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Monitoring the Cooling Air Effluent From the Brookhaven Graphite ResearchReactor at the Point of Release and in the Environment*

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PASTE

Description of Reactor and Effluent Monitoring Systems

The Brookhaven Graphite Research Reactor^{1,2,3} is an air-cooled, graphite moderated reactor with a nominal power level of 20 megawatts. The present core consists of some 4900 enriched fuel elements loaded in 615 horizontal fuel channels passing through a 7.6 meter cube of graphite moderator and reflector. Each element is 61 cm long and consists of three curved plates containing 5 grams of U²³⁵. The U²³⁵ is in the form of a Uranium-Aluminum alloy, 0.5 mm thick which is clad on all surfaces by 0.5 mm of aluminum. The core is divided into two halves (north and south) by an 8-cm wide vertical gap in the center of the graphite. A diagram of the cooling air system is shown in Figure 1. The cooling air is drawn into the reactor through inlet filters at the rate of 7600 cubic meters per minute. It passes into the central gap and flows bi-directionally through the north and south halves of the core into the respective plenum chambers. It then enters the north and south exit air ducts. The air is monitored at this point by the exit air monitors. These are moving filter tape monitors with beta scintillation detectors. They are operational monitors and are maintained by reactor operations. The air then passes through the exit air filters, heat exchangers, venturi and on to the fan house where the two ducts join. The exit air filters are not absolute filters but were designed to remove 95% of the particulate down to three microns in the unloaded condition. In their present, partially loaded, condition they are more efficient. The particulate material in the filtered effluent has been found to have a geometric

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mean diameter of 0.88 microns with a standard deviation of 2.38 determined graphically from a log probability plot of the particle size distribution⁴. After passing through the fans, the air is monitored for Argon-41 by a Kanne ionization chamber system. The sample of air is filtered before entering the chamber. At full power the rate of Argon-41 release is about 16,000 curies per day. Just before the air enters the base of the stack, it is monitored for gross particulate activity and for Iodine-131. The Health Physics Division maintains and operates these systems. The air is then released to the environment via a 100 meter stack.

The particulate activity in the stack effluent is measured by a moving filter tape monitor with a beta scintillation detector. The monitor has been calibrated to determine the amount of activity per unit volume of air released. The calibration was obtained by taking separate two-minute samples of the effluent on type HA Millipore filters and counting in an internal proportional counter. The counter efficiency is based on a Cl^{36} standard which has a maximum beta energy of 0.71 MeV. Thus, under normal operating conditions, the data obtained from the particulate monitor represents to a first approximation, the amount of particulate activity removed by a Millipore filter. The activity decays rapidly so it is necessary to choose a reference time at which to make the determination. This is ten minutes after the air leaves the base of the stack. The data are reported in terms of equivalent curies based on the beta disintegration rate.

The release of I^{131} is monitored by continuously passing a measured volume of effluent air through a high efficiency filter and activated charcoal trap in series. The charcoal trap is similar to that developed by Sill and Flygare⁵. The filter and trap are normally changed at 2 or 3 day intervals. The quantity of I^{131} on the trap and the filter 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 provides an accurate measure of the amount of I^{131} released. Under normal conditions, this is about 8 mc per day.

Prior to the continuous use of the activated charcoal traps, the filterability of the iodine in the effluent air was investigated. It was found that for short sample times, ranging from ten minutes to several hours, about 10% of the iodine was filterable on Millipore filters. However, when longer sample times mentioned above were used, the filterability of the iodine dropped to 1% or less. Under conditions of increased iodine release, to be discussed later, the quantity of iodine found on the filter ranged from 1 to 7%. A variety of factors, including the concentration of iodine, the dust loading and the adsorptive capacity of the filter media, are believed to cause this variation in the filterability of the iodine. It is obvious that ordinary filter media are not suitable for iodine sampling.

Environmental Monitoring

Argon-41, which is released in large quantities, is not metabolized by the body and merely represents an external radiation exposure hazard. Environmental radiation levels, including natural background and increments attributable to the BGRR cooling air effluent are monitored continuously at eight fixed stations. As shown on Figure 2 three of these stations are on-site at about 500 meters from the stack, four are on or near the site perimeter about 2500 meters from the stack and one is 8700 meters north of the stack.

The equipment in each station includes an ionization chamber and dynamic-condenser electrometer assembly which has been described in detail elsewhere⁶. These units are capable of accurately measuring external radiation levels in

the order of a few $\mu\text{r/hr}$. The background at a given station is assumed to be the radiation level prevailing when the meteorological record or reactor shut-down record indicates that the reactor effluent with the Argon-41 content has not been in the vicinity. The Argon-41 exposure is the difference between the gross radiation level and the background. As reported by Hull⁷, the net weekly radiation levels during 1962 attributable to Argon-41 are shown in Table I. The highest on-site annual average was 3.1 mr/week. The highest annual average at the perimeter was 1.08 mr/week.

The I^{131} concentration in the effluent cooling air is about $8 \times 10^{-10} \mu\text{c/cc}$. Figure 3 is a plot of the Predicted Annual Mean Ground Concentrations computed by the Brookhaven Meteorology Group. It is apparent that atmospheric dilution is such that the anticipated ground concentrations represent no direct inhalation problem when compared to a permissible non-occupational concentration of $5 \times 10^{-12} \mu\text{c/cc}$ calculated from the Federal Radiation Council Radiation Protection Guide⁸ for I^{131} . High volume samplers containing a particulate filter and an activated charcoal trap in series are operated at each of the perimeter stations. The filter is changed on a weekly basis and the charcoal traps on a bi-weekly basis. Gamma spectrum analyses of the samples have indicated that the average air concentrations are below the limit of detection which is about $1 \times 10^{-15} \mu\text{c/cc}$.

At a given concentration of radio-iodine, indirect intake by the ingestion of milk from cows feeding on contaminated pasture may be more significant than inhalation by at least an order of magnitude. Assuming the deposition velocity of 0.5 cm/second, a grass cover of 0.15 gms/cm^2 suggested by Hull⁹ and the 10:1 grass to milk I^{131} ratio observed during the Windscale incident¹⁰, it can be shown that an average air concentration of $4 \times 10^{-13} \mu\text{c/cc}$ would correspond to a grass concentration of $1 \times 10^{-6} \mu\text{c/gm}$ and a milk concentration of 100 $\mu\text{c/liter}$.

As indicated in Figure 3 the calculated BNL perimeter concentration of I^{131} average 4×10^{-16} $\mu\text{c/cc}$ or lower. The I^{131} concentration in milk from a cow pastured at the perimeter should be $0.1 \mu\text{c/liter}$ or less. Routine milk samples are obtained from the nearest dairy farm, some 5 km distant from the stack. The analysis procedure employs resin column collection of I^{131} from several liters of milk. The present average I^{131} concentration of the milk samples is below our limit of detection, which is about $5 \mu\text{c/liter}$.

Effluent and Environmental Monitoring Under Emergency Conditions

The adequacy of a monitoring program may be judged by its ability to provide the necessary information under emergency, as well as routine, conditions. 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 fuel elements. Although the amount of activity released was not of serious proportions, it was considerably greater than normal and served to provide valuable experience in reactor effluent monitoring under emergency conditions.

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

1. The particulate monitors at the north duct and at the base of the stack both showed peaks of modest proportion which indicated that some particulate activity has been released from the north half of the reactor core.
2. The Kanne ionization chamber monitor showed a rapid rise to full scale and returned immediately to normal, indicating short, concentrated releases of gaseous activity.

The first release occurred at 2100 hours on September 10, 1962. When the charcoal trap was removed from the sample line and placed in the gamma spectro-

meter, 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 has 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} photo-electric peak is quite small and is often obscured by the other iodine activities present. Further analysis of the sample showed that the ratio of I^{133} activity to I^{131} activity present in the effluent air at the time of the release was about 1.8. This ratio is characteristic of slightly aged equilibrium fission products. The ratio found in the effluent air under normal conditions is about 8 which is characteristic of one-day fresh fission product iodines. The normal rate of release of I^{131} from the reactor is about 8 mc per day. The first release was 70 mc and the second and third releases contained 180 and 300 mc of I^{131} respectively.

In the time interval between the third release and the removal of the defective fuel element, an additional 300 mc were released. After the element was removed another 600 mc were released over a period of 20 days. The rate of release then returned to normal. Approximately 65% of the iodine released from the element was immediately absorbed on the reactor structure and exit air filters and subsequently exchanged off into the air stream. This was confirmed three days after the last release by taking simultaneous samples at the north and south duct sample points and at the base of the stack. It was found that about half of the I^{131} being released originated from the filters.

When it became apparent that unusual amounts of I^{131} had been released to the environment, the BNL Meteorology Group was alerted and requested to predict the location of the maximum deposition and the probable distribution of activity based on the release data available. This was done and was continuously updated as more release data became available.

These predictions indicated that the maximum deposition had occurred on site within 1 km and in the northeast to southeast sector downwind from the stack. Although the largest number of grass samples were obtained from this sector, others were obtained up to a distance of 10 km from the stack. This was done to provide adequate definition of the affected area and also to provide an estimated of the prevailing background of fallout I^{131} deposition from nuclear weapons tests.

The predicted location of the maximum deposition and the probable distribution of activity at 1600 on September 13, as calculated by the Meteorology Group using estimated hourly release rates, are shown in Figure 4. The predicted maximum integrated ground concentration of I^{131} as of that time was $1.5 \times 10^{-6} \mu\text{c-sec/cm}^3$. From this, a grass concentration of $5 \times 10^{-6} \mu\text{c/gm}$ would be anticipated assuming the deposition velocity and grass cover previously indicated.

The location of the measured maximum grass concentration of I^{131} and its distribution on-site and nearby on the afternoon of September 13 are shown in Figure 5. Peaks, both of about $4.5 \times 10^{-6} \mu\text{c/gm}$, are apparent northeast and southeast of the stack. A fallout I^{131} background averaging $1.0 \times 10^{-6} \mu\text{c/gm}$, as determined from upwind and distant samples, was assumed for all the samples in the stack deposition area.

Although additional grass samples were obtained after September 13, definition of the deposition from the stack was obscured by an apparent doubling in fallout background levels during the following week. However, at no time did the measured off-site deposition of I^{131} from the release approach that which would have resulted in a significant milk contamination. The average concentration of I^{131} in milk samples from the nearest downwind dairy farm averaged $25 \mu\mu\text{c/liter}$ during the two weeks after the release. This was less

than the average concentration during the same period in milk from other dairy farms upwind and at a greater distance from the BNL site.

Conclusions

The fuel element failure which resulted in the release of fission product iodines from the reactor, though unfortunate, did provide valuable experience in effluent monitoring under emergency conditions. It was found that the monitoring program currently in use at BNL was capable of providing the required information which includes the quantity and identity of the activity released and the location and concentration of the activity in the environment.

It is of interest to review some of the records on the release of activity from the BGRR. The quantity of particulate activity and I^{131} release is reported monthly. It is summarized and presented here in terms of activity per megawatt day:

(1) Monthly release of particulate activity for 1962 (mc/MWD)

	<u>Except September</u>	<u>September</u>
Average	80	88
Range	71 - 87	

(2) Monthly release of I^{131} for 1962 (mc/MWD)

	<u>Except September</u>	<u>September</u>
Average	0.51	3.8
Range	0.49 - 0.56	

These data demonstrate the failure of particulate monitoring equipment to provide information under the conditions experienced in September of 1962 and the need for a separate system, such as that used at BNL, to reliably and accurately measure the release of vaporous iodines.

TABLE I

A⁴¹ Radiation Levels, 1962
(mr/wk)*

Month	<u>On-Site</u>			<u>Perimeter</u>				<u>Off-Site</u>
	E-10	E-11	E-12	E-2	E-4	E-7	E-9	O-6
January	0.75	1.94	2.60	0.21	0.33	0.51	0.47	0.16
February	1.52	3.89	1.50	0.63	0.06	0.77	0.25	0.36
March	2.39	2.49	1.02	0.64	0.87	0.86	0.39	-0-
April	0.62	2.26	3.97	0.32	-0-	0.84	1.26	-0-
May	2.73	2.00	3.83	0.80	0.51	0.24	1.56	0.09
June	1.35	2.07	5.35	0.86	0.40	0.26	1.83	0.21
July	1.41	2.00	4.21	0.34	0.17	0.44	2.27	0.14
August	1.38	2.44	4.33	0.79	0.42	0.28	1.14	0.14
September	1.06	1.69	2.92	0.38	0.35	0.24	1.15	-0-
October	1.12	1.99	3.03	0.36	0.28	0.73	1.21	0.11
November	1.55	4.25	2.28	0.21	0.53	0.60	0.80	0.13
December	1.76	2.50	2.11	0.13	0.45	0.38	0.66	0.16
Average	1.47	2.46	3.10	0.47	0.36	0.51	1.08	0.13

* Error estimated at ± 0.25 mr/wk, at a 90% confidence level.

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COOLING AIR SYSTEM BROOKHAVEN PILE

FIGURE 1

LOCATION OF BNL AREA SURVEY STATIONS

FIGURE 2

ANNUAL MEAN GROUND CONCENTRATION OF IODINE 131

FIGURE 3

PREDICTED GROUND CONCENTRATIONS OF I^{131} FOLLOWING STACK RELEASE

FIGURE 4

MEASURED I^{131} CONTAMINATION ON VEGETATION FOLLOWING STACK RELEASE

FIGURE 5