

AN AIR SAMPLING SYSTEM TO ESTIMATE CONCENTRATIONS
OF ARGON-41 IN THE V.P.I. PHYSICS BUILDING DURING
REACTOR OPERATION

by

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III. INTRODUCTION

The object of this thesis was to determine the A^{41} concentrations at different locations in the V.P.I. physics building during reactor operation under normal conditions. From these values a linear extrapolation was made to estimate the concentrations for higher power operation.

The V.P.I. nuclear reactor is designed for a maximum operating power of 10 kw. However, with a few alterations this reactor can be converted for higher power operation. Increased power involves an increase in the potential reactor hazards and a careful examination of all components and operating conditions is necessary. One of the more important considerations is the amount of radioactive argon (A^{41}) released to the surroundings.

A^{41} is the product of the following n- γ reaction,



The natural atmosphere consists of ~ 1%, by volume, A^{40} . Argonaut-type reactors have an open region between the top closures and the top of the core region which is exposed to a relatively high neutron flux. Since the reactor is not sealed, the activated air from this area may leak into the reactor room and into adjacent areas.

Due to the low concentrations of A^{41} present a very sensitive detection system is required, and a counting area where the background

radiation is independent of reactor operation.

The principle problem encountered in this investigation was the calibration of the counting system. Once the system was calibrated the air sampling technique was fast and straight forward. However, the activity of some of the samples was so low that long counting times were required.

IV. REVIEW OF LITERATURE

In the past few years the need for air monitoring systems has increased because of the increasing number of reactors going into operation. There have been relatively few reports published concerning this subject. A^{41} , a product of the (n,γ) reaction between the A^{40} in the atmosphere and a neutron, decays with the emission of -1.20 Mev β -particle and a 1.29 Mev γ -ray. Some detector systems are based on detection of the β -particle while others utilize the γ -ray.

Judd⁽¹⁾ used a gamma scintillation spectrometer to detect the γ -rays from A^{41} , and the gaseous fission products xenon and krypton. Four different sizes of NaI crystals were tested and because of the estimated geometry factor and photopeak efficiencies, the 3" x 3" proved to be the best choice for counting A^{41} . The calibration of his counting system was based on equilibration of a gas sample in a standard gas counting chamber used routinely on nuclear propelled naval craft and a one-gallon polyethylene gas sampling bottle. Also used were calculated efficiencies for cylindrical radiation detectors from Heath et al⁽²⁾. He used a "batch" gas sampling method in which the sample was collected in a bottle and taken to the counting area and analyzed. The detection limit of this system was $\sim 5 \times 10^{-8}$ $\mu\text{c/ml}$ for A^{41} corresponding to a count rate of ~ 10 cpm.

Ghatak⁽¹¹⁾, used a halogen-quenched G-M counter to detect the -1.20 Mev beta particle given off by decaying A^{41} . The G-M counter

was mounted in a ventilation duct and connected to a counting rate meter to give the activity of the air being expelled from the reactor system. For the particular counter and geometry used, a counting rate of 970 cpm corresponded to a concentration of approximately 2×10^{-6} $\mu\text{c/ml A}^{41}$ with an overall accuracy of 10%. His calibration was based on the activation of a cylinder of pure argon which was then transferred to a previously evacuated cylinder containing a G-M counter to be calibrated.

The air monitoring system used aboard nuclear powered ships⁽¹³⁾ utilizes a "batch" sampling method which has a G-M tube as the detecting element. A 100 cm^3 gas collection chamber fitted with two stop-cocks, a detachable flush-bulb and filter for the particulate activity is used to obtain the air samples. Several compressions and releases of the flush-bulb circulates the air to be sampled through the collection chamber. The chamber is then taken to the counting room to determine the A^{41} concentration of the air sampled.

Problems involved in calibrating cylindrical radiation detectors are examined in some detail by Heath⁽³⁾. He suggests the convenient quantity: "photo-efficiency" or "peak-to-total ratio" for determining the counting efficiency of cylindrical radiation detectors such as NaI(Tl). The peak-to-total ratio is defined as the fraction of the total events which fall in the photo-electric peak.

The maximum permissible concentrations of A^{41} for 40 hours in any seven consecutive days is 2×10^{-6} $\mu\text{c/ml}$ for a restricted area

and 4×10^{-8} $\mu\text{c/ml}$ in an unrestricted area⁽¹²⁾. A restricted area is defined as any area, access to which is controlled by an authorized person. Such small concentrations suggest the need for high sensitivities and long counting times.

V. THEORY

A. Activation Analysis

When a sample is exposed to a neutron flux, radioactive atoms will be produced by the capture of neutrons. If there are N' radioactive atoms present at time t' and the production rate is P , then the rate of change of the number of radioactive atoms is

$$\frac{dN'}{dt'} = P - \lambda N' \quad (1)$$

where, λ = decay constant for the induced activity (A^{41})

$$P = N\sigma_a \phi$$

N = total number of atoms (A^{40}) in the sample

σ_a = neutron activation cross-section

ϕ = neutron flux in neutrons/cm²-sec

If one assumes that no radioactive atoms are present before exposure, then integration of equation (1) yields the activity, A_0 , present after an exposure time, t_e ;

$$A_0 = \lambda N' = N \sigma_a \phi [1 - e^{-\lambda t_e}] \quad (2)$$

A_0 = absolute activity at the end of the exposure, in disintegrations per second (dps)

$(1 - e^{-\lambda t_e})$ = build-up term to account for decay during exposure

If the exposure time is long compared with the half-life of the radioactive isotope the build-up term becomes unity and A_0 approaches the

saturation activity, A_s :

$$A_s = N \sigma_a \phi \quad (3)$$

Rearranging the saturated activity,

$$A_s = \frac{A_0}{[1 - e^{-\lambda t_e}]} \quad (4)$$

Since the sample cannot be counted at the same time it is removed from the neutron flux a term must be added to account for the exponential decay during the waiting time. Hence, the source strength, A , at any time, t , after exposure, is given by

$$A = A_0 e^{-\lambda t} \quad (5)$$

The total number of counts, C , observed in the counting time, t_c , after waiting a time, t_w , before counting, is mathematically equivalent to integrating the activity over the time, $t = t_w$ to $t = t_w + t_c$:

$$C = \int_{t_w}^{t_w + t_c} A dt = \int_{t_w}^{t_w + t_c} A_0 e^{-\lambda t} dt$$
$$C = \frac{A_0}{\lambda} e^{-\lambda t_w} [1 - e^{-\lambda t_c}] \quad (6)$$

Rewriting this expression in terms of saturated activity;

$$C = \frac{A_s}{\lambda} [1 - e^{-\lambda t_e}] e^{-\lambda t_w} [1 - e^{-\lambda t_c}] \quad (7)$$

$$A_s = \frac{\lambda C}{[1 - e^{-\lambda t_e}] e^{-\lambda t_w} [1 - e^{-\lambda t_c}]} \quad (8)$$

where,

C = number of counts, corrected for background recorded
in time t_c

t_w = waiting time

t_c = counting time

t_e = exposure time

If the sample is counted for a period of time long in comparison with its half-life then the relation between the number of counts observed and the source strength is given by Eq. 8. In the case when the counting time is much less than the half-life ($t_c \ll T_{1/2}$) of the sample, the amount of decay during the counting time is negligible and the last equation can be simplified:

$$A_s = \frac{R}{[1 - e^{-\lambda t_e}] e^{-\lambda t_w}} \quad (9)$$

where R is the corrected counting rate obtained by dividing corrected number of counts by the counting time.

B. Absolute Counting Efficiency

The detection efficiency of a solid phosphor (NaI) may be calculated with a high degree of precision if one knows the cross-sections of NaI, for γ -rays of a given energy and source-detector geometry. Vegors, Marsden and Heath⁽²⁾ have made calculations for point and disk sources located on the central axis of a cylindrical detector. When the detector efficiency for a particular detector and source-detector geometry is known the emission rate from the source can be calculated from the data obtained on the scintillation spectrometer. If the spectrum due to background radiation is subtracted from the total spectrum then integration under the resultant pulse height distribution will yield the total number of photons from the sample detected by the crystal. If a multichannel pulse height analyzer is used, then the integration can be obtained by simply adding the individual channel contributions.

In many cases it is more convenient to relate the source strength to the number of events under the photo-peak by introducing the quantity; the "peak-to-total ratio" or "photo-efficiency". The area under the photo-peak is defined in terms of a symmetrical "gaussian" shape, fit to the high energy side of the peak, as shown in Figure 1. This fraction P , is the ratio of the number of events under the peak to the total number in the spectrum. With this convention the emission rate of a single gamma-ray is given by:

$$N_0 = \frac{N_p}{\epsilon a P} \quad (10)$$

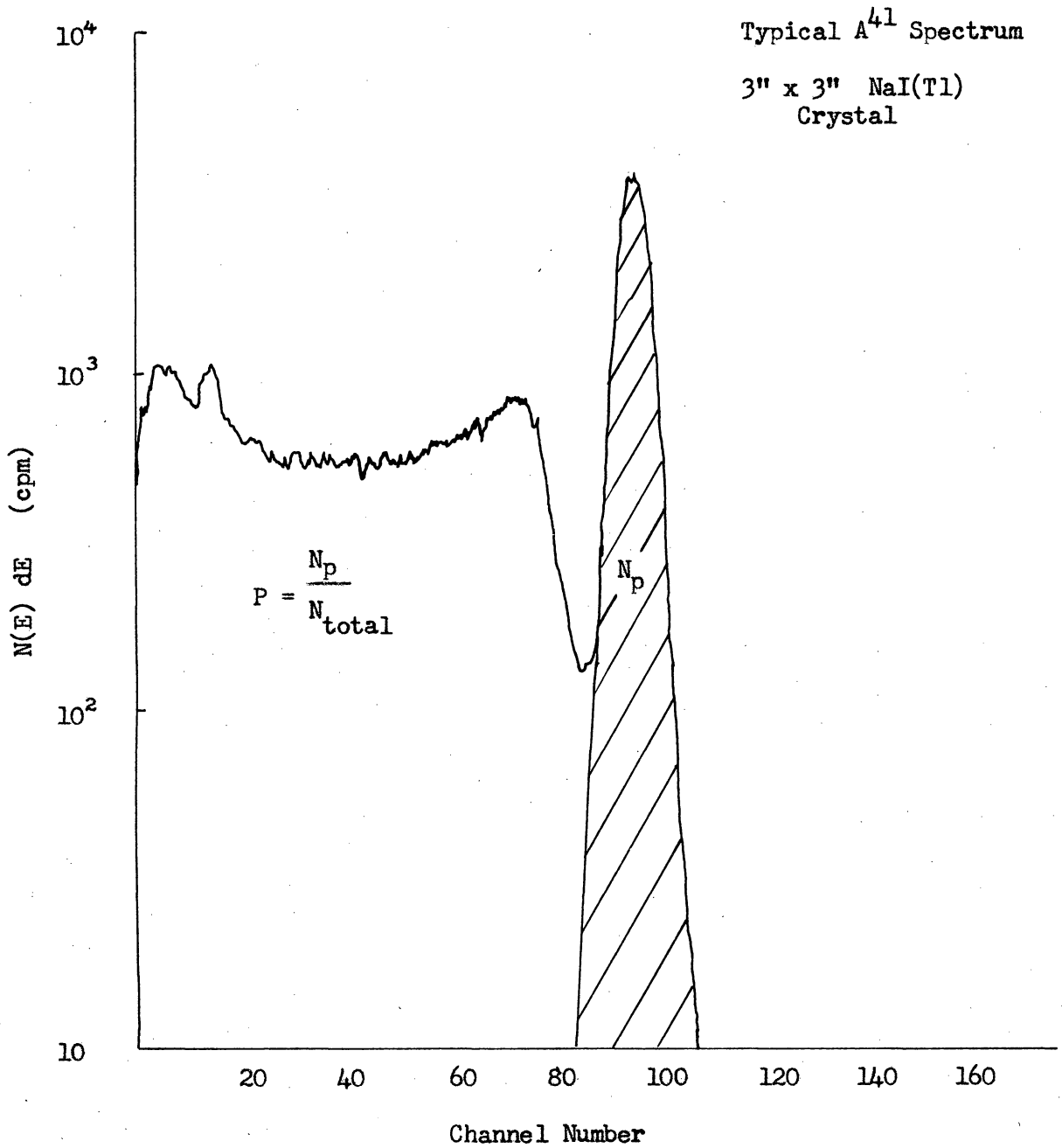


FIGURE 1 ILLUSTRATION OF PEAK-TO-TOTAL RATIO

where

N_o = the number of gamma rays emitted per unit time, (dpm)

N_p = the number of events registered under the photo-peak per unit time, (cpm)

P = peak-to-total ratio for the particular isotope.

ϵ = absolute detection efficiency

a = correction factor for self absorption (source, absorber, etc.)

If one considers a standard source having a disintegration rate A , counted on a gamma scintillation spectrometer, and N_p events per unit time registered under the photopeak, then equation (10) can be written to give the counting efficiency:

$$\epsilon_s = \frac{N_p}{A} \quad (11)$$

Note that equation (11) is for a particular detector, source-detector geometry and radionuclide. Then ϵ_s is the probability that a photon emitted will undergo a photo-electric interaction. Hence, if a sample of the same material is counted under the same conditions as the calibration source, then by knowing the counting rate under the photopeak the emission rate can be determined.

In order to relate the counting efficiency of one bottle to that of another bottle, where both contain the same concentration of radioactive material, say A^{41} , the following expression is used:

$$\epsilon_2 = \frac{N_{p2} V_1}{N_{p1} V_2} \epsilon_1 \quad (12)$$

where,

V = volume of the bottle

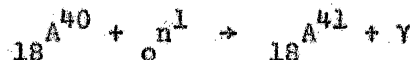
N_p = counting rate under photopeak

ϵ = counting efficiency

and subscripts indicate the bottle number.

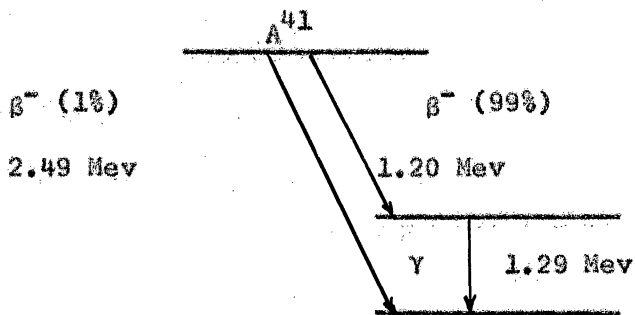
C. Activation of A^{41}

Normal air consists of approximately 1% by volume A^{40} . If A^{40} captures a neutron, radioactive A^{41} is formed in the following reaction;



The release of A^{41} is generally a problem around research reactors utilizing beam ports, pneumatic tubes, and etc.

A^{41} decays with a half-life of 110 minutes in the following scheme,



VI. EXPERIMENT

A. Apparatus

The apparatus used in this experiment consisted of a gamma-scintillation spectrometer and polyethylene bottles. Two small bottles (525 ml) containing normal air were irradiated in a known neutron flux⁽¹⁴⁾ and used as calibration sources. A one gallon bottle with a well sealed in the bottom was used to collect the air samples.

Pulses from the detector were amplified, sorted and stored according to pulse height by a 512-channel analyzer. Components making up the spectrometer are shown in Figure 2, and are as follows:

(1) A 3" x 3" cylindrical NaI(Tl) crystal was used to detect the 1.29 Mev γ -ray emitted by A^{41} . The choice of the size crystal to be used was determined by similar measurements made by Judd⁽¹⁾ and Heath et al⁽³⁾ who found the 3" x 3" crystal most suitable for detection of A^{41} . With the crystal mounted on a DuMont 6363 photomultiplier tube, 6% resolution for the Cs^{137} photopeak was obtained. The signal from the phototube is fed directly into a cathode follower which shapes the pulses and matches the impedance of the signal to that of the transmission cable.

(2) A Nuclear Data 512-channel pulse height analyzer was used to amplify, sort and store the pulses from the detector. It was more convenient to use only 256 channels for all measurements because of the extra time involved in typing the full 512 channels.



FIGURE 2. SPECTROMETER COMPONENTS

(3) A Hammer model N401 high voltage power supply provided a steady positive voltage for the phototube (750 V).

B. Sampling

The air sampling bottle was a one gallon polyethylene jar with a well sealed in the bottom large enough to fit over the 3" x 3" NaI(Tl) crystal. The cylindrical well approximately 3 3/4" x 3 3/4" was made from a portion of a quart polyethylene bottle. The walls of the well were approximately the same thickness as the standard bottle (-0.040 in).

The sampling procedure was extremely rapid and straight forward. The air sampling bottle was filled with water before entering the area to be sampled. By pouring the water from the bottle, the bottle was filled with air representative of that area. The bottle was then taken to the spectrometer to be counted. Figure 3 shows the air sampling bottle in counting position and an end view of a second sampling bottle.

After each sample was counted, the sampling bottle was then rinsed with ordinary water to remove any traces of the previous activity.

Measurements were made to determine the amount of water that adsorbed onto the walls of the bottle during sampling and the results showed that less than 0.1% of the volume of the bottle was taken up by the remaining water. So no correction was made for this.

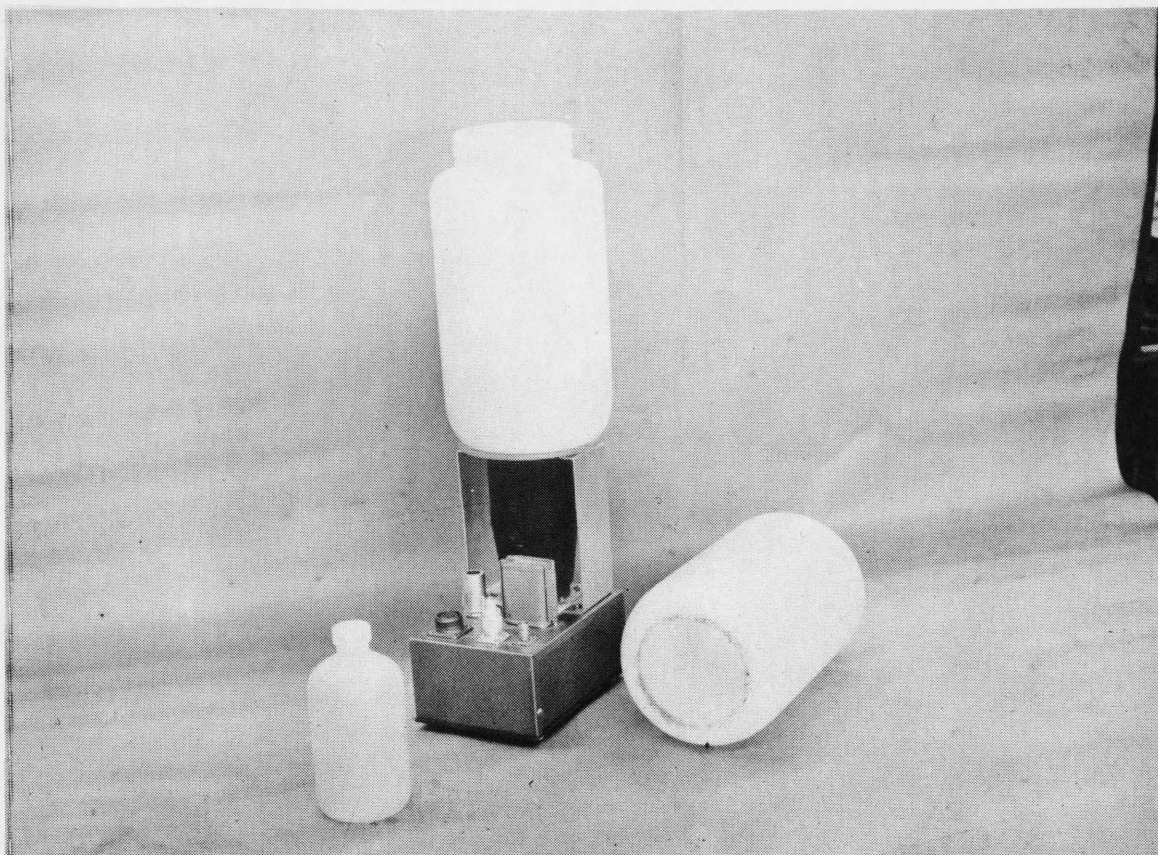


FIGURE 3. AIR SAMPLING BOTTLE IN COUNTING POSITION

C. Results

1. Calibration Constant for Standard Bottle

After irradiation, the polyethylene bottle to be used as the calibration source became slightly activated. Hence, two identical bottles were irradiated simultaneously and the active air was flushed from one. By subtracting the activity due to the bottle itself, the counting rate under the A^{41} photo-peak due to the A^{41} alone was determined. It was found for short exposure periods (30 min) that ~5% of the activity under the photo-peak was due to the activated polyethylene.

The two bottles were exposed to a flux of 1.28×10^{11} n/cm² sec⁽¹⁴⁾ for 30 minutes in the central stringer of the reactor. The source strength, A'_0 , at time zero after exposure was calculated by using equation (2) and found to be

$$A'_0 = 9.448 \pm 0.092 \times 10^7 \text{ dpm.}$$

After irradiation the calibration sample was taken to the gamma-scintillation spectrometer to be counted. The standard bottle was placed directly on top of the crystal for a one minute count and then the second bottle was counted for the same time to subtract the activity due to the activated bottle. From the counting rate under the photo-peak corresponding to the 1.29 Mev γ -ray emitted, and the above calculated source strength, a calibration constant, ϵ_s , for the standard bottle was determined. This constant, ϵ_s , is

obtained by combining equations (5) and (11) and is written in the following form,

$$\epsilon_s = \frac{N_p}{A'_o e^{-\lambda t_w}} \quad (13)$$

The results of the calculations are recorded in Tables 1 and 2. The two bottles did not receive the same amount of induced activity when irradiated. The results of two identical exposures in which the two bottles were alternated showed that the ratio of the two activities was 1.066, where bottle #1 received the most activity. Hence, the values listed in Table 1 must be multiplied by 1.066 to normalize both sets of data to the same source strength. A final value for the calibration constant was obtained to be,

$$\epsilon_s = 0.01319 \pm 0.00016$$

2. Determination of Absolute Counting Efficiency

After the calibration constant for the standard bottle was calculated, this value had to be related to an absolute detector efficiency for the source-detector geometry to be maintained by the one-gallon air-sampling bottle. The absolute calibration was based on the equilibration of a gas sample in the standard bottle and the one gallon air sampling bottle. Air pumped from the core region of the reactor served as a source of A^{41} and was circulated through the bottles until equal distribution was reached. The bottles were then counted for one minute intervals and the number

TABLE 1
 CALIBRATION CONSTANT FOR STANDARD BOTTLE
 (Bottle 2)

Set	t_w (min)	$e^{-\lambda t_w}$	$N_p \times 10^{-5}$ (cpm)	$A'_0 \times 10^{-7}$ (dpm)	ϵ_{S_2} $\times 10^2$
1	181	0.31956	3.613	9.448 ± 0.092	1.200
3	189	0.30420	3.449	↓	1.200
4	210	0.26634	3.093		1.233
6	220	0.25000	2.863		1.249
8	240	0.22050	2.520		1.213
10	360	0.10352	1.231		1.262
11	375	0.09419	1.125		1.258
12	385	0.08844	1.060		1.273

TABLE 2
CALIBRATION CONSTANT FOR STANDARD BOTTLE
(Bottle 1)

Set	t_w (min)	$e^{-\lambda t_w}$	$N_p \times 10^{-4}$ (cpm)	$A_0' \times 10^{-7}$ (dpm)	ϵ_{s_2} $\times 10^2$
6	450	0.0592	7.328	9.448 ± 0.092	1.314
7	460	0.0551	6.887	↓	1.327
9	470	0.0523	6.463		1.312
10	475	0.0501	6.270		1.330

of events per unit time under the photo-peak for each bottle was determined. This procedure was repeated several times to insure uniformity of distribution. The ratio (α) of the activity of the air sampling bottle to that of the standard bottle was calculated using the following equation:

$$\alpha = \frac{N_{pas}}{N_{ps}} \quad (14)$$

where,

N_{pas} = Counting rate under photo-peak for the air sampling bottle

N_{ps} = Counting rate under photo-peak for the standard bottle

The results of this calculation are listed in Table 3, and the average value was found to be

$$\alpha = 6.345 \pm 0.05$$

The relationship between the calibration constant and the absolute counting efficiency is given by the combination of equations (12) and (14):

$$E_t = \frac{\alpha V_s E_s}{V_{as}} \quad (15)$$

where,

V_s = Volume of standard bottle (525 ml)

V_{as} = Volume of air sampling bottle (3600 ml)

and was calculated to be,

TABLE 3

RATIO OF ACTIVITY IN AIR SAMPLING BOTTLE TO STANDARD BOTTLE

Set	Bottle	t_w (min)	$e^{-\lambda t_w}$	$N_p \times 10^{-4}$ (cpm)	$N_{p0} \times 10^{-4}$ (cpm)	α
5	S	0.000	1.000	5.591	5.591	
6	AS	3.375	0.979	34.785	35.531	6.355
7	AS	0.000	1.000	32.603	32.603	
8	S	2.500	0.984	5.017	5.097	6.396
Recharged the Bottles						
10	S	0.000	1.000	6.723	6.723	
11	AS	2.625	0.984	41.657	42.352	6.300
12	AS	0.000	1.000	38.904	38.904	
13	S	2.334	0.985	6.070	6.160	6.316
Recharged the Bottles						
15	S	0.000	1.000	6.580	6.580	
16	AS	2.625	0.984	40.920	41.601	6.322
17	AS	0.000	1.000	39.212	39.212	
18	S	2.709	0.983	6.036	6.140	6.383

Note: S refers to the standard bottle

AS refers to the air sampling bottle

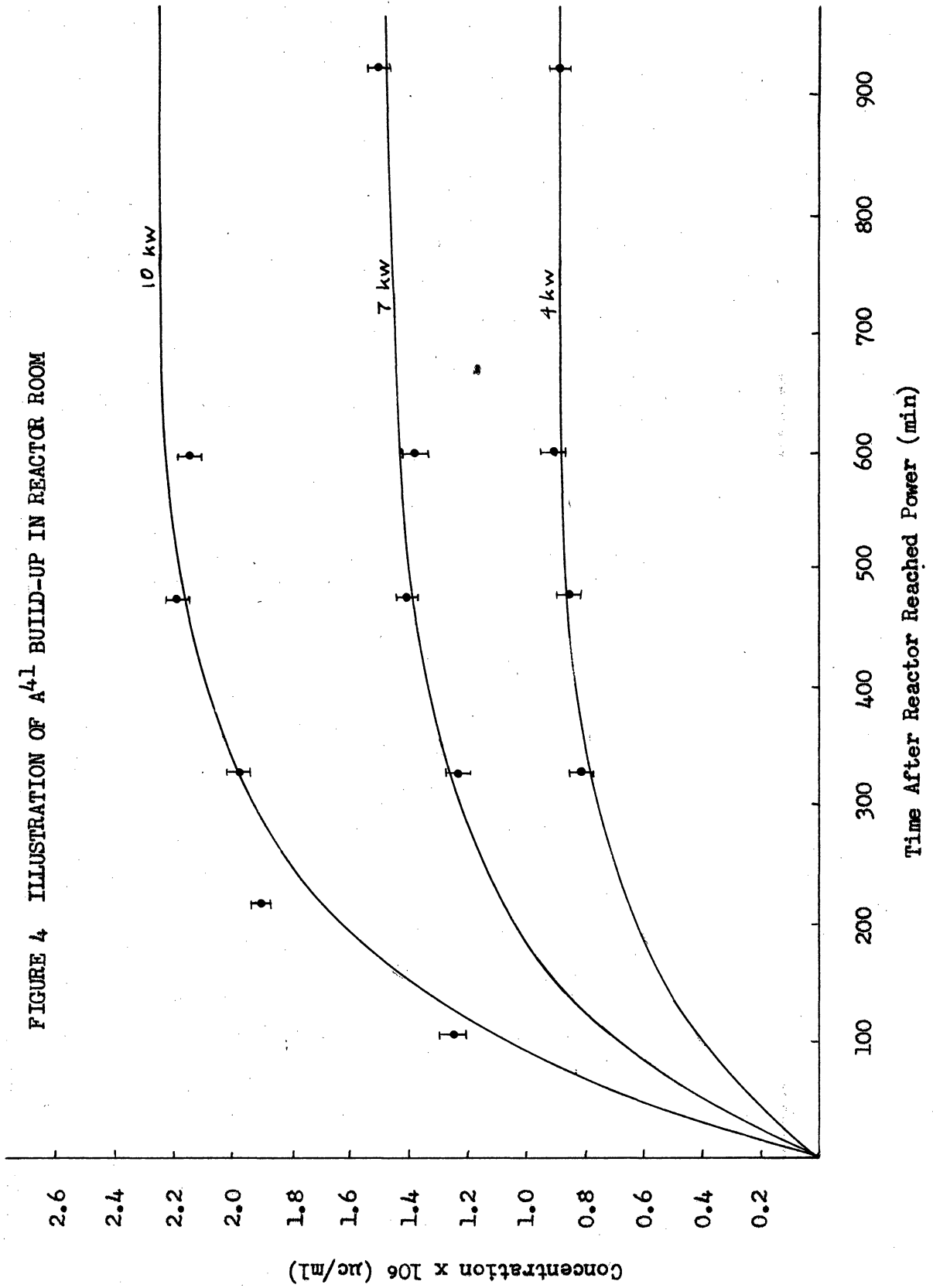
$$\epsilon_t = 0.01218 \pm 0.000063$$

3. Analysis of Air-Samples

The variation in A^{41} concentration with reactor power was determined for several different locations in the physics building. The reactor was operated at 4, 7, and 10 kw under normal operating conditions. Air samples were taken in the reactor room periodically to determine approximately when A^{41} reached secular equilibrium. Figure 4 illustrates the exponential build-up for different power levels. The solid curves are based on theoretical values.

After the A^{41} had reached equilibrium, air samples from other rooms were taken and analyzed. All of the samples were allowed to decay for the same length of time before counting. Because some of the samples contained very small amounts of the radioactive gas, a long counting time was required to give the statistical accuracy needed. To make the necessary corrections uniform, a waiting time of four minutes and a counting time of 40 minutes was maintained for all samples. Since some of the samples were very weak it was necessary to subtract two numbers very nearly the same magnitude to determine the A^{41} contribution. For this reason very long background counts were taken before each reactor run in order to reduce the statistical error in the background to a minimum. The total corrected number of events recorded under the photo-peak during the 40 minute counting time was R_p . The concentration of

FIGURE 4 ILLUSTRATION OF A^{41} BUILD-UP IN REACTOR ROOM



A⁴¹ in the air samples was calculated utilizing the following expression,

$$C_o = \frac{A_s}{\epsilon_t V_{as} (2.22 \times 10^6)} \quad (16)$$

where A_s is defined by equation (8). Rewriting this expression using equation (8) and setting C = R_p, the concentration is then defined by,

$$C_o = \frac{\lambda R_p}{[1 - e^{-\lambda t_c}] e^{-\lambda t_w} [\epsilon_t V_{as} 2.22 \times 10^6]} \quad (17)$$

Since all parameters are held constant except R_p, a new factor, f, can be defined:

$$f = \frac{1}{[1 - e^{-\lambda t_c}] e^{-\lambda t_w} [\epsilon_t V_{as} 2.22 \times 10^6]} \quad (18)$$

When the appropriate parameters were inserted and the necessary calculations were made this factor was determined to be

$$f = 2.98 \pm 0.01 \times 10^{-10}$$

Hence,

$$C_o = 2.98 \pm 0.01 \times 10^{-10} R_p \text{ (}\mu\text{c/ml)}$$

The results of the calculations are recorded in Tables 4, 5, 6, 7, and 8. Note that all error due to statistical variations are reported in terms of the standard deviation.

TABLE 4
 AIR SAMPLES FROM REACTOR ROOM
 (Restricted Area)

Power (kw)	Time (min)	R _p (counts)	f x 10 ¹⁰	Concentration x 10 ⁶ (uc/ml)	Per Cent of M.P.C.	
10	110	4172±55	2.98	1.244±0.017	62.2	
10	223	6300±68	↓	1.878±0.020	93.6	
10	330	6602±68		1.969±0.020	98.5	
10	482	7292±67		2.173±0.020	108.7	
10	600	7118±67		2.122±0.020	106.1	
7	330	4162±56		1.240±0.017	62.0	
7	482	4659±58		1.389±0.018	69.5	
7	600	4569±58		1.362±0.018	68.1	
7	930	4976±60		1.484±0.019	74.2	
4	330	2736±50		0.815±0.015	40.8	
4	482	2825±50		0.842±0.015	42.1	
4	600	3016±51		0.899±0.015	44.9	
4	936	2963±51		0.883±0.015	44.2	
10 *	330	54±40			BDL	
10 *	486	87±99			BDL	

BDL = Below Detection Limit
 time = time after reactor reached power
 * core purge experiment

TABLE 5
AIR SAMPLES FROM REACTOR CONTROL ROOM
(Restricted Area)

Power (kw)	Time (min)	R _p (counts)	f x 10 ¹⁰	Concentration x 10 ⁶ (uc/ml)	Per Cent of M.P.C.
10	660	1580±43	2.98	0.471±0.013	23.6
10	866	1510±43	↓	0.451±0.013	22.6
7	660	1842±46		0.549±0.014	27.5
7	880	750±40		0.224±0.012	11.2
4	660	562±36		0.168±0.010	8.4
4	985	568±36		0.169±0.010	8.4

TABLE 6
AIR SAMPLES FROM ACCELERATOR LABORATORY
(Unrestricted Area)

Power (kw)	Time (min)	R _p (counts)	f x 10 ¹⁰	Concentration x 10 ⁸ (uc/ml)	Per Cent of M.P.C.
10	755	247±37	2.98	7.364±1.103	184.1
7	755	395±38	↓	11.780±1.133	294.5
7	990	158±36		4.710±1.073	117.8
4	760	147±37		4.383±1.103	109.6
4	1039	66±35		1.967±1.103	49.2

TABLE 7
AIR SAMPLES FROM AUDITORIUM**
(Unrestricted Area)

Power (kw)	Time (min)	R _p (counts)	f x 10 ¹⁰	Concentration x 10 ⁸ (uc/ml)	Per Cent of M.P.C.
10	915	256±37	2.98	7.631±1.103	184.0
10	964	81±36	↓	2.414±1.073	60.4
7	705	1±35		BDL	
4	710	9±35		BDL	
10 *	758	365±99		↓	10.88±2.98

* Core Purge Experiment

TABLE 8
AIR SAMPLES FROM CLASS ROOM (Rm 204) **
(Unrestricted Area)

Power (kw)	Time (min)	R _p (counts)	f x 10 ¹⁰	Concentration x 10 ⁸ (uc/ml)	Per Cent of M.P.C.
10	815	86±37	2.98	2.593±1.073	64.8
10	1012	439±38	↓	13.087±1.113	327.2
7	805	127±37		3.786±1.103	94.7
7	1045	- 8±35		BDL	
4	876	- 1±35		↓	BDL

** The air intake for the auditorium and the windows in Room 204 are located downwind from the exhaust vent of the reactor room. The worst conditions were simulated by closing all doors, operating the ventilation system in the auditorium and opening windows in Room 204.

The maximum permissible concentrations (M.P.C.) for continuous exposure to A^{41} for 40 hours in any seven consecutive days are 2×10^{-6} $\mu\text{c/ml}$ for a restricted area and 4×10^{-8} $\mu\text{c/ml}$ for an unrestricted area. Since the reactor room and reactor control room are classified as restricted areas, the first value is used in calculating the per cent M.P.C. for the locations. The latter value is used for all other areas sampled. The variation of per cent M.P.C. with reactor power for three areas sampled is shown in Figures 5, 6, and 7.

During the first few hours of the 7 kw reactor run it was necessary for the reactor room door to be opened and closed frequently. Figure 5 illustrates how the concentration in the reactor room is decreased by opening and closing the door. Figures 6 and 7 illustrate how the A^{41} concentration is increased in the control room and the adjacent Accelerator Laboratory by this same effect.

The one high value obtained for the accelerator laboratory during the 4 kw run can be explained by the fact that an auxiliary room air conditioner was on during this run. This air conditioner pulls air in from the hall-way directly across from the reactor room and hence, there was an abnormal flow rate of air into this room. This condition did not prevail during other runs.

FIGURE 5 PER CENT OF M.P.C. VERSUS REACTOR POWER
(Reactor Room)

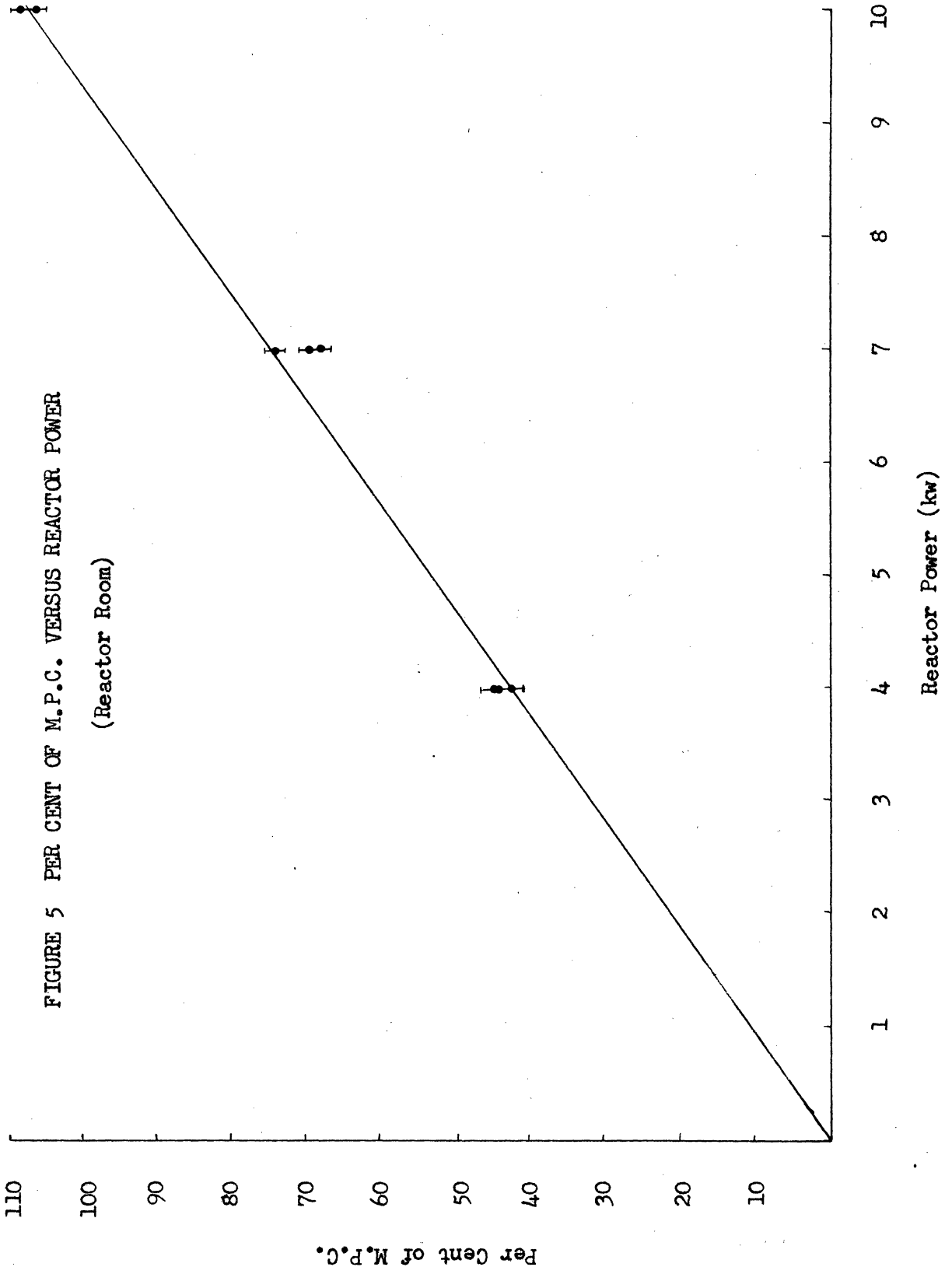


FIGURE 6 PER CENT OF M.P.C. VERSUS REACTOR POWER
(Reactor Control Room)

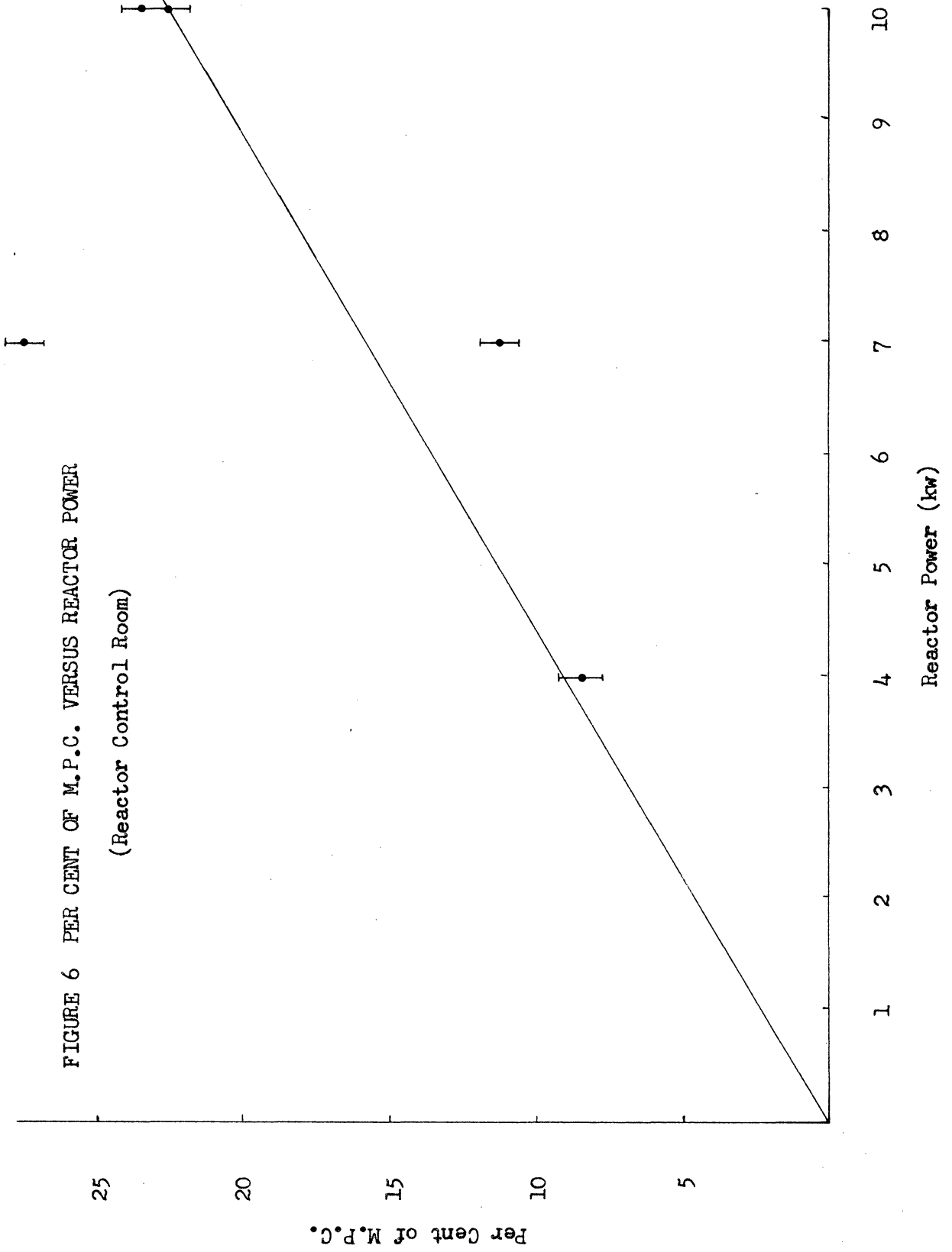
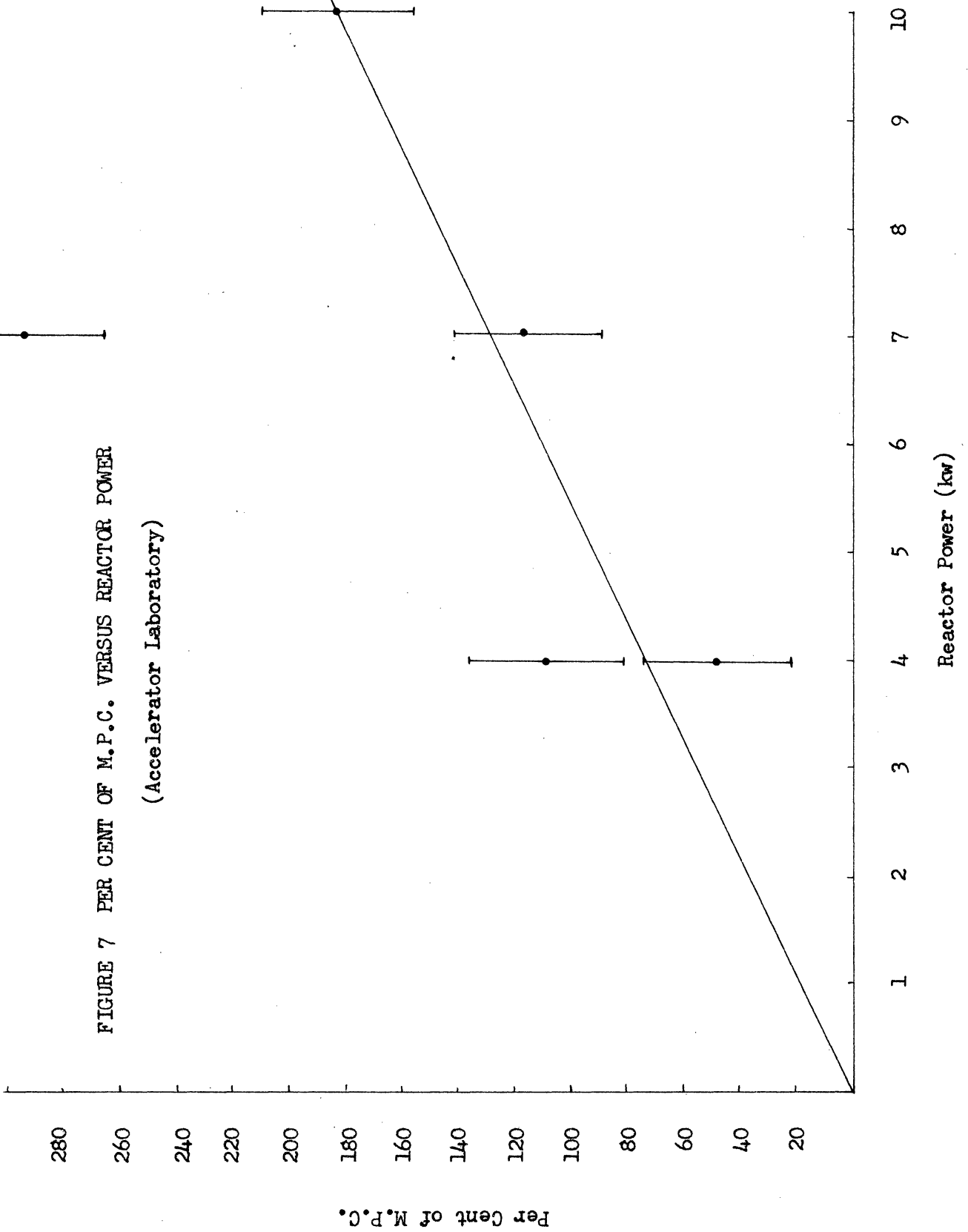


FIGURE 7 PER CENT OF M.P.C. VERSUS REACTOR POWER
(Accelerator Laboratory)



4. Core Purge Experiment

Under the present operating conditions, at full power the A^{41} concentrations are very close to the maximum permissible for a 40 hour week. Hence, if the maximum operating power level is increased by a factor of 10 then some system may be required to remove the activated air from the building. One method is to purge the core region and discharge the air through a ventilation stack. When the air leaves the stack it is quickly diluted and dispersed to the surrounding atmosphere.

A run was conducted in which the air from the core region was pumped from the top central access port and discharged through the ventilation window. The air pumping system is shown in Figure 8. The centrifugal pump (United Blower Co., model 8-P) had a capacity of approximately 2500 ft³/min.

The results of this run were not entirely satisfactory because of unusual fluctuations in the background radiation. It was necessary to use 40 minute background readings to approximate the true background at any time. This shorter background period increased the error in the final results. Nevertheless, it was shown that the A^{41} concentration was decreased by a factor greater than 10 by purging the core. Because the concentrations in other locations were very near the detection limit of the equipment no definite evidence was found of a change in concentration for those areas. The results are tabulated in Tables 4 and 7.

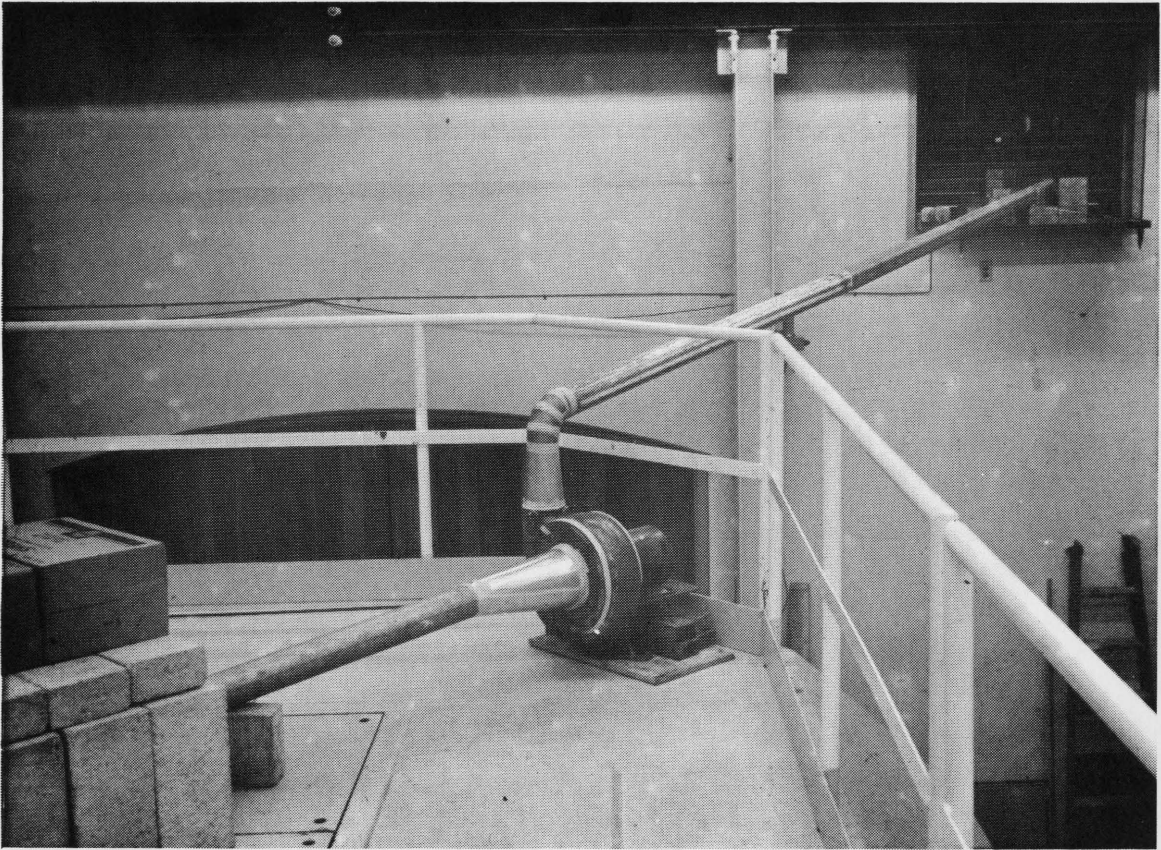


FIGURE 8. PHOTOGRAPH OF AIR PUMPING SYSTEM USED TO
PURGE THE CORE

5. Detection Limit

The main purpose of an experiment of this type is to produce a counting system which will have a very low detection limit. The detection limit will depend upon a number of factors including:

(1) Counter efficiency, (2) Abundance of the isotope in the sample, and (3) Portion of the spectrum utilized.

The detection limit was determined by the number of counts under the photo-peak during the counting time. A concentration of 4×10^{-8} ($\mu\text{c/ml}$) corresponded to -135 counts under the photo-peak.

6. Estimate of Concentrations at 100 kw

Estimates are made for the reactor room and a second floor classroom (room 204) only, since these two areas had the highest concentration in a restricted area and an unrestricted area respectively. The maximum measured concentrations are extrapolated linearly to estimate the maximum concentration to be expected for 100 kw operation. These values are then related to the limited operating time for this reactor. A table of maximum permissible concentrations (M.P.C.) for different continuous exposure periods follows.

From Figures 2 and 3 the maximum concentration to be expected in the reactor room during 100 kw operation would be 25×10^{-6} $\mu\text{c/ml}$. However, after five hours of operation the concentration will have

reached only 85% of the maximum value, i.e., a concentration of -21.3×10^{-6} $\mu\text{c/ml}$. From this last value and Table 9 the A^{41} concentration in the reactor room would be below M.P.C. if the reactor were operated continuously at 100 kw for a period of four hours. An operating period of -10 hours is required for the concentration to reach its maximum value and most experiments are for a fraction of this time. Table 10 shows the per cent of maximum concentration in the room for different exposure periods. The results of the unrestricted areas were so near the detection limit of the equipment that a uniform curve could not be obtained for the A^{41} build-up in these areas. Hence, the estimates are based on a factor of 10 increase over the maximum value -13×10^{-6} $\mu\text{c/ml}$, for 10 kw.

From the maximum concentrations for the reactor room and the second floor classroom given above, the A^{41} concentrations would be below maximum tolerances if the reactor were operated continuously at 100 kw for a period of two hours. This would not be too much of a restraint since the limitations of the license would permit on the average, one hour per week at 100 kw, per year. However, if the 100 kw operation were restricted to times when the classrooms above and other laboratories adjacent to the reactor room were not being used then longer periods of operation could be safely scheduled.

TABLE 9
MAXIMUM PERMISSIBLE CONCENTRATIONS FOR DIFFERENT EXPOSURE PERIODS

Exposure Period (hr/wk)	M.P.C. for a Restricted Area (uc/ml)	M.P.C. for an Unrestricted Area (uc/ml)
40	2×10^{-6}	4×10^{-8}
10	8×10^{-6}	16×10^{-8}
5	16×10^{-6}	32×10^{-8}
4	20×10^{-6}	40×10^{-8}
2	40×10^{-6}	80×10^{-8}
1	80×10^{-6}	160×10^{-8}

TABLE 10
PER CENT OF MAXIMUM CONCENTRATION (M.C.) FOR DIFFERENT EXPOSURE PERIODS

Exposure Period (hr)	Per Cent of M.C.
1	32
2	53
4	78
5	85

Note: The above tables apply to A⁴¹ only.

The previous estimates do not take into account purging the core region. If the core is purged and the air discharged through a ventilation stack outside the building, then an improvement by a factor of at least 10 should be expected.

VII. SUMMARY AND CONCLUSIONS

The purpose of this investigation was to determine the amount of radioactive argon that is released to the surroundings of the V.P.I. physics building during reactor operation. The results were related to the maximum permissible concentrations of A^{41} for continuous exposure in any seven consecutive days.

In some locations under full power operation the A^{41} concentrations exceeded the maximum permitted values for 40 hours of continuous operation. Since approximately 10 hours of operation is necessary for the argon build-up to reach its maximum and on the average most reactor experiments are for only a fraction of this amount of time, the radiation exposures are nominal. Reactor operation is also limited to 5000 kw-hrs per year by the terms of the license and consequently the annual permitted exposures would not be reached.

Detailed information on unrestricted areas could not be obtained because the detection limit was approximately the M.P.C. for such an area. However, it was possible to check such areas for excessive concentrations of A^{41} .

The results of the core purge experiment showed that the A^{41} concentration in the reactor room itself was decreased by a factor greater than 10.

Thus, one concludes that, if the air is pumped from the reactor core region during operation at higher power levels, and discharged through an exhaust stack outside the physics building, the hazard due to A⁴¹ would be safely controlled.

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Finally the author wishes to thank

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IX. BIBLIOGRAPHY

1. Judd, W. C., "Estimation of Airborn Argon-41 and Fission Products Xenon and Krypton", Reactor Technology Report No. 10 - Chemistry, K.A.P.L.-2000-7, pp. 34-38, (1959).
2. Heath, R. L., et al, Calculated Efficiencies of Cylindrical Radiation Detectors, IDO-16370, (1958).
3. Heath, R. L., Scintillation Spectrometry Gamma-Ray Spectrum Catalogue, IDO-16408, (1957).
4. Crouthamel, C. E., Applied Gamma-Ray Spectrometry, Pergamon Press, (1960).
5. Overman, Ralph T. and H. E. Clark, Radioisotope Techniques, McGraw-Hill, (1960).
6. Price, W. J., Nuclear Radiation Detection, McGraw-Hill, (1958).
7. Benedict, Manson, and T. H. Pigford, Nuclear Chemical Engineering, McGraw-Hill, (1957).
8. Glasstone, Samuel and Milton Edlund, The Elements of Nuclear Reactor Theory, D. Van Nostrand Co., Inc., (1960).
9. Kaplan, Irving, Nuclear Physics, Addison-Wesley Publishing Co., Inc., (1958).

10. Jaffey, Arthur H., "Statistical Tests for Counting", *Nucleonics*, Vol. 18, No. 11, 1960.
11. Ghatak, Ajoy Kumar, "Argon Detector to Monitor Reactor Environment", *Nucleonics*, Vol. 21, No. 2, p. 48, 1963.
12. Code of Federal Regulations, Title 10, Part 20, U. S. Government Printing Office.
13. Christianson, C. and Maggio, R. C., Counting Techniques, Procedures and Instrumentation for Measuring the Activity of Radioactive Samples Aboard Nuclear-Powered Ships, NAVSHIPS-93393A, 1960.
14. Stam, E., Performance Characteristics of the V.P.I. Training and Research Reactor (UTR-10), Thesis, Virginia Polytechnic Institute, Blacksburg, Virginia, 1961.

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ABSTRACT

A gamma scintillation spectrometer was assembled and calibrated in a low background radiation area to analyze air samples. Air samples were taken from different locations in the physics building during reactor operation to determine the concentrations of A^{41} released to the surroundings of the building during reactor operation. The samples were collected in a one gallon polyethylene bottle with a $3\frac{3}{4}'' \times 3\frac{3}{4}''$ cylindrical well sealed in the bottom. The air samples were taken to the counting area and the sampling bottle placed on the detector, a $3'' \times 3''$ cylindrical NaI(Tl) crystal which fitted into the well. The pulses from the detector were fed to a multi-channel analyzer. From the number of pulses per unit time under the photo-peak, the concentration of A^{41} for each location was determined.

Samples were analyzed for 4, 7, and 10 kw from which a linear extrapolation was made to estimate the concentrations of A^{41} in these locations if higher power operation is authorized.