids from the gas. In our pilot plant calciners a condenser follows the venturi cyclone to reduce the volume of water in the off-gas. Sample taps exist to permit evaluation of the off-gas equipment and determine how calcination variables are affecting the off-gas. There are two

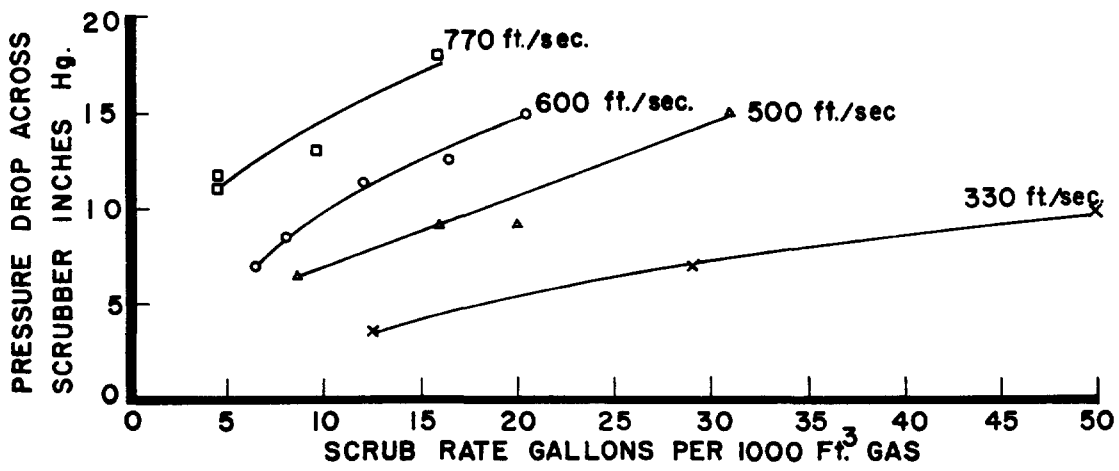
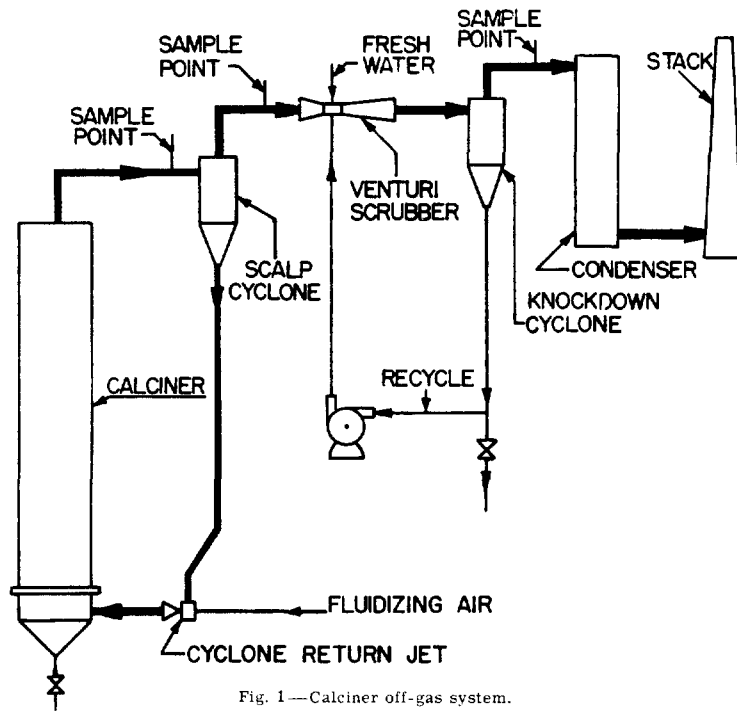


Fig. 2—Effect of scrub rate and air velocity on pressure drop across venturi scrubber.

calciner pilot units on which studies are being made: (1) a 6" diameter unit and (2) a two-foot square model. Table I tabulates the average off-gas flows as well as scrubber dimensions.

Scrubber equipment also included a short section of Yorkmesh Demister between the venturi scrubber and knock out cyclone during the early evaluation period. Occasional clogging of the Yorkmesh necessitated its later removal, however. The clogging was caused from entrained solids in air continuously bled to the spray nozzles to prevent their plugging when the calciner was down. The Yorkmesh did appear to increase scrubber efficiency somewhat.

Present equipment on the calciner includes an enlarged section downstream of the scrubber cyclone. All cyclones thus far used have permitted a film of liquid to be carried over at high gas velocities. Naturally a venturi scrubber can be no more efficient than the knock out cyclone. Re-entrainment of this film has been eliminated by using the enlarged section.

The independent venturi scrubber equipment consists of (1) bubble cap column one foot in diameter, (2) a venturi scrubber with a 1/4 inch diameter throat, (3) a US Hoffman type cyclone, and (4) a packed column downstream of the cyclone to eliminate water carry over film at high air rates.

The bubble cap column was used as the aerosol generator and was chosen to simulate the fluidized bed of the calciner. Aluminum oxide product from the calciner was pulverized in a ball mill for 10-16 hours and then put into the bubble cap column. Average particle size of this ground  $Al_2O_3$  as determined by a Fischer Sub Sieve Sizer was less than 2 microns. Microscope examination showed the material ranged from 0.01 to 3 microns in size. Some agglomeration into larger particles was apparent, however. Metered air entering the bottom of the column generated an aerosol which was diluted 60 to 90 per cent with additional air before it was forced into the venturi scrubber at throat velocities of 300 to 800 feet per second.

Water was injected into a 1/16" hole bored perpendicular to the venturi scrubber throat at rates between 5 and 55 gallons per 1000 cubic foot of gas.

An electrostatic precipitator to separate  $Al_2O_3$  particles from air is also being evaluated independently of the pilot plant calciner. The precipitator consists of 1.76" ID-12" long, wetted wall, column with a twisted stainless steel rod as a discharge electrode. A power pack capable of supplying 20,000 volts completes the precipitator.

The electrostatic precipitator has not been operated to date at voltages exceeding 10,000, however, because of a tendency to arc as the voltage builds up. Once this arcing starts, the voltage drops off. The original straight electrode was replaced by a twisted rod with 1/16" barb projections. This electrode has made it possible to increase ionizing voltages from 7-8000 to 9-10000 volts.

The wetted wall column section of the precipitator is usually operated with film of water 0.003 inches thick flowing continuously over the walls. The water film removes deposited solids thereby increasing overall efficiency. Splashing occurs when the water film is too thick causing excessive current discharge from the center electrode. Space velocities exceeding three feet per second tend to cause splashing also, which results in current discharge.

Aerosol for the precipitator is generated in a manner similar to that for the independent venturi scrubber. The solids concentration of the gas is then reduced to about  $1.5 \times 10^{-3}$  grains per cubic foot with a venturi scrubber prior to entering the electrostatic precipitator.

All gas samples are taken with a cascade impactor backed up with a millipore filter. The impactor is one designed by J. A. Brink<sup>(4)</sup> of Monsanto based on work of Ranz & Wong<sup>(5)</sup>. Ordinarily the impactor is operated such that the particles caught on the plates are the characteristic particle sizes listed in Table II. The millipore filter catches those particles escaping the final plate of the impactor. Work performed at KAPL<sup>(3)</sup> indicates the millipore filter removes more than 90 per cent of those particles exceeding 0.01 microns.

TABLE II

<u>Impactor Stage</u>	<u>Characteristic Size Microns</u>
1	2.40
2	1.72
3	1.15
4	0.57
5	0.28

---

Samples of gas taken upstream of the venturi scrubbers were isokinetic because of the possible existence of large particles. Downstream samples were not necessarily isokinetic because the particles were primarily submicron in size.

EXPERIMENTAL:

Most of the data for the venturi scrubber evaluation were taken on the scrubber which was independent of the pilot plant calciner. Lack of flexibility of the calciner off-gas system necessitated this. The calciner for the most part was operated to determine what variables affected the process rather than for venturi scrubber evaluation. Thence, the very control of primary calcination variables severely limited the scrubbing conditions for any one venturi scrubber design. The independent venturi scrubber permitted exploration of a wide range of scrubber conditions on an  $Al_2O_3$  aerosol. After optimum precipitator conditions have been established a precipitator will be designed and installed on the calciner.

Inlet loadings on the independent venturi scrubber were all considerably lower than those at a similar point on the calciner; that is, 0.7 to 1.5 grains per cubic foot as compared to 0.07 to 0.2 grains per cubic foot. This situation, of course, was undesirable but was necessitated by the gradual coating of the walls of the scrubber with  $Al_2O_3$  upstream of the water injection point at higher aerosol concentrations. Water injected into the venturi throat apparently splashes somewhat, thereby wetting the upstream walls. The dry  $Al_2O_3$  particles tend to stick to the wetted surface over a period of 8 to 10 hours, and build up the overall pressure drop. This phenomenon was also observed on the calciner venturi scrubbers when they were operated without feed injection. When feed is being sprayed into the calciner the off-gas is composed of vapor as well as nitrates and air. The presence of the vapor in the off-gas apparently prevents the solids from sticking to the venturi scrubber walls.

Gradual build up of pressure drop on the independent venturi scrubber was eliminated by keeping the solids concentration in the gas low and by inserting a 1/16" tube upstream of the venturi scrubber throat. The tube outlet was facing the throat and about 10 per cent of the scrub water was bled continuously through this tube and parallel to the venturi throat walls to wash the solids away.

#### DATA PRESENTATION:

The data are presented as a series of graphs. Data on the experimental venturi scrubber were collected under two sets of conditions: (1) with a section of Yorkmesh between the venturi scrubber and knock out cyclone to act as a coalescence surface and (2) without the Yorkmesh.

Figure 2 is a plot of the pressure drop data across the venturi scrubber and knock out cyclone without the Yorkmesh section. This figure shows that pressure drop is a function of both the aerosol velocity in the throat of the venturi scrubber and liquid scrub rate. Increasing velocities and scrub rates both increase overall pressure drop; however, the aerosol velocity contributes to the greater power loss. Data collected when the Yorkmesh was used reflect a somewhat higher pressure loss for corresponding aerosol velocities and scrub rates. The data points in Figure 2 represent pressure drop information collected during air cleaning periods. No definite attempt was made to obtain data primarily for pressure drop versus scrub rates and velocities.

Several investigators (2)(4)(6) have correlated venturi scrubber efficiency as a function of power loss. Figure 3 represents a similar correlation for our data. This study utilized higher aerosol velocities and scrub rates, resulting in much higher pressure losses across the venturi scrubber than did the previous investigations.

Penetration rather than efficiency has been plotted to convey the data more vividly. All data collected on the venturi scrubber set up is plotted in Figure 3 with no consideration given to gas velocity or scrub rate. Considerable data scatter is evident; however, there is a definite trend showing that the venturi scrubber efficiency is a function of the pressure drop or power utilized.

These data show that for the  $Al_2O_3$  aerosol specific gas velocities or scrub rates are relatively unimportant. A combination of scrub rate and gas velocity will set a corresponding pressure drop which will result in a particular scrubber efficiency. Any extrapolation, of course, can be carried too far. It is erroneous to pick a high gas velocity with essentially no scrub rate and expect the same cleaning that a low gas velocity plus high scrub rate would produce even though the corresponding pressure drops were similar.

Other workers (2)(4)(6) have achieved excellent efficiencies on venturi scrubbers at lower power requirements; however, the concentration of solids was higher and/or the particle size was larger.

Figure 4 is similar to Figure 3 in that it shows how power consumption affects venturi scrubber cleaning. Off-gas loading downstream of the venturi scrubber as a function of the power requirements again follows a straight line variation on semi-log paper. The power requirements here for the scrubber are much more pronounced; i.e., the data are not scattered as much as in Figure 3.

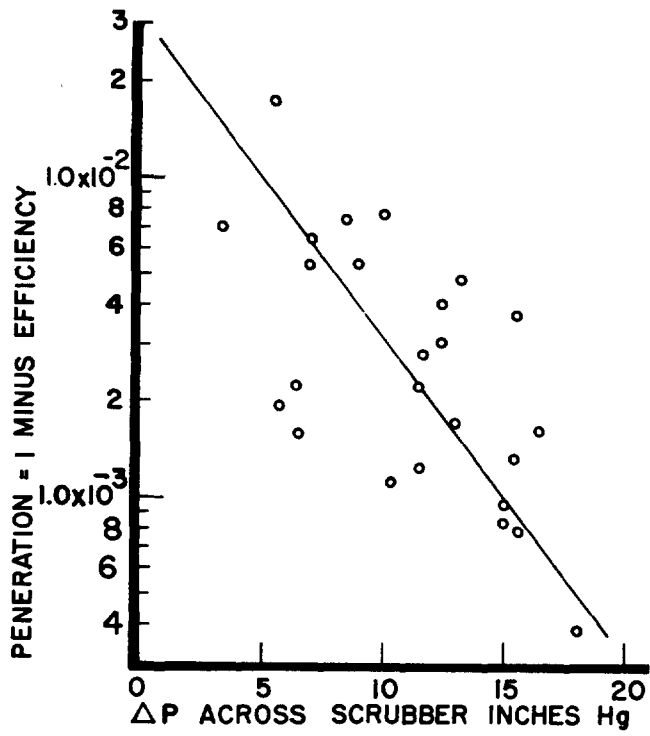


Fig. 3—Experimental scrubber efficiency for particles in the 3.0 to 0.01  $\mu$  range.

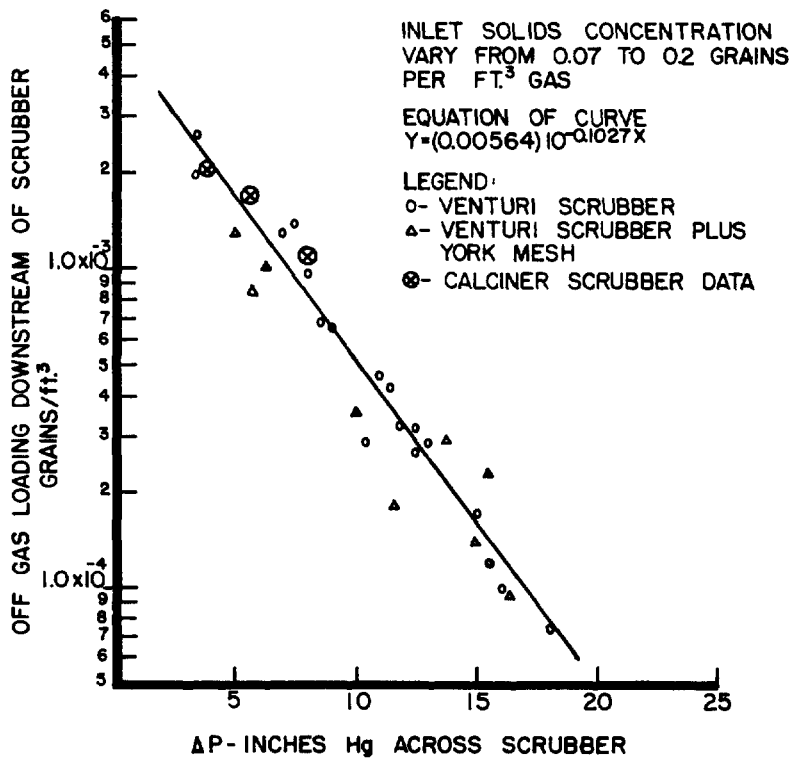


Fig. 4—Concentration of solids in gas downstream of venturi scrubber.



The empirical equation:

$$y = 0.00564(10^{-0.1027x})$$

describes the data in Figure 4.

Where:  $y$  = grains of solid per  $\text{ft}^3$  of off-gas downstream of venturi scrubber  
 $x$  = pressure drop across scrubber inches of Hg.

It may be assumed that if the upstream gas-solids concentration were the same for all data points the scatter in both graphs, Figures 3 and 4 would be identical. (The slope of the data is the same for both graphs.) Since the upstream loading varies by as much as threefold for different points, while the outlet loadings for similar pressure drops are essentially the same, it is evident that efficiencies must have more scatter. The data indicate that the importance of the upstream solids loadings on the venturi scrubber efficiency is nil in comparison to the power requirements. It is evident that the pressure drop is the most important of the variables examined in this study; i.e., scrub rate, aerosol velocity, and solids concentrations.

Figure 4 points up that extremely good gas cleaning may be performed on particles smaller than 3.0 microns if a large power loss can be expended.

Figure 5 illustrates how various aerosol velocities and scrub rate affect the off-gas loading downstream of the venturi scrubber. With the exception of the runs which had the Yorkmesh demister in the venturi scrubber it is the same data previously plotted as a function of pressure drop. The off-gas cleaning is somewhat better with the demister; i.e., the velocity parameters would be shifted downward. Slightly higher pressure drops were encountered though for similar velocities and scrub rates; hence the reason for better cleaning. This further indicates that the power requirements reflect the scrubber efficiency more than the other variables do. The Yorkmesh data were not included in Figure 5 because the parameters are shifted slightly and tend to distort the picture.

Figure 5 clearly shows that the best off-gas cleaning occurs at high aerosol velocities and high scrub rates. These conditions both give the higher pressure drop.

TABLE III

Scrubber	% Scrub Recycled on Calciner	Off-Gas Loading - Grain/ $\text{ft}^3$	
		Calciner Scrubber	Independent Scrubber (% Recycle)
	100	$6.0 \times 10^{-3}$	$7.0 \times 10^{-5}$
	92	$3.0 \times 10^{-3}$	$1.0 \times 10^{-4}$
	70	$6.0 \times 10^{-4}$	$1.0 \times 10^{-4}$
	0	$1.6 \times 10^{-3}$	$1.3 \times 10^{-3}$

NOTE: Like pressure drop data are being compared

The major portion of the data collected downstream of the venturi scrubber on the calciner off-gas system was collected at a time when the scrub solution was being recycled. As might be expected the recycle of the scrub solution reduces scrubber cleaning ability by a considerable amount. Table III gives some comparative data collected for like

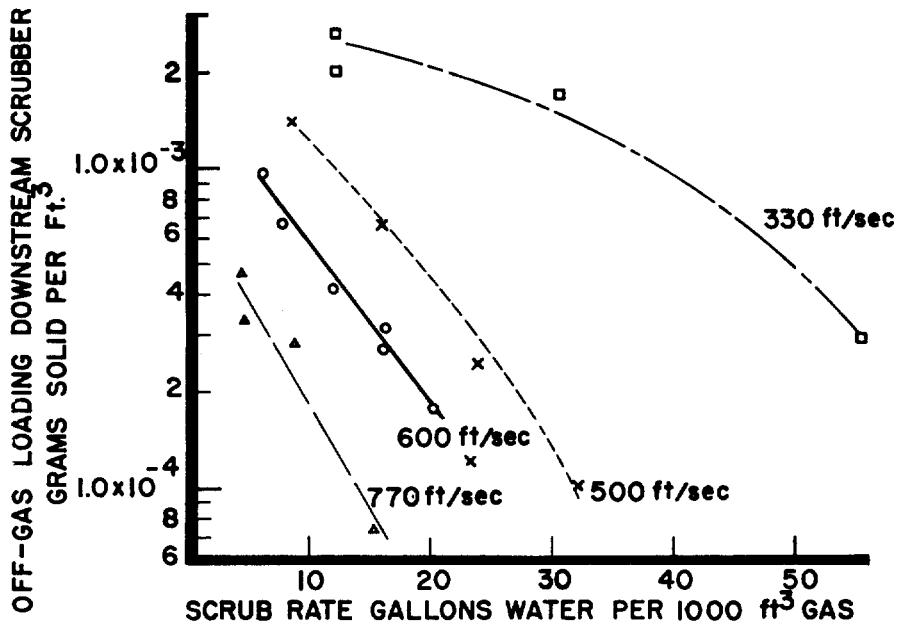


Fig. 5—Concentration of solids in gas downstream of experimental venturi scrubber.

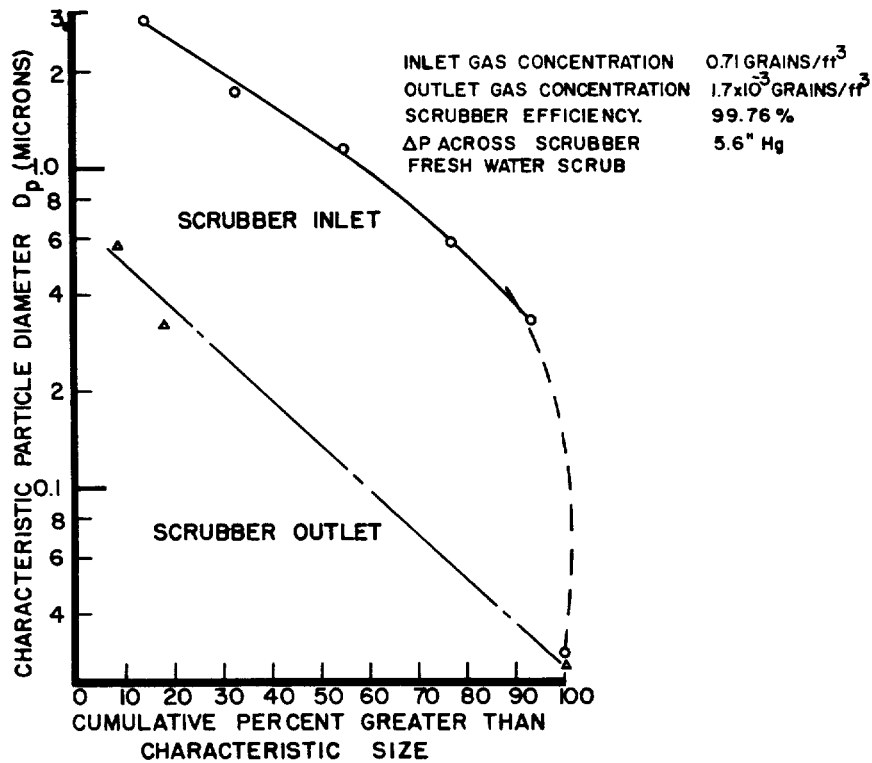


Fig. 6—Particle size distribution of off-gas entering and leaving the calciner venturi scrubber.

pressure drops across the calciner scrubber and independent scrubber. It is important to note that even at 100 per cent scrub recycle that portion of the liquid condensed in the calciner venturi scrubber was continuously bled off to insure equilibrium of (1) liquid volume and (2) solids concentration in the liquid. Solids concentration in liquid was dependent on the per cent of the scrub recycled as well as calcination conditions. Insufficient data prevent a comparison of liquid solids concentration instead of the scrubber recycle comparison in the table.

The table shows that venturi scrubber data for pilot plant and independent scrubbers are nearly the same when fresh water is used as scrub solution. As the per cent of recycle is increased the difference between the scrubbers becomes larger. At 100 per cent recycle the scrubber difference in downstream off-gas loading varies one hundredfold. (A portion of this may be due to the higher upstream gas-solids loading, tenfold higher. The zero per cent recycle data disqualifies this, however.)

Figure 6 represents the particle size distribution before and after the venturi scrubber on the two-foot square pilot calciner. For the particular day these data were taken, scrubber efficiency was 99.76 per cent with fresh water scrub. Naturally the scrubber was most efficient (100%) on particles greater than one micron. The scrubber efficiency decreased (97.2%) as the particle size decreased to those retained on the millipore filter.

Figures 7 and 8 pertain to data collected with the electrostatic precipitator. Off-gas loadings to the precipitator were in the range of  $1.5 \times 10^{-3}$  grains of solid per cubic foot of gas. Figure 7 shows that the wetted wall electrostatic precipitator efficiency drops from 90 per cent to 52 per cent for space velocities varying from 1 to  $3\frac{1}{2}$  feet per second. The data were for particulate matter smaller than 1.17 microns average size and 8000 volts. Several points on Figure 7 also indicate that higher voltages give better efficiencies.

At the first glance it appears that the venturi scrubber is a better off-gas cleaning device than is the precipitator. The difference in inlet loadings account for the wide differences in efficiencies. The loading to the calciner scrubber for data in Figure 6 exceeded the loading to the electrostatic precipitator more than two hundred times.

Surprisingly, however, the venturi scrubber is capable of giving almost the same type of gas cleaning as that from the precipitator,  $1.5 \times 10^{-6}$  grains of solid per cubic foot of gas as compared to  $1.5 \times 10^{-5}$  grains per cubic foot. Both numbers represent off-gas loadings downstream of the apparatus. The major difference, however, is that the pressure drop across the precipitator is about one-inch of water while that across the scrubber is about two hundred fifty inches of water.

The effect of voltage on efficiency and particle size is shown in Figure 8. Higher voltages and particles above 2 micron produce 100 per cent efficiency for this study. Once again off-gas loading is very small for Figure 8 data ( $1.5 \times 10^{-3}$  grains per cubic foot.)

#### CONCLUSIONS:

(1) The major contributing factor to a venturi scrubber efficiency for particles smaller than 3.0 microns is amount of pressure drop taken.

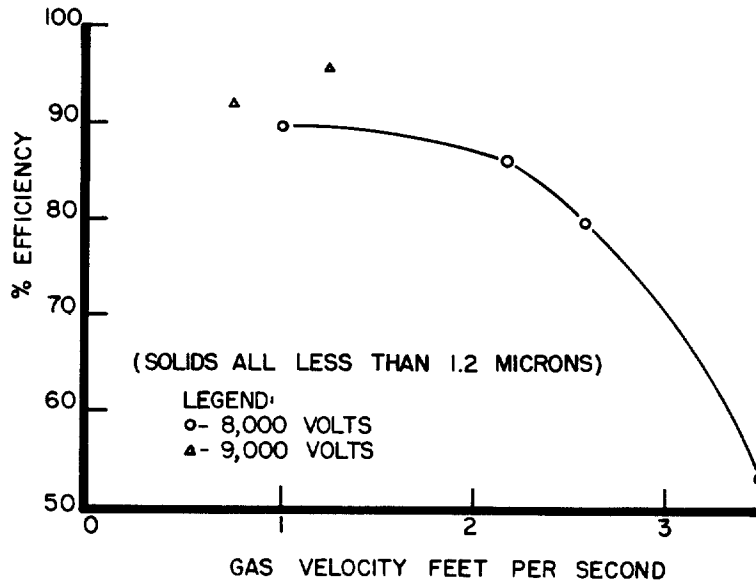


Fig. 7—Effect of air velocity on electrostatic precipitator efficiency.

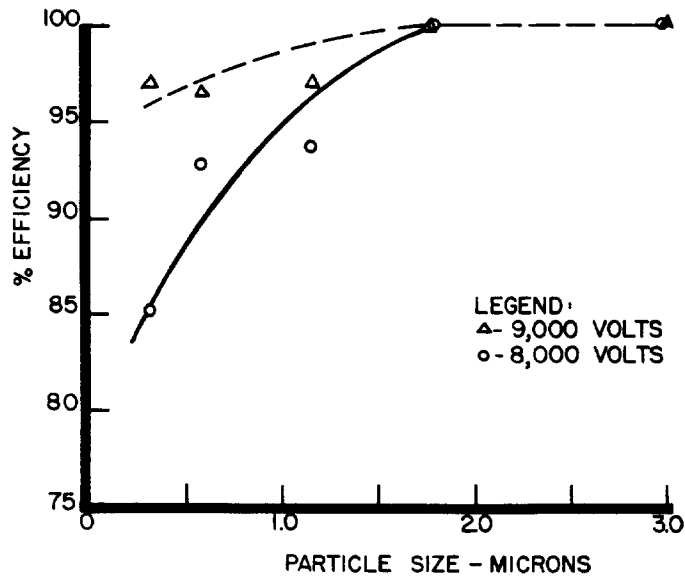


Fig. 8—Voltage effect on electrostatic precipitator efficiency.

Once the decision is made of what concentration of solids in off-gas can be tolerated data in this paper will indicate the power requirements in the form of pressure drop. For the area studied it matters very little whether high gas velocity plus low scrub rates or low gas velocity plus high scrub rates are used in the venturi scrubber.

(2) Variation in upstream solids concentration will not contribute to the gas cleaning ability of a venturi scrubber in the range 0.07 to 0.2 grains per cubic foot of gas.

(3) Electrostatic precipitator efficiency is best at high voltages and low gas space velocities. Higher voltages may be obtained with a barbed center electrode.

(4) The venturi scrubbers clean-up ability can approach that of the electrostatic precipitator. A pressure drop of two hundred fifty inches of water across the scrubber is expended, however, in comparison to one-inch of water across the electrostatic precipitator.

#### LITERATURE CITED

- (1) BRINK, J. A. Jr. - Paper presented at Boston Annual AIChE Meeting, December 1956
- (2) EKMAN, Frank O., and Johnstone, H. F. - Industrial and Engineering Chemistry, 43, 1358 (1951)
- (3) FITZGERALD, J. J. and Detweiler, C. G. - KAPL Report, 1592 (1952)
- (4) LAPPLE, C. E., and Kamack, H. J. - Chemical Engineering Progress, 51, 110 (1955)
- (5) RANZ, W. E., and Wong, J. B. - Archives of Industrial Hygiene and Occupational Medicine, 5, 464 (1952)
- (6) SEMRAU, K. T., Marynowski, C. W., Lunde, K. E., and Lapple, C. E. Industrial and Engineering Chemistry, 50, 1615 (1958)

#### Discussion

- Q. I wonder why he didn't use higher voltage than 9,000 as this is pretty low for control type.
- A. Reason is that we are using a little model. Not able to get high voltage without arcing or shorting out.

## THE SIGNIFICANCE OF THE AIR CLEANING PROBLEM IN AIRCRAFT NUCLEAR PROPULSION

ROBERT E. BAKER

*Aircraft Nuclear Propulsion Department,  
General Electric Company, Cincinnati, Ohio*

Most of you are familiar with the effort being made by General Electric to produce a direct cycle nuclear power plant for aircraft propulsion. Figure 1 is a picture of the Heat Transfer Reactor Experiment (HTRE #1) as it is being moved by a shielded locomotive in the Idaho Test Station. The reactor and shield assembly is carried in the center of the Core Test Facility (CTF) which is literally a laboratory on wheels. This

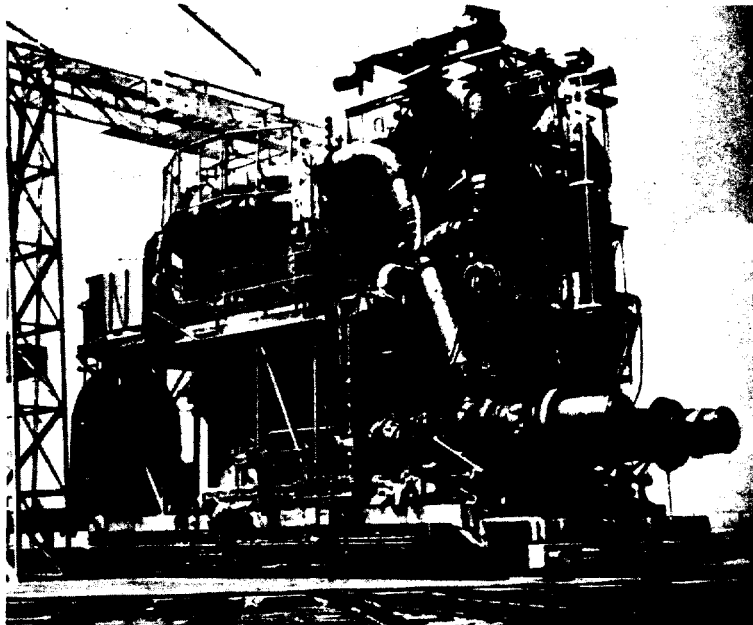


Fig. 1 — Heat transfer reactor experiment (HTRE) power plant.

apparatus was the first to demonstrate that jet engines could indeed be operated by nuclear power. The apparatus obviously was not intended to fly.

The mode of operation of the HTRE is shown schematically in Figure 2. Air is taken in through the compressors and routed through ducting and a torus to the reactor. After passing through the reactor, air again passes through ducting and a chemical burner can before passing out through the turbine and being exhausted to the atmosphere through a 150 foot stack.

Routine operation of the direct cycle reactors releases a negligible amount of radioactivity. This activity is carried out of the stack and dispersed without a problem. However, aircraft reactors require high power densities in order to minimize shielding requirements and therefore have rather stringent coolant requirements. Loss of coolant during oper-

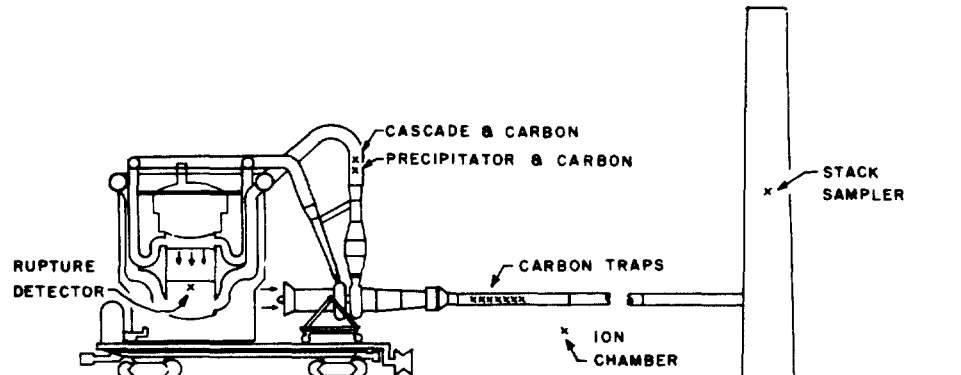


Fig. 2—In-facility sampling locations.

ation at power or shortly after shutdown can cause considerable damage to the fuel elements and release significant amounts of activity in the form of fission products. Testing fuel elements in early development stages might also reasonably be expected to release activity. While it is not practical to clean the exhaust air of a flying jet bomber, the problem of cleaning the effluent from a stationary power plant during testing might be possible.

In order to get some idea of the degree of difficulty of air cleaning problems involved. Let us look at some typical numbers which might be expected when we are dealing with the testing of an operational power package. Consider the requirements for a conventional B-36 bomber. This plane is powered by six piston engines of about 3500 horsepower each and four J-47 turbojets of about 5000# thrust each. Each plane then requires somewhere around 25 MW. of power and if we consider that all power is to be supplied by a reactor which is relatively inefficient (and to generate nice round figures) a 100 MW. (thermal) reactor is reasonable. Typically, a J-47 operates on an air flow of about 50 pounds per second and exhausts air at about 900°F. If the six piston engines of the B-36 were replaced by J-47 jet engines, the total effluent would then be expected to be about 500 pounds of air per second at about 900°F which is a lot of air to clean!

Before one can determine specifications for a cleaning system, it is necessary to determine the physical characteristics of the contaminant—in this case fission products. Laboratory scale experiments have been performed to learn what fractional release can be expected in the event of overtemperatures to fuel elements. These studies will continue. However, it is difficult to mock-up a reactor malfunction in a laboratory, therefore a reactor was modified to provide in-situ experiments on fuel

element damage studies. Figure 3 is a schematic of such an experimental arrangement. A valve has been installed to limit the coolant air to a fuel element of a reactor while the reactor is operating at power. One such experiment has been done at the Idaho Test Station to study fractional release and reactor damage. Figures 4 through 6 are pictures of some of the typical residue resulting from this experiment. From such tests

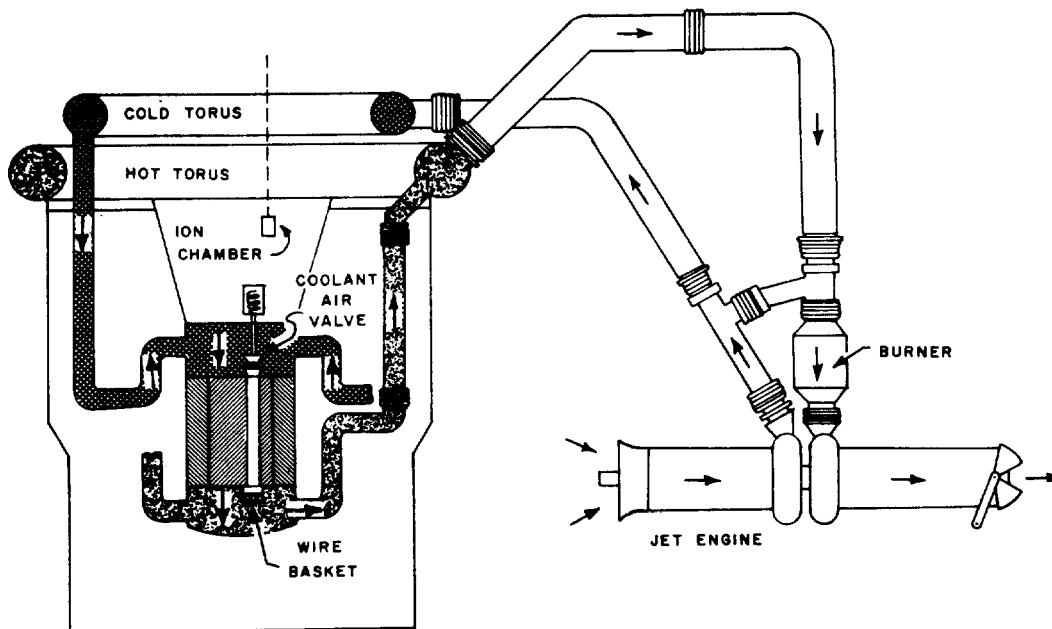


Fig. 3—Schematic of HTRE with insert.

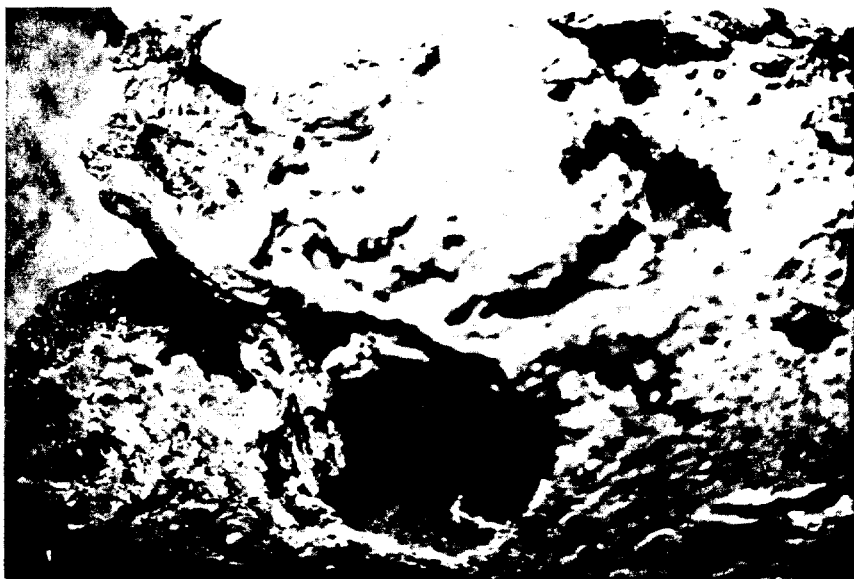


Fig. 4—Fuel element residue.

we hope to learn mechanisms of reactor damage, the fraction of specific isotopes released, physical characteristics of the contaminants, and prediction of dispersion patterns for fission products once it is released.



Meteorological control, which has been used to date, has proven to be a workable method but it does impose some restrictions on operating time. The following papers will provide more detailed information on both meteorological control of operations in lieu of air cleaning and operations in which air cleaning has been used successfully.

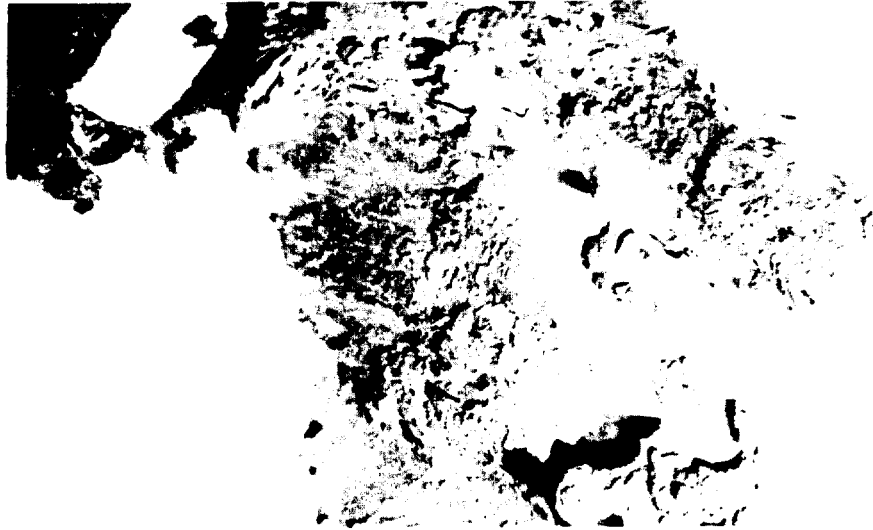


Fig. 5—Fuel element residue.



Fig. 6—Fuel element residue.

## Discussion

(C.) Reactor heat assembly - the heat transfer experiment, is it intended to fly? - routine operations produce only negative amounts of radioactivity. The radioactivity is carried out of the stack and therefore, they have very strict collecting requirements.

## EXPERIENCE WITH NON-FILTERED AIR SYSTEMS AT THE IDAHO TEST STATION

R. B. O'BRIEN

*Aircraft Nuclear Propulsion Department,  
General Electric Company, Lockland, Ohio*

### Abstract

Experimental direct cycle aircraft nuclear power plants are ground tested at the ANP Area, National Reactor Testing Station, without air cleaning. Effluent hazards are minimized by a system of meteorological control. To date, this system has proved to be successful in controlling the doses to humans and may prove to be adequate for full scale aircraft powerplant tests. Even if air cleaning of the effluent proves to be necessary in the future, a meteorological control program will serve to reduce the requirements of the air cleaning system, thereby reducing its cost.

\* \* \* \* \*

The ground testing of experimental aircraft nuclear power plants of the direct cycle type presents a significant challenge in the field of air pollution control. Since the air which cools the reactor makes a single pass through the power plant system and is exhausted to atmosphere, any radioactive materials released within the reactor core can be carried by the winds to some point where human beings might be exposed.

Radioactive materials may be present in this coolant stream for several reasons. First, some experimental fuel elements are not designed for complete retention of the radioactive fission products. Secondly, some tests may be deliberately operated at conditions designed to induce failures in fuel cladding. Third, minor cladding defects may appear even in non-destructive testing. Fourth, activation products may be released. And, of course, there is always the possibility of an accident of some kind.

Ordinarily one would immediately suggest that an air cleaning system be installed to remove all or part of the offending materials. Unfortunately this approach is a difficult and expensive one due to the extremely large volumes of high temperature air pumped by the turbo-jet engines and the fact that high back pressures cannot be tolerated.

Various air cleaning systems are under consideration, but some of the more conventional systems have already been proved impractical.

Since the beginning of the ANP testing program at the National Reactor Testing Station, in early 1956, these power plants have been operated without any attempt at air cleaning. To minimize the effects of the radioactive materials released to the atmosphere, a system of meteorological control is utilized. This control system has been under continuous development and will, of course, continue to be developed in order to keep pace with the increasing demands of the ground test program. To date, the meteorological control system has been highly successful in limiting the dose to humans to very low levels, but, in the early stages especially, the extremely conservative approach to the problem has not always permitted the most expeditious progress of the test program.

In its present form, the ANP meteorological control program is operated as follows:

The possible releases of radioactive materials are divided into two cases; the Accident Case and the Continuous Release Case. This is done because the permissible limits as well as the dose calculations are different for the two cases.

In any nuclear system there are a number of different types of accidents which could occur, even though elaborate precautions are taken to prevent them. For each different ANP reactor, a hazards report is prepared for evaluation and approval by the AEC. These hazards reports contain discussions of the causes and effects of the various possible accidents. For a given reactor, then, controlling accident is selected for each phase of the planned test program by weighing the assumed probability of occurrence and the magnitude of the hazard which would be created. This is done by evaluating such factors as; fission product inventory, degree of reliability of various components of the system, probable effect upon the system should a given component fail and the state of knowledge concerning the operating characteristics of the power plant.

Each time the reactor is operated, a series of calculations are made of the significant doses to the various critical organs at occupied areas downwind should this controlling accident occur. If the controlling dose, which is dependent on the atmospheric diffusion conditions, were calculated to be higher than accepted permissible limits, we would not operate the reactor until the meteorological conditions improve to the point that the accident dose is again within acceptable bounds.

During the periods in which radioactivity is released continuously, the applicable critical organ doses to a given receptor area, per hour of reactor operation, are calculated based upon current atmospheric diffusion conditions and measured activity release rates. The meteorological portions of these calculations are then verified by means of measurement in the field. The accumulation of dose at a given location may then be controlled by selecting desirable meteorological conditions and by controlling reactor operating time.

It is obvious that the calculation of dose at some point remote from the test facility is an essential part of this control system. These calculations are made using a modified form of Sutton's diffusion equation. This modified equation may be thought of as containing three terms: one describing the effluent materials at the source; one describing the diffusion, transport, and deposition of these materials in the environment; and the third describing the biological factors which enter into the conversion of a concentration of material in a given media to a critical organ dose in the human body.

Indications are that these calculations can be made with reasonable accuracy provided the correct input data is used.

The Source Term. The source term must, of course, be derived differently for the two different cases. For the accident this is done by computing the current fission product inventory in the reactor for the particular isotopes of interest and then multiplying by a release fraction which varies from isotope to isotope and is also dependent upon the type of accident under consideration. Some experimental evidence is available on the release fractions for a limited number of accident cases, but this type of information is by no means complete.

In the case of the continuous release of fission products, more complete data is available because the materials can be monitored on their way through the ducting and stack system before they escape to the atmosphere. Since all sampling and monitoring devices for airborne fission products have inherent limitations, several different types of devices having different characteristics are used so that there are as few unknowns as possible.

A rupture detector, which utilizes a wet cyclone to remove a fraction of the particulate from the air, is closely coupled to the hot ducting of the power plant. The liquid from this system is passed over a shielded scintillation counter. This system, while not quantitative, has a response time in the order of a few seconds and serves to warn of sudden increases in the discharge activity. This device will actuate the reactor scram system in the event of a sudden increase above pre-set limits.

Two external radiation monitoring devices are located adjacent to the discharge ducting at some distance from the power plant. One of these is a gamma sensitive ionization chamber and the other is a fission chamber which measures delayed neutrons.

As the air passes up the exhaust stack, a sample is continuously drawn from the 80' level to a stack monitoring device which is located in a shielded vault underground. Here it is passed through a continuously moving filter tape which is scanned by two beta-sensitive scintillation counters - one immediately beneath the collection point and one at a point corresponding to a 30 minute decay time. After passing through the filter, the air then goes into a shielded tank which is scanned by a gamma ray spectrometer. This gas chamber has provisions for holding up a portion of the gas for decay counting.

In addition to these continuous monitoring devices, samples are collected at many different locations for later analysis in the laboratory. Extensive use is made of deep bed charcoal traps for the collection of radioactive iodine. Other sampling devices utilize aerosol filters, electrostatic precipitators and cascade impactors. These devices do not usually provide data which is immediately useful in the control of the current test run, but they are very useful for calibra-

ting the various monitoring devices and they provide more detailed data on the characteristics and quantities of the various effluent materials.

The Meteorological Term. At present, the most useful meteorological data is obtained from a single 200 foot weather tower at the release point. A direct line to the U. S. Weather Bureau office at Central Facilities provides a means of effectively utilizing the services of this organization.

Four additional weather towers are currently being installed at radial distances of between 3.5 and five miles from the release point, thus forming a rough "box" around the ANP Area with the exhaust stack at the center. These stations will be equipped with radio-telemetering devices so that the resultant wind and temperature profile data can be transmitted to the control center on a continuous basis. The data from these outlying stations will be used to plot air flow stream lines across the ANP Area for a more accurate determination of plume trajectory, and will provide a more complete analysis of the micro-meteorology of the test site and vicinity.

The important variables in the meteorological portion of the dose calculation are the diffusion coefficient "c", the stability parameter "n", and the effective stack height. At present, these are all determined by generalized methods. The effective stack height is estimated by using the Davidson Bryant equation. The stability parameter is defined by the temperature profile, and the diffusion coefficient is determined from a general method suggested by Barad and Hilst. These generalized methods do not provide the desired degree of accuracy for this type of program. Studies are being conducted to provide new or improved methods and meteorological instrumentation.

A group of 26 radiation monitoring stations are operated in a circle roughly ten miles in diameter around the area. These stations are also being equipped with radio-telemetering devices so that the airborne activity as well as the external gamma dose from the cloud are continuously printed out at the control point. These stations - supplemented by similarly equipped mobile units - serve as calibration points for the dose calculations. Another ring of 16 continuously recording gamma sensitive ion chambers is operated at a distance of about one mile from the stack to assist in the initial trajectory determination.

In addition to these automated monitoring stations a number of manually operated air sampling and fallout stations are operated on grids at various distances from the stack to obtain more detailed data, and a comprehensive vegetation sampling and analysis program is also conducted. Supplementary monitoring and data collection programs are also operated by the AEC-IDO Health and Safety Branch.

The results of the meteorological control program to date may be summarized by stating that, to our knowledge, off site doses resulting from ANP testing have been less than 1% of the annual non-occupational limits each year. Actually, the full effectiveness of the system has not been fully demonstrated, since the actual fission product release rates have not been high enough to approach the permissible dose limits in any occupied area. In fact, the nuclear weapons tests conducted in Nevada and elsewhere in the world have added more radioactivity to the environment of the ANP site than have the ANP tests themselves.

Beyond a certain point the actual reduction of hazards by meteorological control can only be accomplished by delaying the test program. Another important benefit of this type of program, however, is that it allows the recognition and use of the permissible limits which have been established for non-occupational exposure. Without this type of program there is no means of determining the extent of the hazard created by the release of airborne radioactive materials until after the damage is done. There are still important gains to be made by refining the present system. For example, the results of field verifications, to date, indicate that for typical daytime operating conditions the atmospheric diffusion calculations are conservative by a factor of 3 on the average. If, by learning more about atmospheric diffusion, this over-conservatism can be reduced a net gain in operating flexibility will be realized.

Extrapolations have been made to determine whether a meteorological control program will be adequate to cope with the releases expected from the full scale flight-type power plants to be ground tested in the future. This, of course, depends to a great extent upon the amount of time available in which to perform each test, but indications are that it will be a marginal situation. In other words, the operation of these future power plants, within the non-occupational dose limits may be possible without filtration of the exhaust air. Even if air cleaning is found to be necessary this type of control will certainly reduce the requirements of the cleaning system thus saving the taxpayers considerable expense. For this reason, the methods and techniques of meteorological control will continue to be developed, even as filtration systems for ANP ground testing are developed on a parallel path.

## AIR CLEANING IN ANP EXPERIMENTS AT THE ETR AND MTR

D. C. FOSTER

*Aircraft Nuclear Propulsion Department, General Electric Company,  
Idaho Test Station, Idaho Falls, Idaho*

The accomplishment of ANP test objectives at the Idaho Test Station often results in an expected release of radioactive effluent. These releases can occur for several reasons:

1. Testing of fuel element types not designed for complete retention of fission products
2. Destruction testing to determine feasible operating parameter limits and life expectancy
3. Destruction testing to study kind of failure and effect.

It should be pointed out that the degree of effluent contamination we have experienced to date would not constitute a serious problem as a flying source. As a continuous source of radioactive effluent being released from a testing facility stack such as those at Materials Test Reactor or Engineering Test Reactor, the problem is somewhat more acute, and during adverse meteorological conditions, the conducting of such tests would have to be suspended if the effluent could not be at least partly decontaminated.

The effluents from tests conducted at ETR or MTR consist of hot air with a variable moisture content, some activation materials, and fission products. Of these, the latter is the most significant as a problem. Some of the fission product activity is associated with particulate matter in the air stream and some - notably the iodines, bromines, xenons, and kryptons - are present in a gaseous state.

In reducing the stack release of radioactive material, it is desirable to reduce the gross activity released as much as possible and to remove in particular those constituents which are most harmful from a biological point of view.

To accomplish this reduction in stack release of radioactive material under the conditions that exist at the MTR (and which will exist at the ETR), the effluent undergoes three phases of treatment before being ducted to the stack for release:

1. Removal of particulate material

2. Treatment to reduce the iodine content
3. Reduction of the inert component and their daughters by decay and subsequent filtering.

The accompanying illustration (Figure 1) is a schematic diagram of a prototype filter system designed and built at the Idaho Test Station to test the effectiveness of a system designed to apply this three-phase treatment to effluents from ANP tests at MTR.

This particular test filter system was designed and sized to handle the coolant flow from a series of tests at the MTR which were expected to provide a suitable source of fission product contaminated effluent. Two of this series of tests were successfully completed earlier this year providing a total of approximately 400 hours of contaminated effluent.

The schematic diagram shows a test sample in a flux region in the MTR as the fission product source. Filtered coolant air flows through the source, through the exhaust ducting (shielded), through the filter system components in series, and then is released, via the MTR stack, to the atmosphere.

The filter system, as indicated, consists of three major components - two filters and a tank system.

The two filters are identical and each is composed of three elements or beds which are six inches in depth and twelve inches in diameter. The first element is silver plated fiber frax. Support and protection is provided by a Regimesh screen on the upstream face.

The second element is an absolute filter rated by the manufacturers as being capable of removing 99.95% of a test aerosol having an average particle size of .3 microns in diameter.

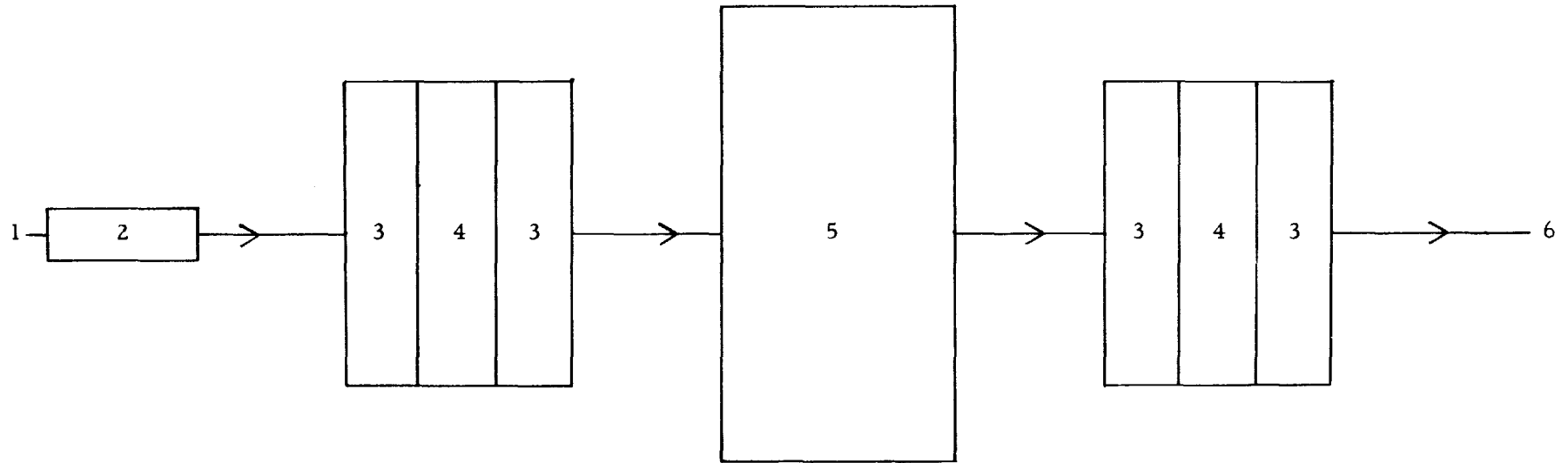
The third element is another six inch deep silver plated fiber frax bed similar to the first and having a Regimesh screen on the downstream face. Special high temperature glass fiber gaskets are employed between the flanges of adjacent elements and the three are bolted together to form a single unit when installed.

The first filter is designed to accomplish two of the treatment phases:

1. Removal of particulate material and the associated activity
2. Reduction of the radioactive iodine content by chemically combining it with the silver on the filter fibers

Quite a few of the fission products that are objectionable biologically have inert Xe or Kr precursors having short half lives. The second of the three-filter system components was so designed that the transit time of the effluent was about two minutes at the design flow conditions. This time is sufficient to permit the decay of many of the inert precursors to their more filterable Cs or Ba daughters which either plate out in the system or are removed by the second filter. In the test set-up, the two-minute transit time was provided by incorporating two appropriately-sized tanks in series with the filters. The tanks were constructed of carbon steel, baffled to insure a non-channeling flow, and designed to withstand a working pressure of 150 psig.





1. Air supply (filtered)
2. Fission product source
3. Silver plated fiber frax bed
4. Absolute filter bed
5. Delay tanks
6. To stack

Fig. 1 — Schematic diagram of a prototype filter system.

The second filter, downstream of the delay tanks, is identical to the first filter. It has silver-plated beds also for two reasons: first, to take advantage of the series setup for removing that iodine which is in the gaseous form due to being formed as a fission product directly or decaying to iodine during the two minutes in transit, and second, to react with that component which may be held up as Te or Sb in the first filter, decay to iodine, and proceed through the system.

At design flow the pressure drop across the entire filter system (including a heat exchanger not shown in the schematic) is 4.9 psig. The pressure drop across each filter is 0.6 psig and across the delay tanks is 0.2 psig.

The two-minute transit time accomplishes one other desirable result. It reduces the total radioactivity of the effluent, disregarding any filtering action it enhances, by taking advantage of the fact that many of the major contributors to the activity of fresh fission products have extremely short half lives.

By comparing the gross effluent activity upstream and downstream of the filter system - and by determining the concentration of various fission product isotopes in samples of the effluent collected upstream and downstream of each component - it was determined that this design did accomplish the desired result. Stack releases of fission products were held well below acceptable limits enabling continued testing during weather conditions which normally would have suspended testing of this nature.

The data from isotopic analysis of samples of the effluent indicated a reduction in concentration of iodine (and iodine precursors) in the effluent by a factor of 10. Inlet concentrations to the system were quite low and there was some indication that had the concentration been higher the decontamination factor would have been greater. Laboratory tests using the same silver-plated filters with greater inlet concentrations of iodine (using  $I^{131}$  tracer) have shown a reduction by a factor of 100 across the filter.

The concentration of Xe and Kr daughters was reduced by a factor of 200 - 400 across the system. Most of this loss was across the combination of delay tank and secondary filter as was anticipated. The specific isotopes for which analyses were made were  $Ba^{139}$ ,  $Ba^{140}$ ,  $Sr^{91}$ , and  $Sr^{92}$ .

The gross activity of the effluent was monitored upstream and downstream of the system by shielded ion chambers adjacent to the effluent line. Data collected from these chambers indicated a reduction in total activity by a factor of 500 to 1000.

## PROPOSED SIDE LOOP PROGRAM AT THE IET

C. L. STORRS and D. C. FOSTER

*Aircraft Nuclear Propulsion Department, General Electric Company,  
Idaho Test Station, Idaho Falls, Idaho*

The two previous speakers have mentioned some of the air contamination problems associated with the Aircraft Nuclear Propulsion test program, and have described two methods which have been used successfully to cope with these problems. I should like to describe a third approach, now reaching the pilot plant stage.

Dependence upon meteorological control, in the manner described by Mr. O'Brien, becomes increasingly less attractive when reactors of higher power are to be tested, when test schedules become tighter, and when several test facilities may be operating simultaneously. We are looking ahead to a time when some form of air cleaning may be needed for the power plant effluent.

The methods described by Mr. Foster which are being successfully used for the MTR and ETR testing are inapplicable to cleaning the effluent from a nuclear aircraft engine because of the large pressure drop they require. What is needed is a filter capable of handling a large volume of air, on the order of 1000 pounds/second, with a pressure drop of no more than 6-12 inches of water. Since the air is initially hot, it is economically advantageous to avoid the expense of cooling it. Our engineering studies indicate that an electrostatic precipitator best meets these specifications. This device will, of course, remove only particulate contaminants from the air stream. Our data show, however, that radioactive gases will, under certain circumstances, adhere to solid particles in the air stream and will be removed with them. We propose, then, to add a material such as smoke after the reactor, and to remove it in the precipitator, along with at least some of the radioactive gases.

Before proceeding to a full scale filter, which would be an installation of considerable size and expense, we have thought it expedient to gain operating experience and to collect more data by constructing a pilot plant. Accordingly, we have designed and are now assembling the side filter loop at the Initial Engine Test Facility. This loop draws off a fraction, 5 to 10%, of the IET exhaust and passes it through an electrostatic precipitator. The precipitator is a commercially available, parallel plate, horizontal flow model. It is capable of handling 14,000 cfm of air at temperatures up to 900°F. It has two sections in series to permit studying the effect of changing the length of precipitator being used. It has provisions for rapping the plates and for a decontaminating wash down, and has a continuously flushed hopper to remove the contaminated material.

Fig. 1 shows the general layout, looking downstream from the IET test building. The IET stack and exhaust duct are on the right in the picture, and the precipitator on the left. A 20 inch diameter duct, not yet installed, will connect the precipitator to the main exhaust duct just downstream from the test building.

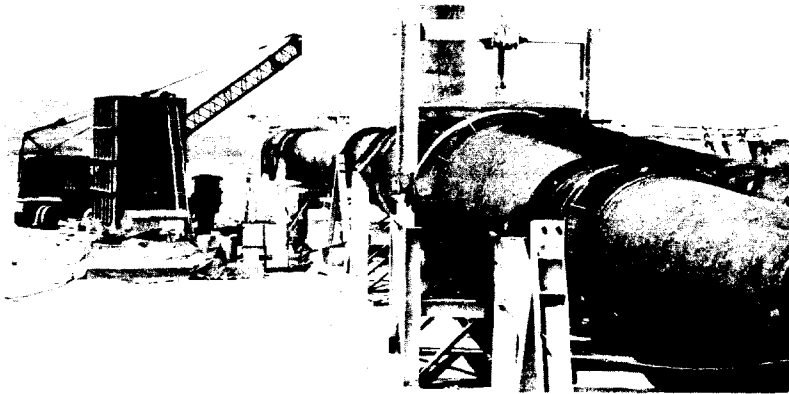


Fig. 1 — Side filter loop looking north from the Test Building.

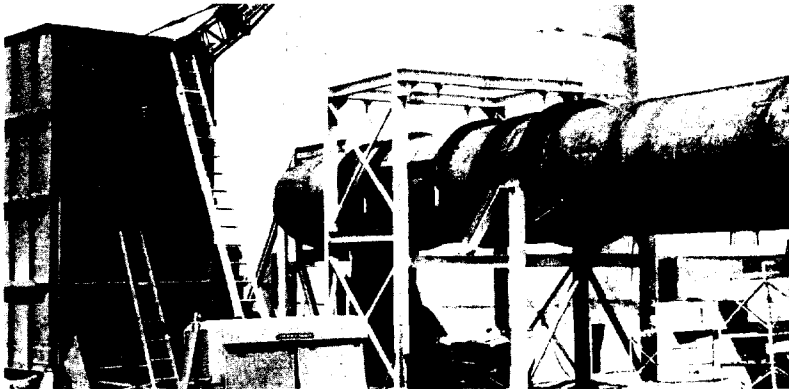


Fig. 2 — Closeup of electrostatic precipitator.

Fig. 2 shows the precipitator from a closer vantage point.

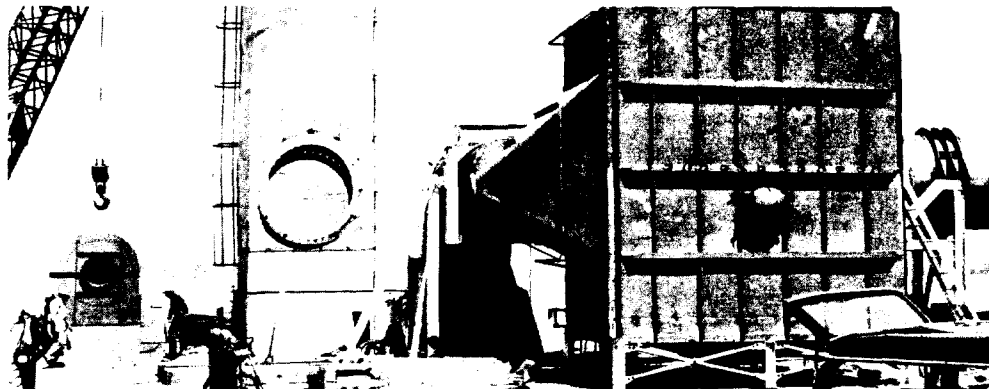


Fig. 3 — Side filter loop from the west.

Fig. 3, taken from the side, shows a blower, with a variable discharge valve, which will be used to control the air flow through the loop for

experimental purposes. We do not plan to use any blowers on a full scale filter.

The loop will be provided with instrumentation for measuring pressure, temperature, and flow at several locations, and for removing samples of the effluent for analysis. There are provisions for adding materials such as smoke to aid in removing radioactive gases, for adding sources to permit testing of the loop when the main effluent is not contaminated, and for cooling the air with water if necessary.

The loop will be ready for checkout next month, and we expect to start collecting data shortly thereafter. Artificial sources will be used at first, and fission products from test power plants when these are available. The first question to be evaluated is, of course, whether the precipitator will do the job. Other investigations will concern the effects of temperature, flow, and additives, and the best methods for removal of trapped material. These data will permit us to design a full scale filter confidently, to predict its performance, and to determine whether its advantages are worth the cost.

## Discussion

- Q. Relative to MTR installation, I was wondering what the air velocity and pressure drop is plus the filter set up?
- A. They are of the designs under conditions tested. Fast velocity - filters beds about 10' per minute.
- Q. What type of smoke --- has there been any work done in generating smokes to trap fission products?
- A. Our first experience with smoke was accidental. We were running power plant with relay of activity, smoke was released and got into the stack. A good deal of our testing involves particular chemical operations so we have smoke from the fuel and we can correlate readings from the stack monitor with the amount of fuel being burned during tests. We have released smoke consisting of burning old engine oil in engine exhausts, but we haven't really pinned the problem down. We don't have the data to know exactly what this machine will do.
- Q. Can you give us some notion of the effectiveness of this smoke?
- A. I am afraid I can't give you any really good answer. I think we are getting a factor 10- 100 increase in our tracing efficiency for radioactive materials (Iodine).
- C. (L. Silverman, Harvard) We ran some experiments - passing the two gases (benzine smoke through iodine). -- In trying to collect I did not - indicating about 35% removal --- so rapidly that the pressure drop got out of line. Activated carbon would be the ideal aerosol.
- A. (C. L. Storrs, ANP-GE) I think that is very interesting and I hope that I am not too optimistic and that it will work out. We really don't know what the thing will do.
- Q (J. J. Sabo, U. S. Public Health) You gave a pressure drop of 20 psi, could you please break that down into components, was it 20 psi or 20 inches of water - seems like a high pressure drop for the system?

- A. (D. C. Foster, ANP-GE) I believe the numbers are 20 psi. I have the tabulation of the breakdown here of 15 points in the system which I would be glad to let you check over, but under the designed test conditions for this particular system the inlet pressure was something of the order of - well it was exactly 100 psia and at the outlet the calculations indicated 81 or 82 psia. I believe the measurements made during the test indicated a somewhat smaller pressure drop but still considerably larger than a matter of inches. I think the test measurements were of the order of 10 to 12 pounds across the air system.
- Q. (B. O. Shaver, GE HAPO) What consideration is given for, say - after the filter following the precipitator in the event of carry over?
- A. (C. L. Storrs, ANP-GE) We won't have anything following the filter. Anything that comes through following precipitation will go up the stack and be diffused into the atmosphere - it undoubtedly will include meteorological control - contact reduce particular elements - Iodine 131 - and if we can reduce this be a factor of 10 with meteorological control - we can then proceed to operate with greater flexibility and without the filter.
- A. (J. Lieberman - AEC, Wash) Matter of control and filtrations will still be working together.
- Q. (Stevens) Have you tried any other dust collection equipment with a reasonable chance of success aside from the electrostatic precipitator?
- A. We have considered other things but haven't really tried anything except on a laboratory scale. High pressure drops are unacceptable in our operations - blowers are prohibitively expensive - the precipitator is about the only one that will do us very much good and stay within reasonable amount of money.
- C. (W. Boone, GE-ANP) The business of introducing carbon particulate smoke into the streams - to properly evaluate this requires a scale model - I tried loading filter papers with carbon in an attempt for better efficiency by burning engine fuel and drawing it through the filter papers - no effect whatsoever. From this I conclude it is essential that the carbon must be introduced beforehand - also in the use of a relative number with chemical fuel flow through generator engines with a factor of  $2\frac{1}{2}$  less my gross efficiency on glass fiber filter paper varied by a factor of 2. When we are running without - however, we get 80% efficiency with a piece of filter paper. - the efficiency drops to about 40% - many of the isotopes it is not influencing at all - our data is preliminary but we have some reasonably solid ground for suspecting that - will improve our efficiency all the way around. We need quite a bit more data, however.