

# PRNC205

PRNC-205

: PUERTO RICO NUCLEAR CENTER

ENVIRONMENTAL MONITORING

oF

ARGON - 41

By

4 Carlos Andrew and Donald S. Sasscor

?OPERATED BY UNIVERSITY OF PUERTO RICO UNDER CONTRACT

NO. © (40.1}1839 FOR US ENERGY RESEARCH AN!) DEVELOPMENT ADMINISTRATION

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ENVIRONMENTAL MONITORING

oF

ARGON - 41

by

carlos Andreu \*

and

Donald S. Sasscer

Work performed at the Puerto Rico Nuclear Center in partial fulfillment of the requirements for the degree of Master of

Science in Nuclear Engineering at the University of Puerto Rico at Mayaguez.

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assTRACT

?The objective of this work is to determine the yearly average maximum concentration and dose of argon-A1 in unrestricted areas adjacent to the Puerto Rico Nuclear Center, and to develop a simple and accurate procedure for measuring argon-A1 concentration, in order to verify the model used for calculation of the attenuation factor

between the off-gas stack outlet and ground level. The model used is

the diffusion equation for average long-period concentration from a continuous point source. In addition, the Kanne Chander, which is the constant air monitor of the reactor off-gas system, was calibrated so as to enable an accurate and easy determination of the future concentration at any power level. The Kanne chamber was satisfactorily calibrated, with a probable error of approximately 5%.

The system used allows concentration at ground level to be measured

to a factor of  $6.75 \times 10^5$  less than the concentration emitted by the

off

stack, of to approximately  $1.6 \times 10^{-10}$  uCi/ec, which is 20% of

the allowable concentration of  $8.0 \times 10^{-10}$  uCi/ec. (2).

The average value of the ratio of the estimated to measured concentration is 1.1, This excellent agreement gives a high degree of

of

confidence in utilizing the model to calculate the yearly average maximum concentration. The largest value of the yearly average maximum concentration occurred during one-shift operation, dry season, at 125 meters from the stack in the ENE sectors and is  $2.69 \times 10^{-10}$  ci/meters<sup>3</sup>

which is 33.67 of the allowable concentration.

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The estimated dose corresponding to the largest yearly average maximum concentration is 0.53 aren per year which is 5.3%

of the allowable dose of 10 aren per year. (1)

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?The sampling of air and the measurement of its radioactive content is often a difficult task due to variable sampling conditions, the diversity of the radioactive substances being sampled, and the fact that there are no standardized air sampling instruments or equipment that can be used for all situations.

?The natural environment has always contained a certain amount of radioactivity. With the discovery and application of radioactive sub-



stances and the incre:

ing use of research and power reactors, another

potential environmental contaminant has been created, thus necessitating a closer scrutiny of various environmental media, including air.

Discharges of radioactive waste to the environment by the Puerto Rican Nuclear Center (PRNC) are regulated by the Energy Research and Development Administration.

The FRNC, functioning as a training and research center, is

engaged in a wide variety of activities some of which involve the TRIGA-FLIP reactor. Such operations invariably produce quantities of gaseous wastes known to contain radioactive nitrogen, oxygen and argon. They are formed from the neutron bombardment of the air contained in the thermal column, beam tubes, gamma room, in-core and side core rabbit system and from the water in the reactor pool.

The formation occurs according to

WALE LUE

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Mie a +e CB) 8 sec

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?The half lives of the isotopes of nitrogen and oxygen which become

radioactive are so short that they decay before escaping from the reactor

building. Substantially all of the activity in the air or water under

normal opt

ating conditions {s due to argon-61.

During normal operation, pool water circulates through the reactor

by natural convection, thereby exposing the air which is dissolved in the

water to the neutron flux of the core. At the same time that the argon in

this air is activated, a fraction of it is driven off from the water due

to the reduced air solubility as the water temperature increases in its

passage through the core. Once the air is driven off, part of it re~

dissolves

it bubbles up through the water and part of it reaches  $\phi$

pool surface and mixes with the building air. It is then exhausted through the normal 16000cfm ventilation system, 69 feet above the site boundary.

In addition, air is circulated through the thermal column: volume and the dry bean tubes and the two rabbit systems by the off-gas system blower.

Of the six beam-tube positions, three contain neutron spectrometer collimators and are not connected to the off-gas system. A fourth one is out of commission and removed from the reactor. Of the remaining two, one is operated in the flooded condition so that only one bean tube is being

swept by the off-gas system. The argon in this air and the air of the

rabbit system is therefore activated and drawn off to the atmosphere by

the off-gas blower. This is @ 150cfm blower exhaust

going to the atmosphere

at a level of 50 feet above the

ee boundary fence.

---Page Break---

Since radioactive argon is the main source of gaseous activity produced by the PRNC TRIGA reactor during normal operation, the primary

objective of this work wa

to develop a simple and accurate procedure

for measuring argon-41 concentration at ground level, in order to verify

4iffusion equation for average long-period concentration fros a continuous

source. This model vas us

for calculating the attenuation factor b $\phi$

tween the off-gas stack outlet and ground level. In addition, the Kanne chamber, which is the air monitor of the reactor off-gas system was calibrated so as to enable an accurate and easy determination of the concentration in the future at any power level.

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## TT REVIEW OF LITERATURE

Research has been performed on this problem by Efigenio Rivera

from Health Physics Division in 1973 (2). This work included the

determination of the concentration of argon-41 bubbles emerging from the pool surface and all of the argon-41 through the off-gas system.

Also, the Kanne chamber was calibrated utilizing the argon-41 produced in the reactor core and emerging through the pool water as the calibrated source.

In addition, Pedro Cruz from the Puerto Rico Nuclear Center Reactor

Division (3) includes the calibration of the Kanne chamber, using

krypton-85 as the standard

The experiment was conducted statically

by

introducing a known concentration of krypton-85 into the chamber. For each concentration the meter readings in cps are recorded and plotted as a function of the known concentration in pCi/cc.

Other research related to the Kanne chamber

refer to the efficiency

determination of a Kanne chamber for detection of radiogases by J. J.

Fitzgerald and B. W. Borelli (4). The inventor of the Kanne chasher, W. R. Kanne, reported a device for the monitoring of gas for radioac~ tivity (5) and for the monitoring of gas for radioactive xenon (6)

These works describe in detai

1 the Kanne chanber and ite principle of operation.

?The Literature survey revealed the existence of other vorks done {in similar areas but not directly related or applicable to this research.

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## INSTRUMENTATION

### A. FOR TRE EFFICIENCY DETERMINATION OF THE DETECTION SYSTEM

A four inch diameter by four inch height cylindrical sodium iodide crystal activated with thalliom was used for the detection

of gamma rays



The detector was connected to a preamplifier and

Linear amplifier, Pulse height analysis was done by a multichannel analyzer (Series 2200 Nuclear Data Analyzer) with the end result typed or displayed on an oscilloscope. A high voltage power supply was

utilized to provide a regulated voltage to the photomultiplier tube.

#### B. FOR THE DETERMINATION OF THE ARGON-41 CONCENTRATION

- 1, In the off-gas system
2. At the surface of the reactor pool

The instrumentation described in

immediately above, was

used to determine the argon-41 concentration in both the off-gas

system and above the reactor pool.

3. At various ground level locations

The m

fenents of the argon-AI concentrations at various ground

Jewel locations required a high-sensitivity counting system. This high-sensitivity counting system was developed by: a) increasing the shielding to six inches of lead, b) connecting two sodium iodide detectors in parallel, and c) obtaining samples of air by using a high pressure

pump to fill a scuba tank to a pre

sure of 2200 psi. The rate of air

entering the tank was found to be constant with time. (See Figure 1).

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OF the ewo detectors used, one was the four inch by four inch detector described above, and the one was a three inch by three

inch sodium iodide cryst

1. Each was connected to a high voltage

supply, @ preamplifier and a linear amplifier, These two connections were introduced to the multichannel analyzer (Series 2200 Nuclear Data Analyzer) for che pulse height analysis, The results were shown

im an oscilloscope and typed.

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## IV METHOD AND RESULTS

### A. OVERALL EFFICIENCY DETERMINATION OF THE DETECTION SYSTEM FOR THE 1.29 Mev CAMWA OF ARCON-A1

An efficiency calibration curve was made for the detection system of argon-t1. A sodium-22 source (which emits 2 gamma of 1.27 Mev 100% of the time) and a cesium-t137 source ( which emits 1 gamma of 0.662 Mev 93.5% of the time) were used as standard sources for the determination of the efficiency, Each source was placed at the same distance from the detector (7.88 ex) and the overall peak efficiency for the count system at each energy was found using

Equation 1

EFFICIENCY (2)

werviny) cProsasryiny \* 1°

PROBABILITY = Probability of emitting a gamma of that energy per disintegration

ACTIVITY = Present activity of source (disintegration/sec)

AREA = Response of a system to the source determined using the Total Peak Area Method (counts/sec)

where

$A_i$  = Number of counts accumulated in channel  $i$

$L$  = Channel number at left hand Limit of photopeak

$R$  = channel number at right hand Limit of photopeak

The overall efficiency for the 1.27 MeV peak of Na-22 and the 0.662 MeV peak of Cs-137 was determined to be 1.317 and 2.29%

respectively, for the detection system described in section III-A.

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Using the efficiency calibration curve of Figure 3, the overall efficiency of the 1,29 Mev peak of argon~K was determined to be 1.3%.

An efficiency curve obtained from reference (7) for 9 typical

four inch by four inch sodium iodide detector is also shown in Figure 2.

## B. DETERMINATION OF THE ARGON-41. CONCENTRATION

### 1. In the off-gas system

Argon-41 concentration in the off-gas system was determined by

taking samples in a 1000 cc spherical #1

O88 container with a stopcock

valve, The container was previously evacuated to a pressure of approximately

0.1 mm of mercury for each sample. Each time a sample was

collected, the time of collection

was noted, start counting time, power level

of the reactor, air flow through the stack and Kanne chamber readings were recorded. (See Table 1).

In order to apply the counting efficiency of the system as

obtained from point sources in Section IV-A to a much La

yt volume, a

Geometry factor is needed. Since this factor had already been obtained by Efigenio Rivers in his thesis (2) and the same container and geometry

4 by Rivera was used in this work (7.88 cm as the distance from the exact center of the 1000 cc sphere to the detector), it was possible

to use his geometry factor for correcting the point source

assumption.

This correction factor is 0.950 for 1000 cc sphere.

Counts of 10000 second, live time, were obtained for each sample.

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EFFICIENCY (2)

on

Figure 2.

n

Efficiency Curve for a  
Typical 4" by 4" Na T  
Detector at 5 cm from  
the source (7)

1.27 Mev Gamma



on 1.07 10

ENERGY (Nev) 1,29 Mew Gacena

of Ard

Overall Efficiency as a Function of Energy of the Four Inch

Diameter by Four Inch High Cylindrical No I Detector, 7.88 cm

from the Souree.

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weashs se9-540 92 Uy YoyaeuTml9s0q Wo}IEIzUeDNOD Ty-UORIY ?tT eTGeL

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BD

In order to keep the same overall efficiency as was determined in Section IV-A, the 1.29 Mev argon-41 peak was determined in the same way as the 1.27 Mev sodium-22 peak. Total Peak Area Method

(explained in Section IV-A) was used to calculate the area under the

Peak. Since this area is not representative of the activity in the

off

system at the ins

int of collection, it had to be corrected

for decay time. (See Appendix).

Therefore, the argon-41 concentration in the off-gas system at

the instant of collection is given by

$C = \frac{A}{V \cdot \epsilon \cdot \eta \cdot WCF \cdot HEC}$

where

$A$

is the total peak area in counts/sec per channel

$t$  is the time decay correction factor

$\epsilon$  is the efficiency of the counting system

$3.7 \times 10^4$  is the conversion factor from disintegration/sec to  $\mu\text{Ci}$

$V$  is the volume correction factor for the 1000 cm<sup>3</sup>

collection sphere

Results are presented on Table 1.

4. Kanne chamber calibration

Radioactive argon-41 concentration in the off-gas system is

continuously monitored by a Kanne chamber. The Kanne chamber was

accurately calibrated in order to have confidence in future readings

The calibration was performed using the data obtained from the of gas system concentration determination. (See Table 1). A Line to correlate the recorded Kanne chamber reading and the determined argon-b1 concentration was drawn by Linear regression analysis. (See Figure 3.)

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; stope =  $2.3 \times 10^8$  4  $0.06 \times 10$

g yetle per sep

=

01x 10 5x 10d

Kanne Chander Reading

(emp)

Figure3 . Kanne Chamber Calibration Curve

---Page Break---

as

?These data gave a conversion factor between Kanne chamber reading and concentration in nCi/ee of

Concentration (nCi/ec =  $(2.3 \times 10^6 + 0.06 \times 10^5) \times$

(Kame chamber Yeading) e

This indicated that the probable error io the conversion of Kanne chanber to a concentration is approxinstely 57.

b, mission rate variation with power level

?The argon-41 emission rate should be proportional to the flux or power level. Since the samples were taken at different power Levels,

(See Table 1) a Line was draun to check the Linearity of the activity

of argon-41 production as a function of power (See Figure 4). This line gave an average emission rate of 543 nCi/min with a probable error of 6% at 1000 Kw.

## 2. At the surface of the reactor pool

Since the area where the argon-41 emerged through the reactor pool water was large, a sheet metal cone was placed on top of the water surface to collect some of the effluent gas. A polyethylene tube was connected to the collector, and to one side of a th

way

glass valve. One valve outlet provided means for sample drawing at any time, the other outlet was connected to another section of the tube with its end under two inches of water. (See Figure 5).

This was done in order to determine the volume rate in cc/min of argon-41 effluents. The area occupied by the collector on top of the pool surface was approximately one half of the total area

through which the argon-41 effluent was emerging. Therefore, since

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the volume rate collected was measured to be 133 ce/min, the total argon-41 effluent released into the reactor building was estimated to be 300 ce/min.

Using Equation 2 the concentration of argon-41 at the pool

surface was obtained and is presented in Table 2.

3. At various ground level Locations

a. Sensitivity of counting system

The high sensitivity of the new counting system (discussed in Section IT1-B-3), without taking into consideration the increased Pressure, was determined before the system was used. This was done

by taking «

sample of the off-gas in a scubs tank (previously evacuated to @ pressure of 0.4 nm of mercury) at the stack. The tank was

filled to atmospheric pres:

re. Activity for periods of 4000 seconds

were obtained for almost 24 hours. In all of the sixteen runs the

argon-41 peak was present even though the last run was counted 12.5 half lives after the sample had been taken. This could be done because the background was reduced and the efficiency was increased by a factor of two.

A curve of the activity as a function of time was obtained and the decay constant was found as  $1.042 \times 10^{-4} \text{ sec}^{-1}$  which is very near to the accepted value of  $1.052 \times 10^{-4} \text{ sec}^{-1}$ ,

The

method explained above gave the sensitivity of the counting



system taking into consideration only the increase in shielding. and in  
the efficiency, This gave an initial to final count ratio of approxi-  
ately 4500, when the air in the tank was at atmospheric pressure

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uv

600

500 Probable error = 61

400

300

Exposure Rate (Ct /min)

200

100

° 250 300° «75040001250

Power Level (iu)

Figure 4. Entssion Rate of Argon

41 im the Off-Cas System as a Function  
of Power Level.

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18

Glass sphere

Three Way Valve

Polyethylene

Tube

supple

collector

Gis

Graduated  
cylinder

Diffuser

Jee

Reactor

Coxe

Reactor Pool

Figure 5+ Schematic Diagram of the Experimental Sample Collection set ups

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Table 2. Argons1 Concentration Determination

in the surface of the Reactor Poot

Foxe Ta Decay?

; \* Emission

Level desintezracion, Tine \$@Meentracion Ale Tow MSSE

co) seeree (see) (WEEE) Cela) ctfnin)

1000 0.751999 22027 6.38x10<sup>7</sup>) 300, 1,698

1000 310207 650 6.98x10<sup>8</sup>) 300, 1.852

wate Recorded

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20

However, the

tivity was increased by a factor of 150 by collecting air in the scuba tank at a pressure of 2200 psi. Therefore, the system can determine concentrations to approximately 150 times 4500  $\times 6.75 \times 10^5$  times less than the concentration leaving the off-pas

stack. The concentration in the off;

system is approximately

$10.8 \times 10^5$  uci/ec at the 1000 Kw power Level, Therefore, the system

can determine concentrations to approximately  $1.6 \times 10^{-10}$  wci/ee, which is less than 20% of the allowable concentration of  $8 \times 10^{-10}$  wei/ec (1).

b. Measured value

Using the high sensitivity system described above, samples of air at various ground level locations were obtained in a scuba tank which

was filled to a pressure

of 2200 pounds per square inch. This pressure

was obtained using a scuba tank air compressor. This tank was previously

evacuated to a

pressure of 0.4 mm of mercury. Counts of 404K) seconds

(five time) were taken for each sample. For every sample the following

was recorded: Kanne chamber reading, air flow coming out of the stack,

distance from the stack to the sampling location, wind speed, wind

direction, wind frequency to the sampling location, meteorological

conditions, time at start of collection, start counting time, and pressure

of the tank after finishing the countings. All of these parameters were

needed. Some to calculate the mass

8 values, others for the estimated

value

Using the Kanne chamber reading, the argon-41 concentration at the stack could be determined from the calibration curve. Using Equation 3.

Concentration at the stack ~ (Kanne chamber) (Conversion factor)

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2

$$10.3 \times 10^{-5} = (4.5 \times 10^{-11})(2.30 \times 10^8).$$

If the tank is filled with argon-41 to atmospheric pressure at

the stack, and counted with the same geometry and of

efficiency as the

one filled at ground level, we can determine the argon-41 concentration

at ground level using the following relation,

Concentration at ground level = Activity at ground level

efficiency at ground level / efficiency at ground level

Concentration at ground level

where appropriate corrections are made for counting and decay time

(See Appendix)

The activity at ground level (Table 4) and the activity at the

stack (Table 3), to which the ground measurements were compared, were

obtained the same day in order to minimize the error determining the

attenuation factor between the stack and ground level.

f. Calculated value

Three meteorological factors are of significance in determining the amount of mixing which will occur of contaminants in the event of diffusion

into the atmosphere. They are: the distribution of temperature

with height, the surface wind direction and speed and the precipitation.

A complete mathematical

analysis of diffusion processes which would

allow quantitative calculations of concentration is difficult because

instantaneous velocities exhibit irregular amplitudes and frequencies

Equations currently in use are based in part on the

empirical analysis

must necessarily include some constants which are empirically determined in nature

?These constants are functions of eddy viscosity

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2%

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bility. Determining suitable values for these coefficients and parameters is the most important operation in their application. Suggested diffusion coefficients for the PRNC, Nayaguer, are shown in Table 5 and are obtained from Reference 8.

The concentration of argon-41 as a function of distance from a continuous, long term, point release of gas was calculated from Equation

3-143 or Reference 9 for the cross wind integrated concentration

where

where the standard deviation of the distribution of material in a plume in the Z direction,  $\sigma_z$ , determined by the Sutton form (Sec 8-4) of (9) to be

$$\sigma_z = M C_2 x^{(2-a)}$$

Then the concentration is averaged

over a 22.5 degree sector (1/16 of a circle),

then the long-term average concentration is given by

$q^2$

$2 \sigma_a$

= toy at @

diffusion coefficient in the z plane (meters)<sup>2</sup>/s

X = distance from the source (meters)

σ = nondimensional parameter associated with stability

f = percent of frequency with which the wind blows from a given sector

U = rate of material emission from a continuous point source (Ci/week)

u = mean wind speed (meters/sec)

h = height of source (meters)

Δx =

Δx = the sector width for each of N sectors at the distance

X = 16)

"concentration with air at (Ci/meter<sup>2</sup>)

---Page Break---

Table 5. Diffusion Coefficients

Weak Lapse Rate

(Adiabatic) 0.25 0.26 0.36

Large Inversion,

Stable Conditions 0.50

---Page Break---

Table 6 presents the estimated argon-41 concentration at ground level and Figure 6 shows the relationship between the estimated and

measured argon-61 concentrations.



#### 4. Yearly average maximum concentration

To determine the yearly average maximum concentration of aryanol, the average velocity and frequency of wind in each 22.5° directional sector was obtained on an hourly basis for the dry and wet season from the climatology data of PRNC-37, Section IV, Appendix A, (8). The hourly values were averaged between 8:00 a.m. and 4:00 p.m. to obtain the average wind velocity and frequency per sector for one-shift operation. For the two-shift operation, the hourly values were averaged between 8:00 a.m. and 6:00 p.m. (11 hours of day), and 7:00 p.m. and 11:00 p.m. (5 hours of night). The division between night and day was necessary due to the fact that in Mayaguez the nights have 90% probability of large temperature inversion and the days a high probability of adiabatic conditions (8). Therefore, to determine the concentration, two sets of diffusion constants had to be used, one for the day and one for the night. (See Table 5)

For the dry season, during both one-shift and two-shift operations,

the concentration was greatest in the EXE sector. In the

rainy season

the greatest value of concentration for both operations was in the W

sector. Although the SW sector and the ENE sector have almost the same

Frequency-to-velocity factor during the dry season, and W and SSW sectors

have also almost the same frequency-to-velocity factor during the rainy

on, the ENE and the W sectors had the highest concentration due to the elevation of land in these sectors, giving a smaller effective stack

height.

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The emission rate for the 1000 Kw power level was assumed to be 9.0 (10°) Ci/see based on the emission rate as a function of power level of Figure 4.

Substituting the values of Table 7 for each separate condition in Equation 7, the concentrations as function of distance were determined and are presented in Figure 7 and 8. The distance from the source, at which the yearly average maximum concentration occurs, was determined from these figures.

The 1000 Kw power level yearly average maximum concentration of argon-41 in uncontrolled areas, during one-shift and two-shift operations,

during the dry and the wet seasons, is shown in Table 7.

The locations of the maximum concentration are shown in Figure 9

### C. YEARLY AVERAGE MAXIMUM DOSE CALCULATIONS

#### 1. Internal dose

The cumulative dose, from the beginning of exposure, due to a finite

inhalation time was calculated using Equation 7.101 of Reference 9.

5.92 (102),

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here

$E$  = energy absorbed in the organ from one disintegration (MeV)

$M_m$  mass of the organ (g)

fraction of radionuclide transferred to the organ retention factor

breathing rate (m<sup>3</sup>/sec)

$X$  = yearly average maximum concentration (Ci/m<sup>3</sup>)

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+9 aren

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@ Estimated (2/27/75)

AMeasuzed 2/27/75)

Concentration (aci/ee)

9200 am, 11:00 10

Tine of

Figure 6. [stinated and Measured Argon~

29

o

it

0 pam 9200

Day

41 Concentration st Ground Level.

---Page Break---

30

Allovable

Dry Season

Rainy Season

yo!

Yearly Average Maximum Concentration

(ct/meters<sup>3</sup>)

so??

400° 800 1209 1600 7000 2400-2800

Distance from the Source (meters)

Figure 7. Yearly average Maximum Concentration at Ground Level for One Shift Operation During the Dry and Rainy seasons as a function of Distance.

---Page Break---



au

Allowable

? Dry Season

Rainy Season

(ct/meters?)

or

Yearly Average Maximum Concentration

102? 4

400 1200 2000 2800

Distance from the Source

(meters)

Figure 8. Yearly Average Maximum Concentration at Ground Level for  
PrepShift Operation During the Dry and Rainy seasons

© Function of Distances

---Page Break---

more?

1 Dry Season

2 Rainy Season (open)

2 at 02 o'clock

Mg - Well Locations

SCALE

Figure 9. Map of the Reactor Site showing the Location of the Yearly  
Average Maximum Concentration.

---Page Break---

3a

$T_y$  = radioactive half Life (see)

$T_y$  ~ biological half Life (sec)

$f$  = time of exposure (see)

$t$  = time following exposure (sec)

Dose to the organ from internal radiation (rad)

When inhaled, argon is assumed to be distributed evenly throughout

the lungs. The lungs are, therefore, the critical organs and  $M = 1000$  g

Using the standard man, (10).

The energy absorbed from the beta and gamma radiation of  $^{40}\text{K}$

was assumed to be one third of the end-point energy of the beta and to

be the amount of gamma energy absorbed when passing through 10 cm of lung

matter, as is given in Equation 7.89 of (1).

ts

Pe Banas en

Ws the energy absorption coefficient of the lung for the gamma

photon  $C_{ea-1}$ )

$F$  = the effective diameter of the organ (cm)

$E_{\gamma}$  = gamma energy per disintegration (MeV)

$E_{\beta}$  = end point energy of the beta (eV)

The fraction of the radionuclide, transferred to the organ by

inhalation,

for  $f_{in}$  assumed to be 0.10, as recommended by DE. Theodore

Agard. (11).

The breathing rate

for standard man was assumed to be

$2.78 \times 10^{-4}$  m<sup>3</sup>/sec.

The biological half-life for argon-41 is short, but not well

Known. A value of 30 sec was assumed, knowing that xenon, also a noble gas, has a biological half-life of less than 30 sec (11). Therefore,

the effective decay constant,  $\lambda$ , is 0.0232 sec<sup>-1</sup>,

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esDTHCETT orvorg og YOTE2NGT

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?The exposure time,  $t$ , equals eight hours for one shift operations

?and eleven hours for two-shift operations. The concentration of argon= $\$1$

uring the last five hours of two-shift operation is more than ten orders

of magnitude Less than the concentration for the day-time operation and

is therefore neglected. The dose received after exposure has stopped

( $t= 300$  see). This is ten times the assumed biological half life.



The results of the internal dose calculations for an average day  
in aren are given in Table

To convert from rad to rea a relative biological effectiveness (REE)

factor is needed which for gannas and bet

is equal to one.

## 2. External dose calculations

calculated

?The external dose in a homogeneous infinite cloud we

by adding the contribution due to the beta and gama rat

tion. The

calculation was made using Equations 7.20 and 7.35a from Reference 9.

$$DY = (0,23 E + 0.25 BE)$$

where

$E$  = end point energy of the beta (ev)

$B$  = gamma energy per desincesration (ev)

$Y$  = yearly average maximua concentration (Ci/meters<sup>3</sup>)

$D$  = dose rate (rad/sec)

1 calculating the yearly average saxime dose, internal plus

external, the internal dose can be neglected since it is a factor of «

thousand less than the external dose. (See Table 7).

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## DISCUSSION OF RESULTS

A. OVERALL EFFICIENCY DETERMINATION OF THE DETECTION SYSTEM FOR  
THE 1,29 Mev GAMMA OF ARGON-61

## The Scintillation Spectrometry Cs-137-Ray Spectrum Catalogue

Index 11), (7) indicates that the total spectral efficiency of a

Standard four inch diameter by four inch deep sodium iodide detector

in response to a sodium-22 point source, 7.88 cm above the detector should be 4.2%. In addition, this same reference gives the peak to total spectrum ratio for sodium-22 in a three inch diameter by three inch deep detector as 0.35. This implies that we might expect our system to have an efficiency of  $4.2\% \times 0.35 = 1.5\%$  rather than 1.31%. This difference is possibly explained by : a) different techniques used in obtaining the Peak spectra (The Spectrum Catalogue does not discuss the technique used in obtaining the peak spectrum), b) a lower intrinsic efficiency of our detector due to detector fatigue or other reasons (our detector is eight years old) and c) lower efficiency of the electronic components of our system,

However, it should be noted that the accuracy of the measurement is not dependent upon determining the absolute efficiency of the system, but rather upon using the ratio technique which assures that the ratio of counts of this detector to activity of the source is the same for argon-41,

cesium-137, and sodium-22.

Thus the ratio

Counts under peak for

Known activity of Na-22 7 1-302

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Using Figure 2 to extrapolate from the 1.27 Mev gamma peak of the

to the 1.29 Mev gamma peak of argon-41 we obtain

Counts under peak for Anil

Known activity of azo ~ 130%

and this is the value we have used for the efficiency of the system,

XE should be noted that great care was taken in determining the efficiency of the system, since all succeeding work depends upon the efficiency and since our value of 1.29% differs by 30% from the value of 1.8% obtained by Efigenio Rivera (2).

## DETERMINATION OF THE ARGON-41 CONCENTRATION

### 1. In the off gas 53

The average argon-41 concentration and emission rate for 1000 kw Power level were found to be  $10.80 \times 10^{-6} + 0.37 \times 10^{-3}$  wCt/ce and 543 417.5 MCi/min, respectively. The difference between these values and the ones used by Efigenio Rivera (2),  $2.7 \times 10^{-5}$  wCi/ce and 116.1 ACi/min are due to the difference in the efficiency and Flow rate measurements,

Kanne chamber calibration

4m estimation of the accuracy of the Kanne chamber reading can be obtained from Table 1 and Figure 3. At @ constant indicated power level of 1000 Kw, both the Kanne chamber reading and the concentration as determined in the experinent show a variation of approximately 5% around the mean value. Woweever, it is not known which part of the variation should be attributed to errors in the indicated pover level, the concontra~

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tion and the Kanne chamber res

ing. Tn addition, the Kanne chanber can be read to only two significant figures, this in itself gives a possible error of approximately 22.

The conversion factor of  $2.3 \times 10^6 + 0.06 \times 10^46$  uci/ee per amp found contains seven of the nine data points, as indicated in Figure 3.

It is therefore concluded that the best estimate in error in converting

Kanane chamber reading to concentration is approximately 5t.

B, Emission rate variation with power level

?The Linearity of the emission rate of argon-41 production as a func~

tion of power level was determined with a 6% probable error.

2. At the surface of the reactor pool!

?The argon-41 concentration and the emission rate measured at the

Surface of the reactor pool are  $6.383 \times 10^{-3}$  wCi/cc and 1.7 Ci/min

respectively

3. At various ground level locations

8, Sensitivity of counting system

?As discussed in Section T11-B-3, three modifications were made to

the standard system in order to improve the sensitivity of the system

for counting argon-41. They were: a) increasing the shielding to six

inches of lead, b) connecting two sodium iodide detectors in parallel,

and c) collecting air in a scuba tank at a pressure of 2200 psi.

?An indication of the accuracy of the system, with both detectors and

---Page Break---

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increased shielding was obtained by determining the half life of

argon-41 using the decay constant obtained as  $1.042 \times 10^{-4} \text{ sec}^{-1}$ ,

Lin 2

Decay Constant ~ 185 hours

Nae

The half life of argon-41 reported by Lederman (12) is 1.83 hours, however, he refers to three works which report 1.83 hours, one which reports 1.82 and the other which reports 1.85 hours. The initial to final count ratio of approximately 4500 indicates the sensitivity of

the counting system considering only the first two modifications,

However, the sensitivity was increased by a factor of 150 by collecting air in a scuba tank at a pressure of 2200 psi. Therefore, when the three modifications were combined, the argon-41 concentration at ground level could be read to approximately  $1.6 \times 10^{-10} \text{ yci/ec.}$ , which is less than



20% of the allowed concentration of  $8.0 \times 10^{-10}$  yci/ec with a probable

error of not more than 10%.

b. and c. Measured and calculated values

?The average value of the ratio of the estimated to measured concen

tration is 1.1, The low frequency case (run No. 2 of Tables & and 6)

was not included in this average because the equation we used for

estimating the concentration is for long term average release, An

implicit assumption in this equation is that the percent of time the

Gas leaves the stack, in the direction of a given sector

is equal to

the percent of time the gas

is found in the sector at some distance x

from the stack. This will be true for both low and high frequency ia

---Page Break---

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4 sector during a long period of time such as a year since the crosswind component is insignificant for the high frequency situation, and since for the low frequency case the crosswind components in a sector would be self compensating. However, for a short period of time, such as the half hour period, during which our samples were collected, this equation will only be applicable for the high frequency situations. For

the low frequency case and short collection time the model does not

apply due to the fact that when the crosswind component is in a constant

direction, very little, if any, of the gas entering @ sector of the stack

would still be in that sector a significant distance from the stack.

?The excellent agreement between the measured and estimated

Results serve as a verification of the model used and give a high degree of confidence that the calculated yearly average

#### 4. Yearly average maximum concentration

The previous value used for the yearly average maximum concentration, one-shift, dry season was  $1.63 \times 10^{-10}$  Ci/meters<sup>3</sup> (13), where the value obtained in this study is  $2.69 \times 10^{-10}$  Ci/meters<sup>3</sup>. Since the presently allowable value is  $8.0 \times 10^{-10}$  Ci/meters<sup>3</sup>, which is 22% of the allowable value specified in 10 CFR Part 20, appendix

Table 11, Column 1, (1)

the yearly average maximum concentration is 33.6% of the allowable.

#### ©. YEARLY AVERAGE MAXIMUM DOSE CALCULATIONS

The previous value used for the yearly average maximum dose one-shift,

?477 season was 0.45 aren (13), where the value obtained in this study is

---Page Break---

a

0.53 mrem, Since the presently allowable value {s 10 aren, the yearly average maximum dose is 5.32 of the allowable

Te can be noted that if a person is exposed to the daily average saximum dose, not for one-shift operation but rather for 24 hours day, seven days 4 week, then the dose would be 50z of the allowable. These

values seem to indicate a reasonable correlation between the concentra:

tion and dose.

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ConciusroN

?The Kanne cha

"eF appears to be satisfactorily calibrated with

@ probable error of approximately St,

The emission rate was found to be Linear as a function of power.

The average en:

ion rate for 1000 Ky, with all of the off-gas

facilities connected, was determined to be 543 pCi/min, with prob:

ble error of 6%.

The syst

using improved shielding, parallel detectors, and scuba tank allows concentration at ground level to be measured to a factor of  $6.75 \times 10^5$  less than the concentration emitted by the off-gas stack, or to approximately  $1.6 \times 10^{-19}$  ci /ce.

The average value of the ratio of the estimated to measured concentration is 1.1, This excellent agreement between the measured and estimated concentrations gives a high degree of confidence in utilizing

the diffusion equation for average long-period concentration from a

continuous point source to calculate the yearly average maximum concentration. The Largest value of the yearly average maximum concentration occurred during one-shift operation, dry season, at 125 meters from the stack in

the ENE

sector, This value was found to be  $2.69 \times 10^{-10}$  Ci/meters<sup>3</sup>, which is 33.62 of the allowable value as proposed in June, 1974. (1)

The dose corresponding to the largest yearly average maximum concentration

tration is 0.53 mrem per year, which is 5.3% of the allowable value.

Since the concentration and dose are 33.6% and 5.3% of the allowable

values, respectively, argon-41 emission is not a limiting value for the

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a

PRNC TRIGA-FLIP reactor operating at 1000 Kw. Although for 2000 kw operation, the concentration and dose would double, the concentration would still be approximately 70% of the allowable and probably indi-

cates a safe operating condition,

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4?

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APPENDIX

DECAY TIME

acTIVITY

a)

DECAY TIME

where

to 7 Activity at start counting time

T's (ty + t,) = counting time period

H = average activity for the counting time period

T= time from start counting until average activity is reached

?Therefore, it can be stated that

ven = 1" ?4 nat

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a?

then

but.

therefore

Teta Ge

now for

1 = 10000 seg

end

4,

tn 2\_ (hour) | 1.052(10"

Decun "Tar he Woo see) = SE

we have the following

« 4 (4.052) (1074) (10000)

Terres SSR ita}

Th = 6566 sec

?The following is a table containing all the T\* corresponding to the

counting periods used.

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|

Ti(seg) \_T(see)

100 49.96

se \_\_\_1930

10K 4566

20K\_\_\_ 8307

?The fact that time had passed from the sample collection time to the start counting time was taken into consideration. So the total decay time  $t_s$

peter

where

1 = decay correction time during counting

T= time between collection and start counting

In the case where the tank was filled up to a pressure of 2200 psi, the pumping time must also be considered. The pumping time was measured and it was determined that the rate of air entering the tank is a constant function with time, therefore, T to determine decay time is equal to half of the pumping, plus the time between the end of pumping and the start of counting.

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