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〈論 文〉

A Study on Gaseous a) Effluents from BMRR

Brookhaven 醫學研究用 原子爐에서의 氣體噴出物에 관한 硏究

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국 문 초 록

Brookhaven 醫學研究用 原子爐에서 放出되는 氣體噴出物에 관하여 연구 되었다. 모든 氣體試料는 γ-線 分光計測에 의하여 분석되었다. 試料 중에 존재하는 것으로 확인된 核種으로서, $^{82}Cl(T_{1/2} = 35.30$ 時間)이 가장 뚜렷하였다. 그 외의 확인된 核種은 $^{38}Cl(T_{1/2} = 37.24$分), $^{41}Ar(T_{1/2} = 1.82$時間), $^{106}Rh(T_{1/2} = 29.80$秒), $^{133}Te(T_{1/2} = 12.45$分)이었다.

3MW 原子爐 出力에서 pre-filter bank를 통과한 氣體噴出物 중에서 ^{41}Ar의 濃度는 2.436Bq/cc로 계산됨으로써 ^{41}Ar의 放射能 放出率은 8.51×10^{9}Bq/MW-h로 산정되었다. filter bank의 放射能 除去效率(%)은 ^{38}Cl의 경우 97.84%, ^{41}Ar은 14.15%, ^{82}Br은 98.70% 그리고 ^{106}Rh은 98.81% 각각 산정되었다. 한편, charcoal trap과 millipore filter 에서 확인된 기타 核種들로서 ^{24}Na, ^{72}Ga, ^{92}Sr, ^{97}Zr, ^{132}I, ^{133}Te, ^{141}Ce, ^{153}Sm과 ^{154}Pm은 filter bank에 의해서 완전히 제거되었다.

INTRODUCTION

The Brookhaven Medical Research Reactor, a 5 MW, is a heterogeneous, tank-type reactor designed exclusively for medical and biological studies. The medical research reactor is water-moderated and has an air cooled graphite reflector surrounding the reactor vessel.

The objectives of this study were identification of the radionuclides present in the gaseous effluent, calculation of their concentrations in the pre-filter bank, determination of their emission rates, and estimation of the % removal efficiency of filter bank.

The air stream flowing into the filter bank from the reactor contains air activation products by neutron and fission products from fuel elements. The data obtained from the gamma spectrum analyses provided information that was valuable in identification of the radionuclides that originate from fission and activation processes.

EXPERIMENTAL METHODS

Air enters the reactor buiding through a 76.2cm diameter pipe and butterfly valve at the bottom of a fresh air shaft located in the basement. An air conditioning unit processes the air before discharge to the

(a) Brookhaven Medical Research Reactor.

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main and upper levels of the building. The air returns to the basement via open stairwells and may take two paths: (1) through a filter, blower, and 40.6cm pipe which leads to the 45.7m stack: (2) through inlet filters, ducts, and channels where the air cools the thermal shield and graphite reflector before going through the filter bank and being discharged to the stack. Figure 1 is schematic diagram showing air flow in the medical research reactor. During normal reactor operation, the air flow rate is $2.93m^3/s$ through the filter bank.

The air samples were collected simultaneously at two sampling ports before and after filter bank in the basement. The reactor operation for air sampling was carried out at the reactor power of 3 MW. Air sampling time was 30 minutes at the air flow rate of about 1180 cc/s through air collection equipment, millipore filter-charcoal trap-air chamber-rotameter-air pump series. Figure 2 shows the sampling sites in pre- and post-filter bank. The practical samplings were done in one hour after reactor power reached at the 3 MW level to insure that the thermal equilibrium of reactor had been established.

The sampling air was drawn through a millipore filter (pore size: 0.8 μ), a charcoal trap (bed length: 7 cm), an air chamber (collection volume: 1710 cc), and a HP vacuum pump with rotarymeter at both sampling ports.

At the end of the air sampling time, the stopcocks of two air chambers were simultaneously shut off and charcoal traps were sealed at their both ends with corks. Two sets of three sample types were removed to the counting room in Building 535A of Safety & Environmental Protection Division where Ge(Li) and NaI (Tl) detectors are available for gamma spectrum analysis.

The millipore filter was placed on a planchet and counted both the 10.2cmϕ NaI(Tl) detector and the 145cc coaxial Ge(Li) detector. The charcoal trap was placed in a 15ml bottle holder and also

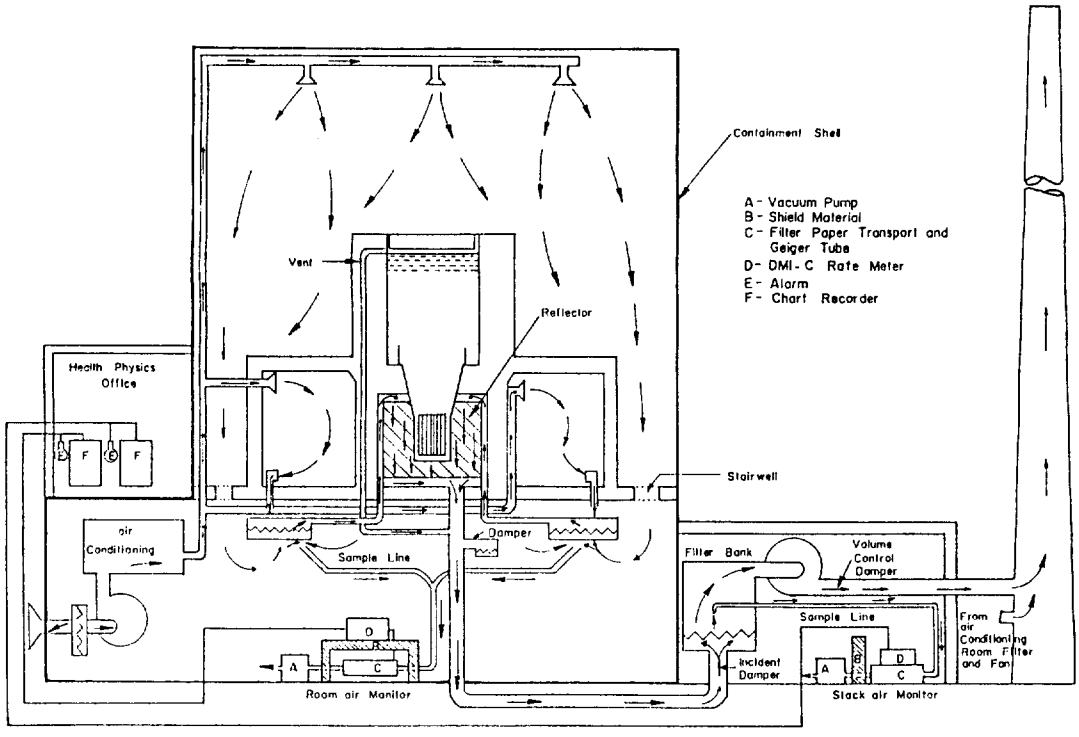

Figure 1. Air flow in medical research reactor at BNL

counted on both the 10.2cmϕ NaI(Tl) detector and the Ge(Li) detector. The Ge(Li) detector was calibrated for the 47mm filter and 15ml bottle geometries. The 10.2cmϕ NaI(Tl) detector has been calibrated for the planchet geometry on daily basis. Since no calibration existed for the 1710cc air chamber geometry, the 20.3cmϕ NaI(Tl) detector was calibrated with ^{22}Na standard sources (5.032 x 10^3 Bq as of 3-18-60) because ^{41}Ar reported in the previous studies [1,2] emits gamma-rays with energy of 1.294 MeV which is similar to ^{22}Na gamma energy of 1.275 MeV.

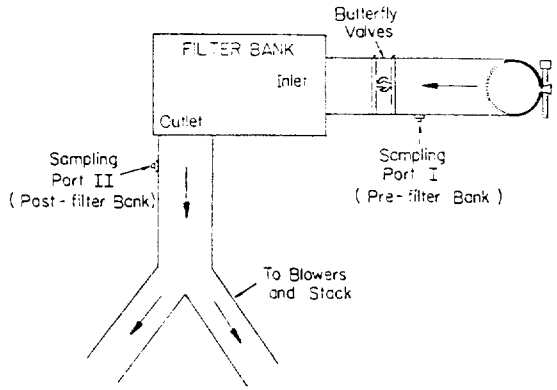

Figure 2. Air sampling sites (Top View)

Figure 3 is a calibration curve which was plotted with the peaks from NBS standards, ^{137}Cs and ^{60}Co, picked out by the 10.2cmϕ NaI(Tl) detector. According to this curve, the energy calibration is averaged to be 6.02 keV/ch.

The data obtained on the Ge(Li) detector provided information for both qualitative and quantitative evaluation on the radionuclides present possibly in the millipore filter samples and charcoal trap samples. Any interference peaks were differentiated from the correct one by using half-life calculated for radionuclide identification.

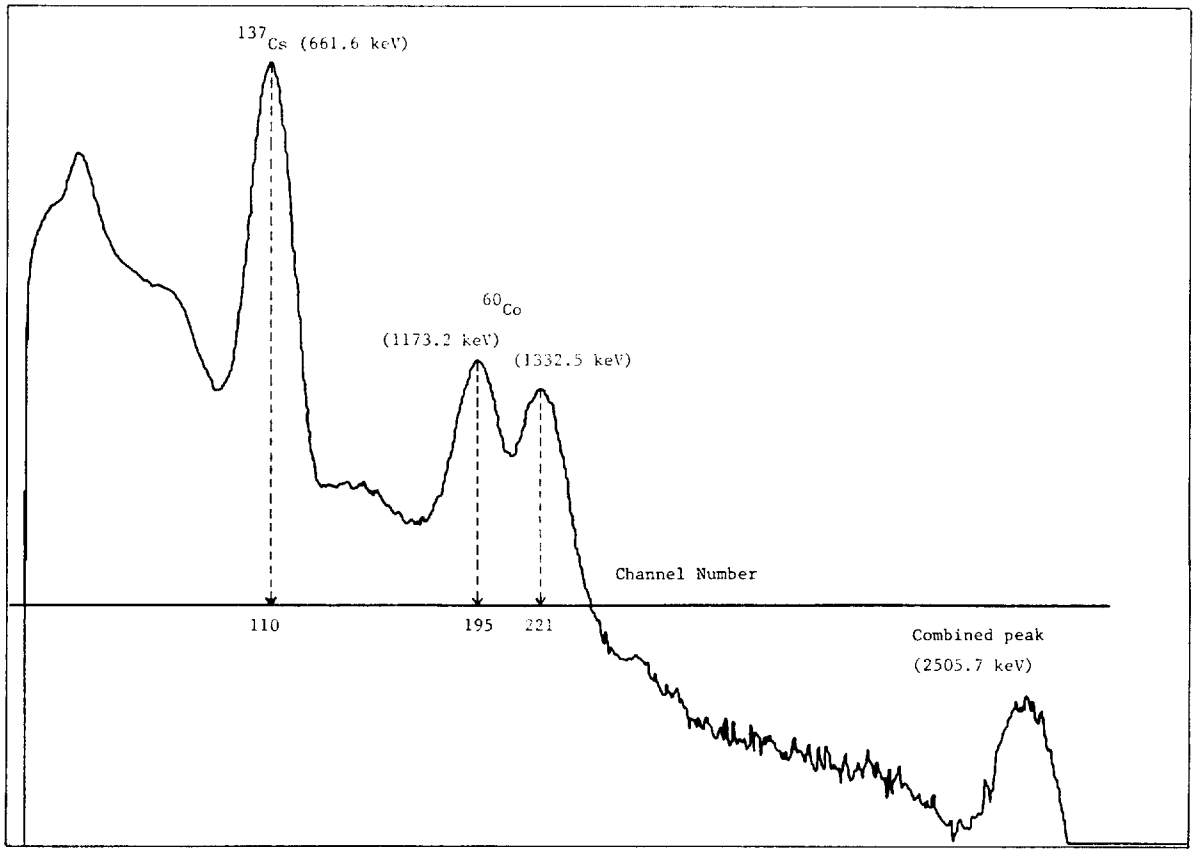

Figure 3. Calibration curve for millipore filter and charcoal trap with 10.2cmϕ NaI(Tl) detector

The analysis for ^{41}Ar was different from the analysis for other radionuclides because the 20.3cmϕ NaI(Tl) detector was used for ^{41}Ar analysis, while others were analyzed by the Ge(Li) detector. The 10.2cmϕ NaI(Tl) detector was used as a qualitative tool rather than to quantitate the radioactivity of the samples.

The following equation was used for determining the concentration of ^{41}Ar: [3]

$$C = \frac{\lambda^2 \cdot N_n \cdot e^{\lambda td}}{Fe \cdot (1 - e^{-\lambda ts})(1 - e^{-\lambda tc}) \cdot E \cdot f} \quad (1)$$

where,

C = concentration of ^{41}Ar in dps/cc

λ = decay constant of ^{41}Ar(=1.058 x 10^{-4} s^{-1})

N_n = net peak counts integrated

F_e = air equivalent flow rate (=19.5cc/s)

E = efficiency of the 20.3cmϕ NaI(Tl) detector (3.36 % for ^{22}Na peak energy of 1.275MeV)

f = abundance for ^{41}Ar gamma energy of 1.294 MeV

t_a = delayed time in sec

t_s = sampling time in sec

t_c = counting time in sec

The concentration of ^{41}Ar in the pre-filter bank was calculated to be 2.436 Bq/cc. Accordingly, the emission rate was estimated to be about 2.57 x 10^4 MBq/h at the 3 MW level. The concentration of other radionuclides present in the gaseous effluent was calculated by using the following equation: [3]

$$C = \sum_{i=1}^{n} A_i \cdot \frac{e^{\lambda td}}{F} \quad (2)$$

where,

C = concentration in dps/cc

A_i = gamma activity in γ's/s of specific i-th gamma-ray.

F = air collection flow rate (= 1180 cc/s)

t_d = delay time in sec

λ = decay constant in s^{-1}

Table 1. Sampling and counting parameters

Reactor Run: 14:00 - 15:00, 7–26–79, 3 MW
Sampling Time: 15:00-15:30

Sample Type		Delayed Time t_d(min)	Counting Time t_c(s)	Detector
(1)	Air Chamber			
		15	100	20.3cmϕ NaI(Tl)
	Pre-filter	155	300	20.3cmϕ NaI(Tl)
	Bank	380	2000	20.3cmϕ NaI(Tl)
		617	3000	20.3cmϕ NaI(Tl)
		25	100	20.3cmϕ NaI(Tl)
	Post-filter	140	300	20.3cmϕ NaI(Tl)
	Bank	425	2000	20.3cmϕ NaI(Tl)
		745	3000	20.3cmϕ NaI(Tl)
(2)	Charcoal Trap			
		29	100	145 cc Ge (Li)
		80	100	10.2cmϕ NaI(Tl)
		165	600	10.2cmϕ NaI(Tl)
	Pre-filter	317	540	145 cc Ge (Li)
	Bank	472	1000	10.2cmϕ NaI(Tl)
		498	1800	145 cc Ge (Li)
		953	2000	10.2cmϕ NaI(Tl)
		1469	1800	145 cc Ge (Li)
		4954	4000	145 cc Ge (Li)
		72	100	145 cc Ge (Li)
		100	100	10.2cmϕ NaI (Tl)
		200	600	10.2cmϕ NaI(Tl)
	Post-filter	360	600	145 cc Ge (Li)
	Bank	503	1000	10.2cmϕ NaI(Tl)
		547	1800	145 cc Ge (Li)
		1049	2000	10.2cmϕ NaI(Tl)
		1382	1800	145 cc Ge (Li)
		4680	4000	145 cc Ge (Li)
(3)	Millipore Filter			
		45	100	10.2cmϕ NaI(Tl)
		115	200	145 cc Ge (Li)
	Pre-filter	225	600	10.2cmϕ NaI(Tl)
	Bank	374	900	145 cc Ge (Li)
		542	1000	10.2cmϕ NaI(Tl)
		605	1800	145 cc Ge (Li)
		1045	2000	10.2cmϕ NaI(Tl)
		1310	1800	145 cc Ge (Li)
		4867	4000	145 cc Ge (Li)
		60	100	10.2cmϕ NaI(Tl)
		187	200	145 cc Ge (Li)
	Post-filter	285	600	10.2cmϕ NaI(Tl)
	Bank	360	900	145 cc Ge (Li)
		580	1000	10.2cmϕ NaI(Tl)
		1411	1800	145 cc Ge (Li)

Table 2. Radionuclides identified by Ge(Li) detector

Energy, E (keV) E_1 (a)	E_2 (b)	Nuclide & Half-life [5]		Energy, E (keV) E_1 (a)	E_2 (b)	Nuclide & Half-life [5]	
Millipore Filter				Charcoal Trap			
				69.7	69.7	^{153}Sm	46.7 h
142.4	145.4	^{141}Ce	32.501 d				
220.8	221.1	^{82}Br	35.30 h	220.8	221.1	^{82}Br	35.30 h
272.7	273.5	^{82}Br	35.30 h	227.7	273.5	^{82}Br	35.30 h
				291.9	295.0	^{82}Br	35.30 h
513.1	511.7	^{106}Rh	29.80 s	531.1	511.7	^{106}Rh	29.80 s
553.2	554.3	^{82}Br	35.30 h	553.2	554.3	^{82}Br	35.30 h
621.2	619.1	^{82}Br	35.30 h	621.1	619.1	^{82}Br	35.30 h
669.1	667.7	^{132}I	2.30 h	669.1	667.7	^{132}I	2.30 h
700.6	698.4	^{82}Br	35.30 h	700.6	698.4	^{82}Br	35.30 h
745.8	743.5	^{97}Zr	16.90 h				
778.6	776.7	^{82}Br	35.30 h	778.6	776.7	^{82}Br	35.30 h
830.6	827.8	^{82}Br	35.30 h	830.6	827.8	^{82}Br	35.30 h
849.8	844.5	^{133}Te	12.45 min	849.8	844.5	^{133}Te	12.45 min
1,047.0	1,044.0	^{82}Br	35.30 h	1,047.0	1,044.0	^{82}Br	35.30 h
				1,296.3	1,293.6	^{41}Ar	1.827 h
				1,321.0	1,317.4	^{82}Br	35.30 h
1,371.7	1,368.5	^{24}Na	15.02 h				
1,386.7	1,386.0	^{92}Sr	2.71 h				
1,478.4	1,474.9	^{82}Br	35.30 h	1,478.4	1,474.9	^{82}Br	35.30 h
1,645.8	1,642.4	^{38}Cl	37.24 min	1,645.8	1,642.4	^{38}Cl	37.24 min
				1,654.0	1,651.2	^{82}Br	35.30 h
1,695.5	1,656.0	^{154}Pm	2.7 min				
				1,824.0	1,822.6	^{82}Br	35.30 h
1,874.8	1,874.0	^{82}Br	35.30 h	1,874.8	1,874.0	^{82}Br	35.30 h
				2,032.5	2,030.4	^{72}Ga	14.10 h
2,172.5	2,167.5	^{38}Cl	37.24 min	2,172.5	2,167.5	^{38}Cl	37.24 min
2,760.0	2,754.1	^{24}Na	15.02 h				
513.1	511.7	^{106}Rh	29.80 s				
553.2	554.3	^{82}Br	35.30 h	553.2	554.3	^{82}Br	35.30 h
				621.2	619.1	^{82}Br	35.30 h
				700.6	698.4	^{82}Br	35.30 h
				778.6	776.7	^{82}Br	35.30 h
				830.6	827.8	^{82}Br	35.30 h
				1,047.0	1,044.0	^{82}Br	35.30 h
				1,296.3	1,293.6	^{41}Br	1.827 h
				1,478.4	1,474.8	^{82}Br	35.30 h
				1,645.8	1,642.4	^{38}Cl	37.24 min
				2,172.5	2,167.5	^{38}Cl	37.24 min

(a) E_1's were obtained from the data given by the Ge(Li) detector.

(b) E_2's were chosen as standard gamma energies from reference 4.

During the medical research reactor operation, air flow is ultimately exhausted from the air cooling system by one of two damper equipped centrifugal blowers and is discharged to the base of the stack. The blowers, each rated at $2.93 m^3/s$, provide the necessary negative pressure to insure the inward flow of active gases and their eventual path through filters to the stack.

In practice, the following equation was used for estimating the emission rates of radionuclides:[3)]

$$R_e = C \times F_s \times f_c \quad (3)$$

where,

R_e = emission rate in Bq/h at 3 MW

C = concentration in dps/ cc

F_s = air flow rate to the stack (= $2.93 m^3/s$)

f_c = conversion factor (=3.60×10^9 cc-s/m^3-h)

RESULTS

Data collections and analyses have been done on the basis of Table 1. Some selected plots are shown in Figures 4 through 9. In general, the millipore filter samples contained less radioactivity than the charcoal trap samples in both pre- and post-filter bank. All radionuclides identified in the samples are presented in Table 2 in order of gamma-ray energies which were detected by the 145 cc coaxial Ge(Li) detector.

The millipore filter sample collected in the post-filter bank contained no radioactivity identifiable with the 10.2cmϕ NaI(Tl) detector, while the

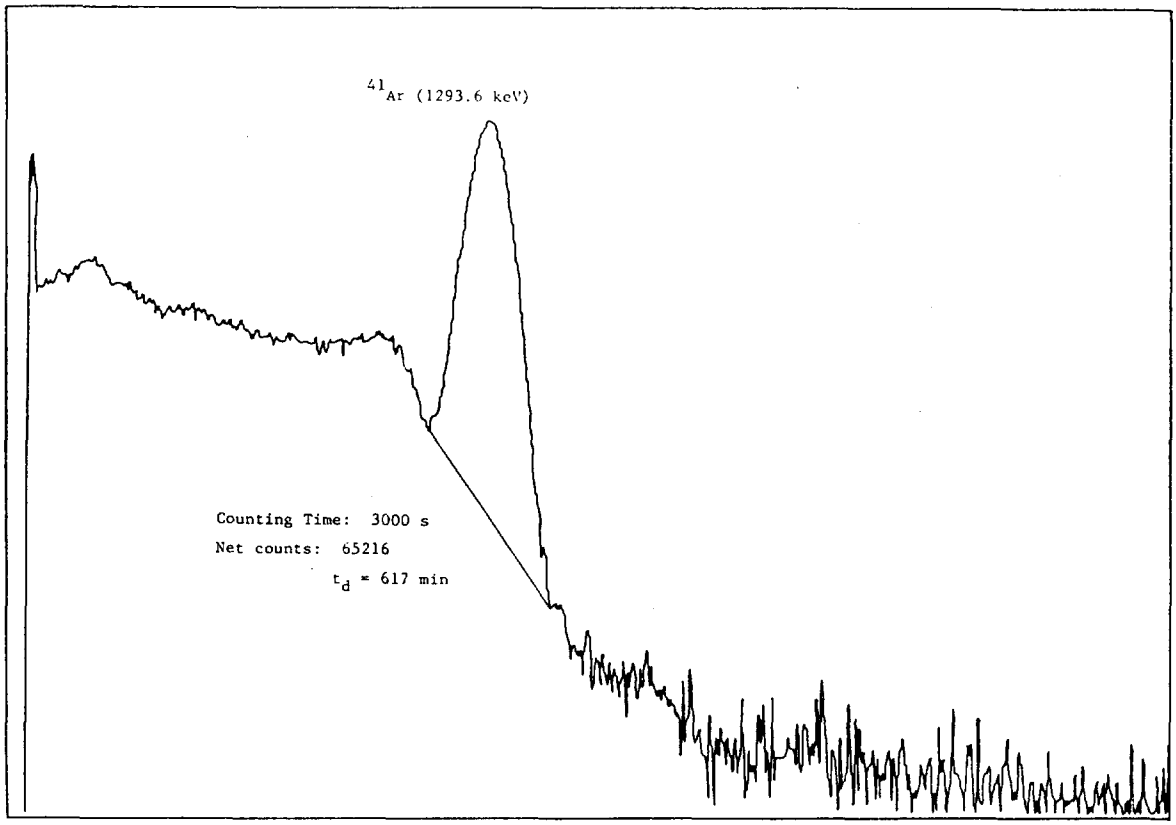

Figure 4. Gamma spectrum of air chamber from Pre-filter bank with 20.3cmϕ NaI(Tl) detector

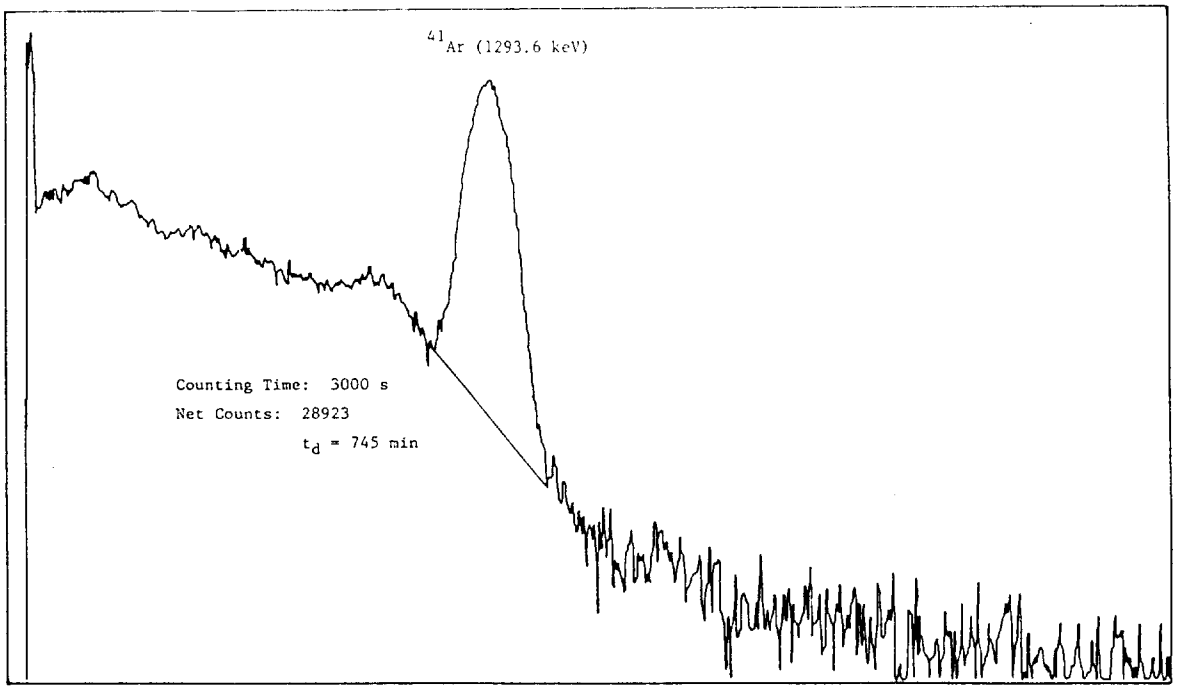

Figure 5. Gamma spectrum for air chamber from post-filter bank with 20.3cmϕ NaI(Tl) detector

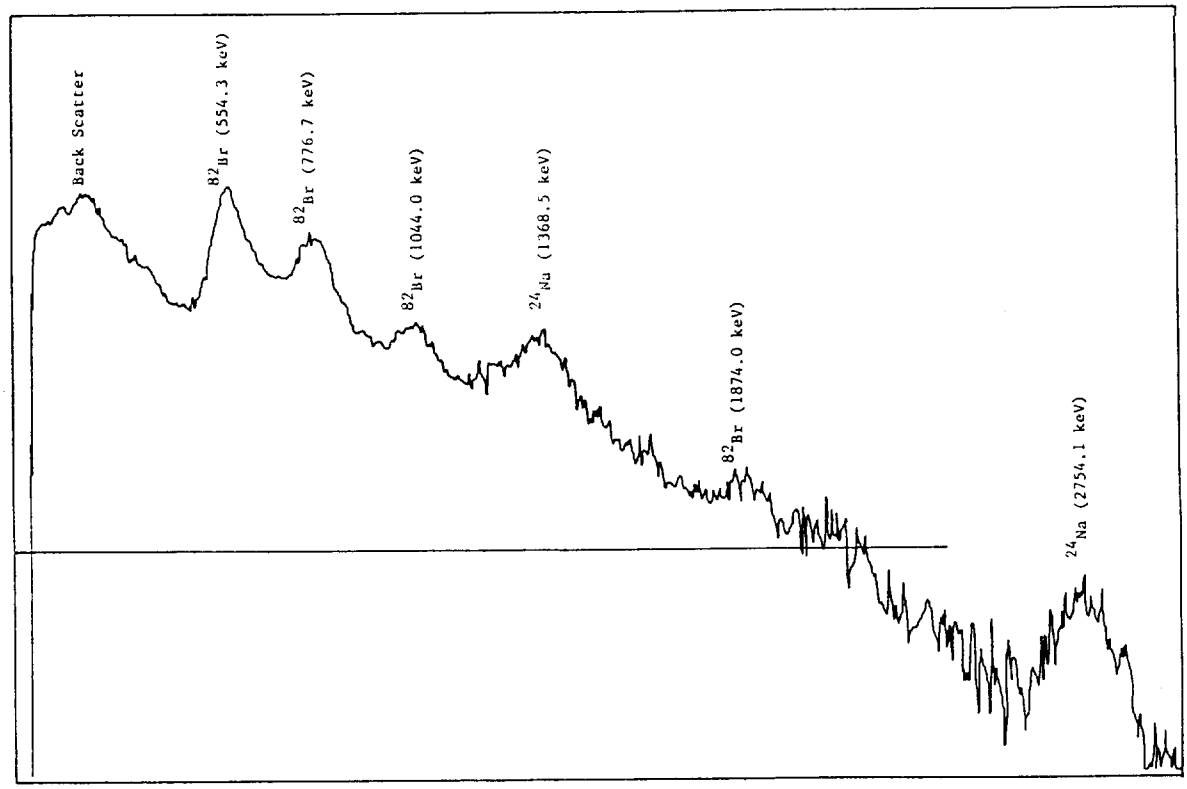

Figure 6. Gamma spectrum of millipore filter from pre-filter bank with 10.2cmϕ NaI(Tl) detector (t_d = 1045 min)

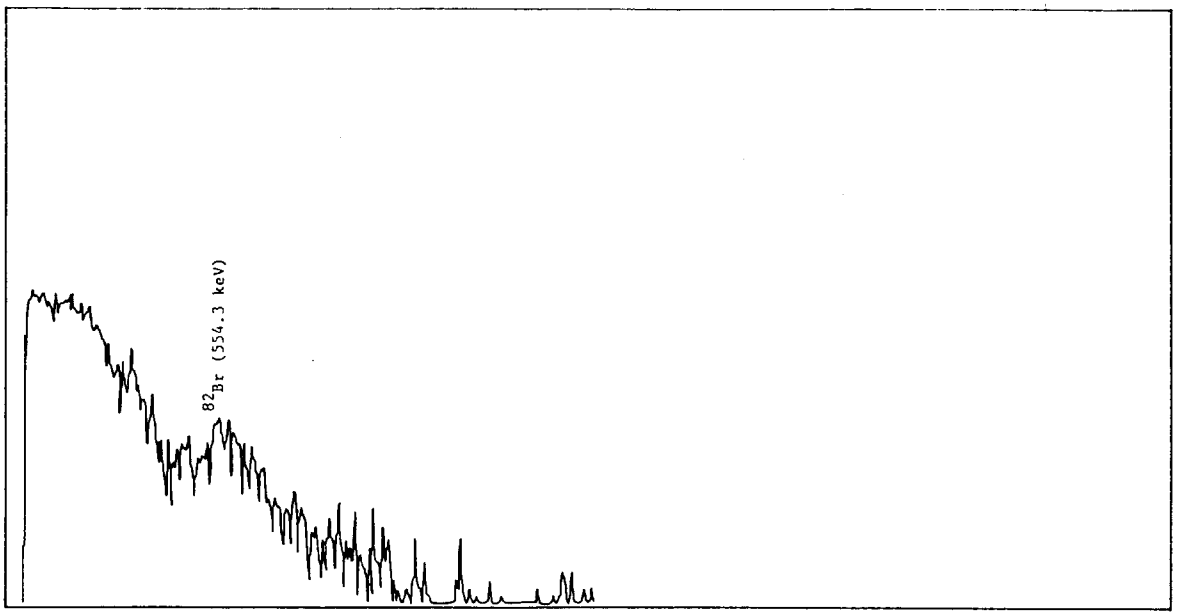

Figure 7. Gamma spectrum of millipore filter from post-filter bank with 10.2cmϕ NaI(Tl) detector (t_d = 580 min)

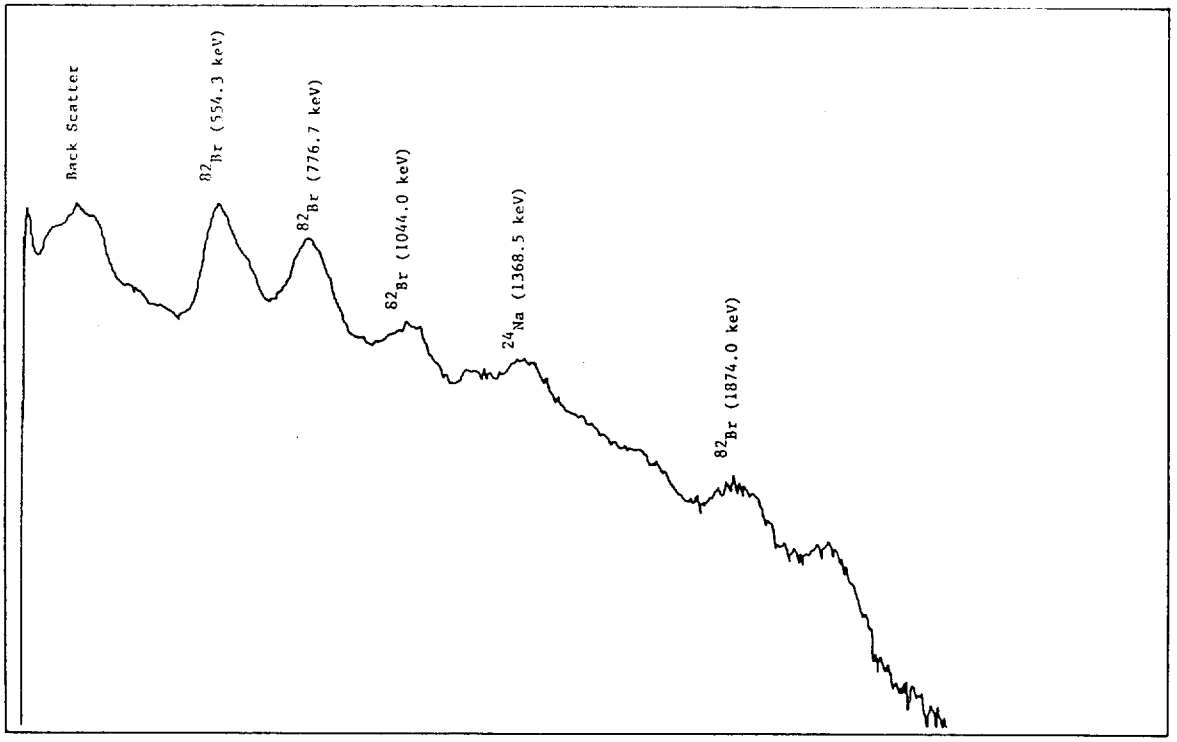

Figure 8. Gamma spectrum of charcoal trap from pre-filter bank with 10.2cmϕ NaI(Tl) detector (t_d = 953 min)

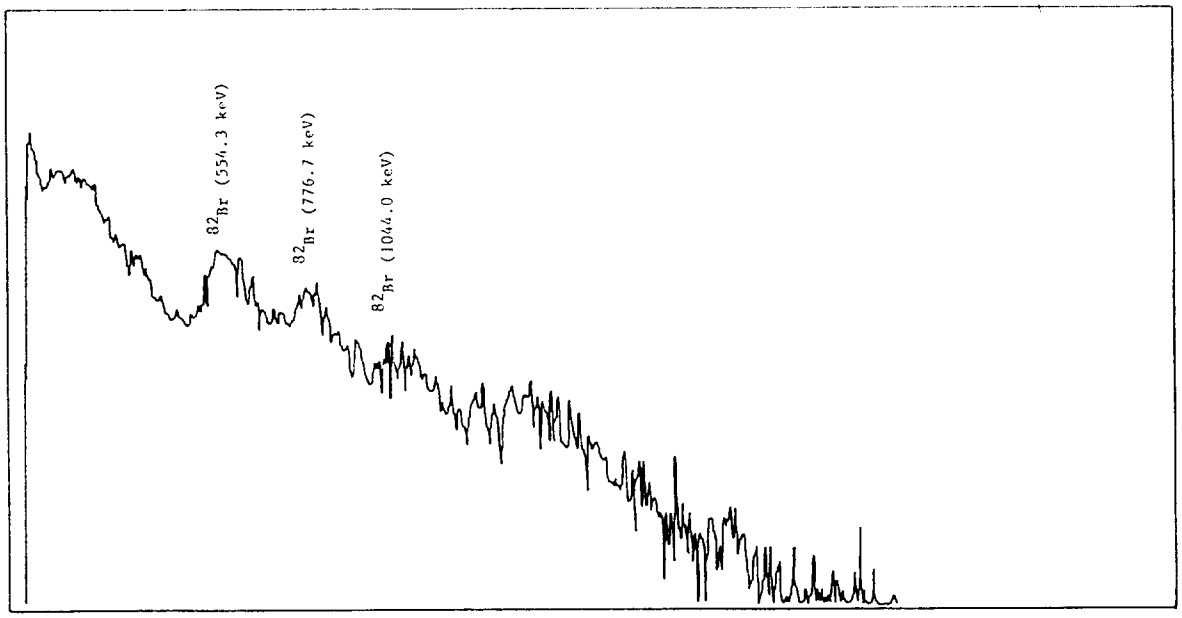

Figure 9. Gamma spectrum of charcoal trap from post-filter bank with 10.2cmϕ NaI(Tl) detector (t_d = 1049 min)

Ge(Li) detector picked out two peaks, ^{82}Br(0.553 MeV) and ^{106}Rh(0.513 MeV).

The air chamber samples from both pre- and post-filter bank contained only ^{41}Ar in large enough amounts to be positively identified as shown in Figures 4 and 5.

In most cases, the gamma spectra obtained with the 10.2cmϕ NaI(Tl) detector were not clear and rather complicated. The positive identification of radionuclides was made possible only with the Ge (Li) detector. However, not all peaks have been identified. Figures 6 through 9 show the bad resolution of NaI (Tl) detector.

The emission rate of radionuclides and the % removal efficiency of filter bank are described in Table 3.

Table 3. Emission rate and % removal efficiency

Nuclide	Emission Rate @ 3 MW (MBq / h)		% Removal Efficiency
	Pre-filter	Post-filter	
^{24}Na	8.10×10^{2}	0	100
^{38}Cl	2.04×10^{5}	4.40×10^{3}	97.84
^{41}Ar	2.57×10^{4}	2.20×10^{4}	14.27
^{72}Ga	5.70×10^{1}	0	100
^{82}Br	2.72×10^{4}	3.55×10^{2}	98.70
^{92}Sr	6.14×10^{2}	0	100
^{97}Zr	6.40×10^{1}	0	100
^{106}Rh	2.14×10^{4}	2.55×10^{2}	98.81
^{132}I	5.29×10^{2}	0	100
^{133}Te	2.02×10^{5}	0	100
^{141}Ce	1.10×10^{1}	0	100
^{153}Sm	2.89×10^{2}	0	100
^{154}Pm	–	0	100

DISCUSSION

The preliminary requirement of this study was that the reactor had to be at the 3 MW power level for about one hour before air sampling began. This was to assure stabilization of the reactor cooling air.

The 30 minute sampling time provided sufficient activity to identify the gamma peaks of components present in the sample.

^{154}Pm peak energy (1.659 MeV) presented in Table 2 may be considered to be interference peak with ^{82}Br which has a gamma energy of 1.654 MeV.

^{138}Cs, ^{133}Xe and ^{135}Xe reported in the previous studies [1,2] were not found in this study. The efficiency of the sampling train can be assumed to be 100 % because as shown in Figures 4 and 5, the air sampling chamber contains nearly 100 % ^{41}Ar activity. The energy calibration for Ge(Li) detector in the gamma spectral analyses was done by using the value, 1.376 keV/ch.

CONCLUSIONS

Radionuclides identified in the gaseous effluent from the Brookhaven Medical Research Reactor during the investigation in 1979 summer included the activated nuclides: ^{24}Na, ^{38}Cl, ^{41}Ar, ^{82}Br, and fission products: ^{72}Ga, ^{92}Sr, ^{97}Zr, ^{106}Rh, ^{132}I, ^{133}Te, ^{141}Ce, ^{153}Sm, and ^{154}Pm.

The results of this study provide the comparable information with the corresponding ones reported in the previous studies.[1,2]

The concentration of selected nuclides in the post-filter bank are as follows:

^{38}Cl: 4.18×10^{-1} Bq/cc
^{41}Ar: 2.09 Bq/cc
^{82}Br: 3.36×10^{-2} Bq/cc
^{106}Rh: 2.42×10^{-2} Bq/cc

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ABSTRACT

The gaseous effluents from the Brookhaven Medical Research Reactor (BMRR) was investigated. All gas samples were analyzed by the gamma-ray spectrometry. Among the radionuclides identified as those present in the samples, ^{82}Br ($T_{1/2}$ = 35.30 h) was the most predominent one. Other major components identified were ^{38}Cl($T_{1/2}$ = 37.24 min), ^{41}Ar ($T_{1/2}$ = 1.827 h), ^{106}Rh) ($T_{1/2}$ = 29.80 s), and ^{133}Te ($T_{1/2}$ = 12.45 min). ^{41}Ar concentration in gaseous effluent was calculated to be 2.436 Bq/cc in pre-filter bank at the reactor power of 3 MW. The emission rate of ^{41}Ar has been estimated to be 8.51×10^{9} Bq/MW-h. The % activity removal efficiency of filter bank was also estimated to be 97.84 % for ^{38}Cl, 14.15 % for ^{41}Ar, 98.70 % for ^{82}Br, and 98.81 % for ^{106}Rh. Other radionuclides identified in millipore filter and charcoal trap such as ^{24}Na, ^{72}Ga, ^{92}Sr, ^{97}Zr, ^{132}I, ^{133}Te, ^{141}Ce, ^{153}Sm, and ^{154}Pm were completely removed by the filter bank.