The Use of Activated Charcoal Iodine Monitors During and Following a Release of Fission Product Iodines*

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Description of Monitoring System

The present core of the Brookhaven Graphite Reactor consists of some 4900 highly enriched uranium fuel elements loaded in 615 horizontal fuel channels passing through a 25 foot cube of graphite moderator and reflector. The core is divided into two halves (north and south) by an 8 cm. wide vertical gap in the center of the graphite (Figure 1). The cooling air enters the reactor through inlet filters, at the rate of 270,000 CFM, passes into the central gap and flows bi-directionally through the north and south halves of the core. It then enters the collecting plenums and flows into the north and south ducts. The air is first monitored by the north and south exit air monitors located within the pile building. These are moving filter tape monitors with beta scintillation detectors. They are essentially operational monitors and are maintained by reactor operations. The air then passes through the exit air filters, heat exchanger, venturi and on to the fan house where the north and south ducts join. After the fan house the air is monitored for Argon-41 by a Kanne ion chamber system. Just before the air enters the base of the stack, the air is monitored for particulate activity and Iodine-131. The Health Physics Division maintains and operates these systems. The particulate monitor consists of a moving filter tape with a beta scintillation detector and is calibrated to measure the rate of release of particulate activity.

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The release of I^{131} is monitored by continuously passing a measured volume of effluent air through a high efficiency filter and an activated charcoal trap[1] in series (Figure 2). The filter and trap are normally changed at 2 or 3 day intervals. The quantity of I^{131} on the filter and trap is determined by counting in a calibrated gamma spectrometer after sufficient time has elapsed to permit the decay of the shorter lived iodines. All necessary decay and sampling efficiency corrections are made and the total stack release of I^{131} is computed. This system of I^{131} monitoring was originally intended as an inventory monitor to accurately measure the total release of I^{131} but was not intended to act as an operational monitor.

Releases of Radioactivity

During a two-day period in September of 1962 there were three distinct short term releases of activity which were the result of a partial cladding failure on one of the 6800 fuel elements. Although these releases were not of serious proportions, they did provide valuable experience in the area of reactor effluent monitoring under emergency conditions.

The three releases were characterized as follows on the operational monitors:

1. The north duct particulate monitor showed a peak which in itself was not large enough to cause concern except that there was no similar peak on the south duct monitor. When both monitors show a peak the presumption is that some material in the cooling air has been activated. The classic example is a vehicle or stationary engine operating near the air intakes and the exhaust fumes being sucked into the reactor. When only one duct monitor shows a peak it indicates that activity has been released in that half of the reactor.

2. The Kanne chamber monitor responded promptly to a full scale reading and returned to normal which indicated a short, concentrated

release of gaseous activity.

3. The particulate monitor at the base of the stack showed a peak of modest proportions which again would not in itself be cause for alarm.

The first release occurred at 2100 hours on September 10, 1962. At 0845 the next morning the charcoal trap was taken from the sample line and placed in the gamma spectrometer within ten minutes after removal. It was immediately apparent from the display of the gamma spectrum on the 'scope of the analyzer that a release of equilibrium fission product iodines had occurred. The 0.365 MeV photo-electric peak of I^{131} was prominent and almost as large as the 0.53 MeV peak from I^{133}. Normally the I^{131} peak is almost entirely masked out and much smaller than the I^{133} peak. Under normal conditions the reactor will release about 8 mc of I^{131} per day. It was calculated that the first release totaled about 70 mc. The second and third releases contained 180 and 300 mc of I^{131} respectively.

Locating the Defective Fuel Elements

Needless to say, a great deal of effort was being expended to find the source of the releases but finding one or two damaged fuel elements out of 3400 elements is a difficult and time consuming task. Between the second and third releases the reactor was shut down and the elements in several suspected channels were inspected. These channels were suspected because of higher temperature readings on the thermocouples in these channels. The elements appeared normal so the reactor was brought up to a reduced power level, and shortly thereafter the third release occurred.

It was decided to reduce power level and to take air samples from each fuel channel. The samples were taken through the charcoal traps and the traps were monitored with a survey instrument. One fuel channel was found to be much higher than the rest. The reactor was shut-down and the elements in this channel were removed and inspected. Two of the four elements appeared to be damaged. One had a spotted appearance and the other was badly blistered. Subsequent hot cell inspections and testing of the elements proved that they were the source of the releases.

The reactor was again brought up to power and no further distinct releases occurred, but the stack samples showed that the I^{131} being released was much higher than normal, initially about 10 times higher and slowly tapering off to normal in about 20 days. It was theorized that some of the iodine from the elements had condensed or adsorbed on the duct work and exit air filters and was slowly subliming or exchanging off into the air stream. The analysis of additional air samples taken on charcoal at the north and south duct monitors and at the base of the stack supported this theory.

Table I shows the ratio of I^{133} activity to I^{131} activity in fresh and equilibrium fission products at various decay intervals.

Table I

Ratio of I^{133} to I^{131} Activity for Fresh and Equilibrium Conditions

Decay Time	Fresh	Equilibrium
0	21	2.3
1 hr	19	2.2
1 day	9.9	1.1
2 days	4.9	0.55
3 days	-	0.27
4 days	-	0.13

The ratio found in the effluent cooling air at the base of the stack under normal conditions is about 8, which corresponds to about 1-day-old fresh fission products. The sample taken during the time of the second release showed a ratio of 1.7 which is consistent with slightly aged equilibrium fission products.

On September 15, about 3 days after the last release, a set of 24-hour samples was started with samples taken at the north and south duct monitor locations and at the stack. When these samples were analyzed the following information was obtained:

	South Duct	North Duct	Stack
Ratio $\frac{I^{133}}{I^{131}}$	8	1.4	1.2

The rate of release and the ratio found in the south duct was normal. The ratio in the north duct and stack samples was not as low as one would expect for 3-or-4-day old equilibrium fission products. However, if the sample results were corrected by subtracting the normal amount of I^{131} and I^{133}, the remaining activity shows a ratio of about 0.1 which corresponds to several day old equilibrium fission products. Also, it was found that the amount of I^{131} being released from the stack was almost twice the amount found in the duct samples indicating that about 50% of the I^{131} being released was coming off the filters.

These samples then, supported the theory that the idoine released from the fuel elements had adsorbed or condensed on the duct work and filters and was slowly subliming or exchanging off into the air stream. A subsequent experiment showed that the adsorption-exchange mechanism was the dominant one. In the course of trying to develop a new operational iodine monitor, stable I^{127} was released into the reactor to produce I^{128} to check the response of the new monitor. Each time this was done, the stack charcoal iodine monitor showed a significant increase in the amount of I^{131} released. The fact that when I^{127} is put in I^{131} comes out indicates that even under normal operating conditions there is iodine adsorbed in the reactor structure which exchanges off into the air stream. This knowledge of the behavior of iodine suggests a method of decontaminating reactors or other structures.[2,3]

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Conclusions

The system of iodine monitoring in use at BNL has proved to be a satisfactory means of measuring the release of I^{131} to the environment under emergency conditions. Although the cooling air effluent is monitored by other means, which indicated that something had been released, only the system using activated charcoal traps and gamma spectrum analysis could reliably indicate that a release of equilibrium fission product iodines had occurred and accurately measure the amount released. Other media tested by BNL and others[4,5] do not have as consistently high collection efficiency as the charcoal traps. The system was also useful in locating the defective fuel elements and in explaining the behavior of iodine within the reactor structure.

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COOLING AIR SYSTEM
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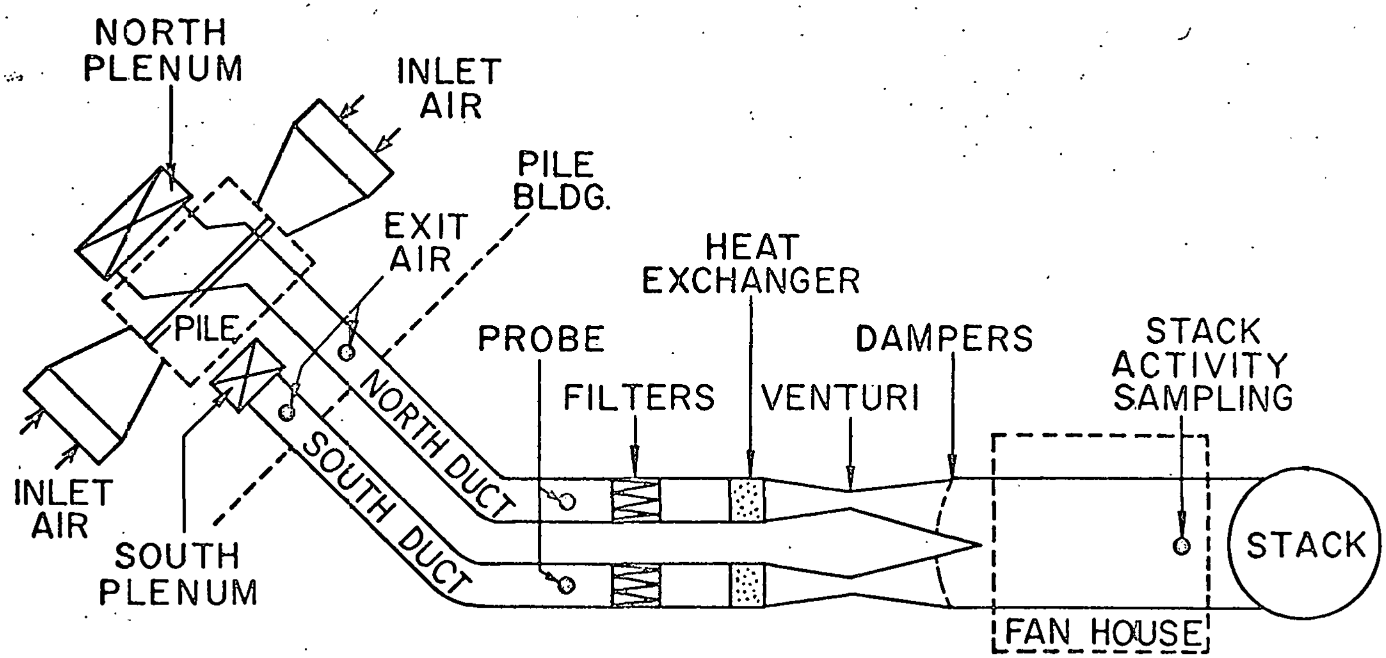
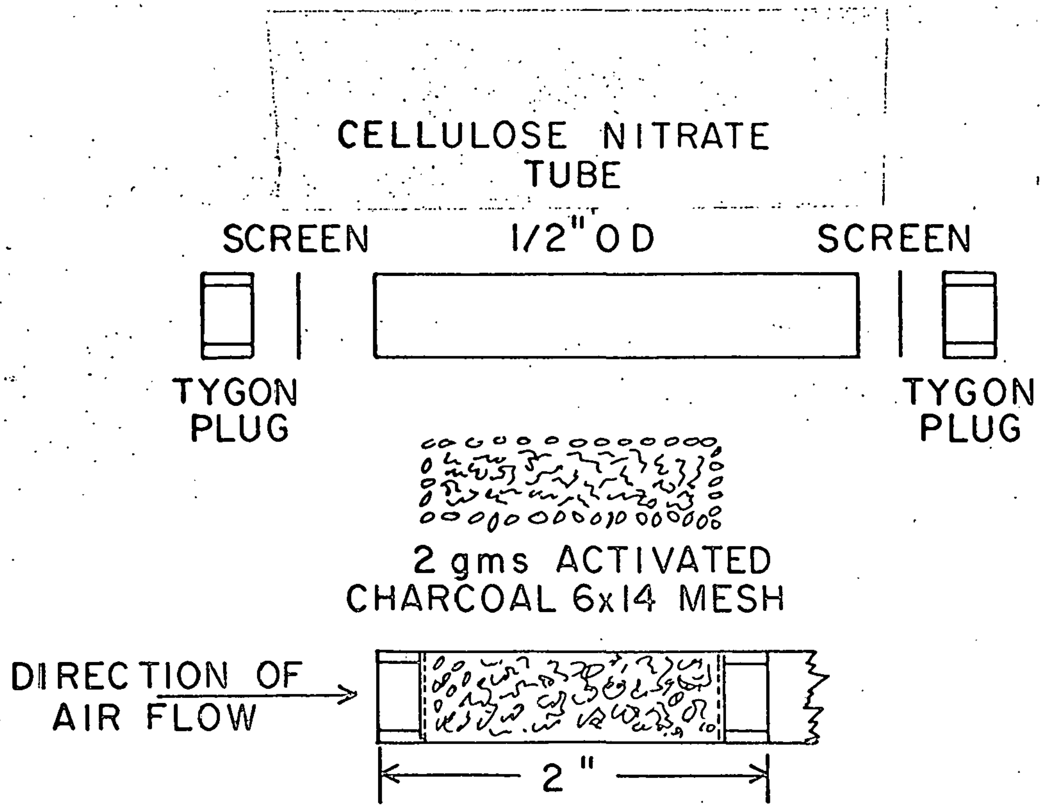


FIG 1



ACTIVATED CHARCOAL TRAP — FITS IN 5/8" NaI WELL CRYSTAL WITH COPPER LINER OR IN JIG FOR COUNTING WITH 3x3 NaI CRYSTAL

ACTIVATED CHARCOAL TRAP