MAR 1 1968

THE EVALUATION OF FISSION PRODUCT AND ACTIVATION ISOTOPES IN
A REACTOR STACK EFFLUENT AND IN THE NEARBY ENVIRONMENT*

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INTRODUCTION

The Brookhaven Graphite Research Reactor (BGRR) is an air-cooled and, as its name indicates, graphite moderated nuclear research reactor. It was originally designed to operate with natural uranium fuel elements at a power level of 30 MW. To raise the neutron flux, this loading was changed during the late 1950's to one highly enriched in ^{235}U . The reactor has, until quite recently, been routinely operated at a power level of 20 MW.

Both inlet and exhaust high efficiency air particulate filters are included in the air filter system. When installed, the latter were described¹ as glass-tex cloth with the following arrestance characteristics: 95% initial to 99% at full load for standard test; 89% initial for 3-4 micron particles. In their present partially-loaded condition, the filters have been indicated by Foelix² to be somewhat more efficient. An experimental study by Billings³ suggests that this should be the case. More recently McClintock⁴ has reported that the efficiency of the exit air filters is only 80% for particulates in the 0.3 micron range. In an earlier study, Potter⁵ found that the particulate material in the filtered effluent had a geometric mean diameter of 0.88 microns with a standard deviation of 2.38 microns. From this, one would anticipate that despite the presence of the filters, the air effluent would contain a perceptible amount of radioactive air particles.

* Research carried out at Brookhaven National Laboratory under contract with the U.S. Atomic Energy Commission.

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In 1965 a by-pass charcoal air filter facility was installed. Although the BGRR air effluent may be diverted through this facility, it is not normally so routed. When the BGRR is operating around the clock at 20 MW, its air effluent contains ^{131}I in a concentration of close to $5 \times 10^{-10} \mu\text{Ci}/\text{cm}^3$. Less than 1% of this ^{131}I has been found by Foelix to be collected (on a membrane filter with a 0.45μ pore size).

The installed devices for operational sampling and control of the stack effluent include a Kanne ionization chamber (primarily to establish the concentration of ^{41}Ar), a moving filter tape air particulate sampler with a beta scintillation detector, and a charcoal iodine trap, of the type developed by Sill⁶.

Until recently, routine identification and measurement of specific isotopes in the BGRR effluent has been limited to ^{41}Ar and ^{131}I . The moving tape monitor data has been quite consistent. The stack release rate of gross-beta air particulates during BGRR full power operation (at 20 MW) is about $17.5 \mu\text{Ci}/\text{sec}$ in a concentration of $1.4 \times 10^{-7} \mu\text{Ci}/\text{cm}^3$. By analogy to the observed⁷ ratio between stack effluent release rate of ^{131}I and the measured ^{131}I concentration at the BNL perimeter, the greatest yearly average gross-beta concentration of air particulates at ground level would be about $6 \times 10^{-13} \mu\text{Ci}/\text{cm}^3$. Decay studies conducted some years ago indicated that the apparent half-life of gross-beta mixture of isotopes in the stack effluent was rather short. It has therefore appeared that the average environmental concentration of any individual isotope contained in the effluent would be much less than $6 \times 10^{-13} \mu\text{Ci}/\text{cm}^3$, and would therefore be difficult to detect in the presence of the relatively larger backgrounds of air activity produced

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by nuclear weapons testing that prevailed during the early 1960's.

A single air particulate sample from the stack effluent, obtained late in 1964, was followed in some detail by a series of gamma spectrum analyses extending over the following year. The isotopes present and their concentrations, as previously reported by Hull⁸ are shown in Table I. The data do confirm that the isotopes present in the largest concentrations do indeed have relatively short half-lives and that (by analogy to ¹³¹I observations referred to above) the environmental concentrations of most, if not all, of the longer lived isotopes in the sample would be in the order of 10^{-15} $\mu\text{Ci}/\text{cm}^3$ and too small to be detectable.

As the prevailing background of air activity produced by weapons testing has declined during recent years, the BNL Health Physics Division and Meteorology Group have become interested in the feasibility of detecting one or more of the isotopes in the stack effluent as both short or long term tracers to supplement the previous longer term studies of stack effluent dispersion phenomena at Brookhaven. The work reported herein was carried on as a portion of this feasibility investigation.

STACK EFFLUENT SAMPLING AND ANALYSIS

Although the Sill charcoal cartridge is satisfactory for stack ¹³¹I emission control purposes, it was too small and operated at too low a flow rate to be useful for these studies. Since a study of the characteristics of the ¹³¹I in the effluent was also an objective, a "May-Pak"⁹ sampler was used. As shown in Fig. 2, the unit comprises, in sequence:

1. Two or more copper mesh discs to remove I_2 .
2. A membrane filter to remove air particulates.
3. One or more charcoal loaded filter papers.
4. A small bed of activated charcoal.
5. A glass fibre paper to retain the charcoal.

The brass cartridge containing these elements has an inner diameter of 47 mm. A flow rate of about 0.75 cfm was maintained, so that the air velocity through the sampler was about 25 fpm and the residence time in the charcoal bed about 0.15 sec.

Daily stack samples were run with the complete "May-Pak" for three months, starting in June 1966. For the balance of the year under consideration in this report, it was modified to a conventional sampler containing a particulate filter, charcoal bed and retaining paper.

The identification and evaluation of the concentrations of specific isotopes present in the samples has been solely by gamma spectroscopy employing two NaI 4" x 2" crystals and a 400-channel analyzer. Two crystals were employed solely to improve counting geometry. The high voltage on the detectors was adjusted so that they produced identical signals, and these signals were then mixed and fed through a common amplifier to the analyzer.

An initial count was made on the complete sampler soon after and again ten days after the termination of the sampling period, primarily for the determination of short-lived isotopes. Following this the samples were disassembled and the individual elements composited into monthly batches. Shortly after the end of each month and again after about a year's decay, these batches were analyzed to determine the intermediate and long-lived isotopes present in each element of the sampler. Since 1% or less of the total activity of any isotope present in the complete sampler was found in the initial individual analyses of the outer brass container and of the retaining paper, analysis of these elements were not continued.

STACK EFFLUENT DATA

Since the initial counts of the complete filter assemblies showed little variation, one representative daily sample from each month was selected to obtain a yearly average stack emission concentration of those isotopes with half-lives of from several hours to a few days. Twenty-one hour ^{133}I and 35 hour ^{82}Br were the only isotopes in this category found in quantifiable amounts. The concentrations of these isotopes found in the representative samples and their yearly average concentrations, as determined by this method, are shown in Table II. The concentration of ^{131}I found in these same samples is also indicated. That the latter is in good agreement with the average of daily ^{131}I monitoring data confirms the representative nature of the selected samples.

Both activation and fission product isotopes were found in most components of yearly samples. These included: 5.3 yr. ^{60}Co , 245 day ^{65}Zn , 65 day ^{95}Zr (with 35 day ^{95}Nb); 40 day ^{103}Ru , 1.0 yr. ^{106}Ru , 60 day ^{124}Sb , 2.7 yr. ^{125}Sb , 8 day ^{131}I , 30 yr. ^{137}Cs , 12.8 day ^{140}Ba (with 40.2 hr. ^{140}La), 32.5 day ^{141}Ce , 285 day ^{144}Ce and 47 day ^{203}Hg . In addition, an isotope tentatively identified as 120 day ^{75}Se was found in monthly composite samples from September on. The yearly average concentrations of these isotopes found in the particulate and charcoal collections in total concentrations greater than 1 pCi/m^3 are shown in Table III. For this purpose the first few months Cu screen collection has been included with that of the membrane filter and the charcoal loaded filter paper with the charcoal bed collection. As is evident in Table III, the major fractions of several isotopes, in addition to ^{131}I , were somewhat unexpectedly found in the charcoal collection.

During the first few months that the complete May-Pak was operated, a nearly uniform ratio of 1:5 was found between the fraction of most isotopes (except ^{131}I and $^{140}\text{Ba-La}$) collected by the screen and that collected by the particulate filter. This suggested that the screen was operating to some extent as a particulate filter. A very small fraction of $^{140}\text{Ba-La}$ was found on the charcoal loaded paper. The concentrations of ^{131}I collected by each component of the pak are shown in Table IV.

The May-pak does not give a precise separation of the different forms in which ^{131}I may be encountered. Following May's scheme, it can be stated from the data that (at the stack) close to 8% of the BGRR effluent ^{131}I was present as elemental I_2 , less than 2% was attached to particulates, as much as 63% was present as readily filterable compounds of iodine and not much more than 27% as relatively less reactive organic compounds (such as methyl iodide).

ENVIRONMENTAL MONITORING AND SAMPLE ANALYSIS

Measurements of the two-week average concentrations of ^{131}I at distances from 1,200 m to 6,200 m from the BGRR stack, have been reported by the BNL Environmental Monitoring group for the past several years^{8, 10}. The primary purpose for these measurements has been the verification of the long-term estimates from diffusion equations made by the BNL Meteorology group. Despite the relatively large 20 cfm flow rate of the positive displacement pumps employed, the amounts of ^{131}I collected in many routine two-week samples have been at or near the lower detection limit of about 10 pCi.

Recent one-day samples were obtained and analyzed within a day after collection to determine if the ^{82}Br and ^{133}I components of the stack effluent could be used for the verification of shorter-term calculations.

Twenty-four hour average concentrations of both isotopes in the order of $5 \times 10^{-14} \mu\text{Ci}/\text{cm}^3$ have been found in each of three one-day samples obtained when other data indicated that the effluent plume was on the ground at a monitoring station on three recent occasions. The first two were at a station (S-13) which is 1,350 m distant and downwind from the stack for winds from 217° , the third at a station (P-2) 2,160 m distant and downwind for winds from 167° .

The sampling periods were subjected to the routine analysis usually used to determine ground level concentrations solely from meteorological data, and the results are summarized in Table V .

The procedure involved the computation of the mean hourly ground level concentration at the sampling point on the basis of dispersion parameters developed in earlier studies at Brookhaven, and the reported stack output rates. For each hour the mean concentration was multiplied by the filter flow rate and a half life correction was made to account for the decay between that hour and the time at which the sample was removed.

The results shown do not agree with the observed values as well as most samples have in the past. In every case the predicted collection is significantly less than that actually observed, whereas similar measurements involving the ^{41}Ar component (in one instance for the same hours) have shown much better agreement. No reason for the discrepancy has as yet been determined.

In an analogous manner precipitation samples have been obtained, using large 1.95 m^2 pan collectors, on two occasions during the past year when it could be established that the plume was over a station (and the collector) during a period of precipitation. The collections were analyzed promptly

for indications of ^{82}Br and ^{131}I . The measured amounts of activity in these collections are indicated in Table VI.

The meteorological conditions associated with the precipitation samples have been scrutinized but not formally analyzed. The variability of the precipitation in both instances was such that it was almost impossible to determine within a factor of 3 what sort of collection might have been expected, and with the relatively long sampling periods involved, it was difficult to dissociate dry deposition from the contribution in the precipitation. Meaningful comparisons with calculated collections, using currently accepted washout factors, have not been practicable. The development of methods for a more precise short-term definition of rates of precipitation is planned, and these efforts to verify washout calculations are being continued.

SUMMARY

A study of one year's duration of the gamma emitting isotopes present in concentrations greater than 1.0 pCi/m^3 in the BGRR stack effluent has been made. For the first three months a May-pak sampler was employed, and a conventional particulate and charcoal filter employed for the balance of the year.

The only relatively short-lived isotopes detected were ^{82}Br in a concentration of $4.2 \times 10^3 \text{ pCi/m}^3$ and ^{133}I in a concentration of $2.2 \times 10^3 \text{ pCi/m}^3$. The May-pak indicated that most of the BGRR effluent radioiodine was in states other than methyl iodide. The intermediate and longer lived isotopes detected included both activation and fission products. Among the activation products, ^{203}Hg was present in the greatest concentration (21 pCi/m^3) and among the fission products, ^{131}I ($4 \times 10^2 \text{ pCi/m}^3$). Ground level air concentrations of ^{82}Br and ^{133}I in the order of $5 \times 10^{-14} \text{ } \mu\text{Ci/cm}^3$ were found in three separate one-day samples. The amounts were somewhat less than those

calculated from meteorological dispersion considerations.

Two precipitation samples have been obtained at times when the BGRR plume was over a collector. Although ^{82}Br and ^{131}I were detected, the definition of the precipitation was found to be inadequate to permit the verification of washout factors.

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TABLE I

Stack Effluent Radionuclide Concentration

Nov. 11 - 13, 1964 Air Particulate Filter

(with half-lives >0.1 hours)

<u>Activation Isotopes</u>	<u>T_{1/2}</u>	<u>pCi/m³</u>	<u>Fission Product Isotopes</u>	<u>T_{1/2}</u>	<u>pCi/m³</u>
²⁴ Na	15.0 h	472 ± 25%	¹⁰¹ Mo-Tc	14 m	7,590 ± 25%
⁸² Br	35.7 h	111 ± "	¹³⁴ I	53 m	5,830 ± "
⁶⁵ Zn	245 d	2.4 ± "	¹³⁵ I	6.7 hr	4,020 ± "
⁶⁰ Co	5.26 y	0.7 ± 50%	⁹⁷ Zr-Nb	17 hr	181 ± "
²⁰³ Hg	47 d	1.0 ± "	¹³³ I	20.8 hr	1,310 ± "
			⁹⁹ Mo	66 h	156 ± "
			¹³¹ I	8.05 d	130 ± "
			¹⁴⁰ Ba-La	12.8 d	108 ± "
			²³⁹ Np	2.10 d	6.8 ± 50%
			⁹⁵ Zr-Nb	65 d	3.3 ± "
			¹⁰³ Ru	40 d	2.0 ± "
			¹⁴¹ Ce	32.5 d	1.1 ± "
			¹⁴⁴ Ce	285 d	1.0 ± "
			¹³⁷ Cs	30 y	0.6 ± "
			¹⁰⁶ Ru	1.0 y	0.4 ± "
Total		587			19,340

TABLE II

^{82}Br , ^{131}I and ^{133}I Average Concentration
 $(10^{-9} \mu\text{Ci}/\text{cm}^3)$

<u>Month</u>	<u>^{82}Br</u>	<u>^{131}I</u>	<u>^{133}I</u>
June	4.13	0.58	1.36
July	6.55	0.50	4.50
Aug.	5.85	0.42	3.42
Sept.	3.55	0.40	1.74
Oct.	4.84	0.52	3.69
Nov.	4.86	0.34	2.76
Dec.	3.94	0.32	1.21
Jan.	2.81	0.42	1.70
Feb.	3.44	0.31	1.56
Mar.	2.24	0.41	1.32
Apr.	3.20	0.47	1.80
May	4.45	0.43	1.89
Yearly Average Conc.	4.16	0.43	2.17
Emission Rate ($\mu\text{Ci}/\text{sec}$)	0.52	0.05	0.27

TABLE III

Yearly Average Concentration of Intermediate and Long-Lived Gamma Emitting Isotopes in BGRR Stack Effluent

<u>Sample Component</u>	Activation Isotopes (pCi/m ³)									
	⁶⁰ Co		⁶⁵ Zn		⁷⁵ Se*		¹²⁴ Sb		²⁰³ Hg	
	Conc.	%	Conc.	%	Conc.	%	Conc.	%	Conc.	%
Particulate	1.7	96	8.0	96	1.4	27	2.3	82	1.0	5
Charcoal	<u>0.1</u>	<u>4</u>	<u>0.3</u>	<u>4</u>	<u>3.8</u>	<u>73</u>	<u>0.5</u>	<u>18</u>	<u>20.3</u>	<u>95</u>
Total	1.8	100	8.3	100	5.2	100	2.8	100	21.3	100

*Identification tentative.

<u>Sample Component</u>	Fission Product Isotopes (pCi/m ³)													
	⁹⁵ Zr-Nb		¹⁰³ Ru		¹⁰⁶ Ru		¹³¹ I		¹³⁷ Cs		¹⁴⁰ Ba-La		¹⁴¹ Ce	
	Conc.	%	Conc.	%	Conc.	%	Conc.	%	Conc.	%	Conc.	%	Conc.	%
Particulate	1.4	77	2.3	60	1.1	79	19.	4.5	2.0	92	9.0	21	0.9	7
Charcoal	<u>0.4</u>	<u>23</u>	<u>1.6</u>	<u>40</u>	<u>0.3</u>	<u>21</u>	<u>413.</u>	<u>95.5</u>	<u>0.2</u>	<u>8</u>	<u>33.5</u>	<u>79</u>	<u>12.1</u>	<u>93</u>
Total	1.8	100	3.9	100	1.4	100	432.	100	2.2	100	42.5	100	13.0	100

TABLE IV

May-Pak Separation of BGRR Stack EffluentJune-August 1966

Pak Element	Concentration (pCi/m ³)	%
Cu screen	34	8
Membrane filter	9	2
Charcoal loaded paper	281	63
Charcoal bed	<u>123</u>	<u>27</u>
Total	447	100

TABLE V

Comparison Between Observed and Calculated Collections

(pCi)

<u>Station</u>	<u>Sampling Period</u>	<u>Calculated</u>			<u>Observed (Ci x 10⁻¹²)</u>		
		<u>¹³¹I</u>	<u>¹³³I</u>	<u>⁸²Br</u>	<u>¹³¹I</u>	<u>¹³³I</u>	<u>⁸²Br</u>
S-13	9/22 to 9/26 (101.2 hours)	1.5	2.6	9.3	6.8	10.6	29.8
S-13	9/26 to 9/27 (23.6 hours)	2.1	5.2	19.0	5.5	11.5	32.2
P-2	10/9 to 10/11 (42.4 hours)	0.9	1.2	9.7	5.1	16.2	25.8

Mean Calculated/Observed Ratios

<u>¹³¹I</u>	<u>¹³³I</u>	<u>⁸²Br</u>
.26	.24	.43

TABLE VI

Precipitation Collections of ^{82}Br and ^{131}I From BGRR Effluent Plume

<u>Date</u>	<u>Station</u>	<u>Downwind</u>		<u>Precipitation</u>		<u>Collected Activity</u>	
		<u>Dist.</u>	<u>Dir.</u>	<u>Hrs. Present</u> *	<u>Amt.</u>	^{82}Br (nCi/m ²)	^{131}I
9/21/66	Met.	1300 m	90°	12	1.82 in.	0.31	0.04
12/13/66	P-4	2250 m	63°	9	0.70 in.	0.30	0.03

* When plume was also indicated over the collector.

FIGURES

Fig. 1 BGRR Cooling Air System

Fig. 2 May-Pak Sampler

FIG. 1

FIG. 2