AN ABSTRACT OF THE THESIS OF

Robert Allen Meck for the Master of Science Degree in General Science, Radiological Physics presented on August 29, 1967

Title: Argon-41 Production by the OSU TRIGA III Reactor

Abstract approved: John C. Ringle

A sensitive ion chamber-electrometer system was used to determine the Oregon State University TRIGA III reactor's $^{41}$Ar production rate as a function of steady state reactor power and production of $^{41}$Ar due to reactivity pulses up to $3.00$. The relative contributions of the argon duct effluent and the pneumatic transfer system exhaust were measured. The minimum $^{41}$Ar production rate at 250 KW reactor power was found to be 76 µCi/min. The equilibrium production rate of $^{41}$Ar was found to be a linear function of reactor power. By linear extrapolation, at 1 MW reactor power the minimum $^{41}$Ar equilibrium production rate is predicted to be 303 µCi/min. The $^{41}$Ar produced due to a reactivity pulse was seen to be a linear function of both pulse worth and nvt. A $3.00$ pulse produces 85 µCi $^{41}$Ar which is released to the atmosphere.
ARGON-41 PRODUCTION BY THE OSU TRIGA III REACTOR

by

Robert Allen Meck

A THESIS

submitted to

Oregon State University

in partial fulfillment of
the requirements for the
degree of

Master of Science

June 1968
APPROVED:

Associate Professor, Mechanical Engineering

Acting Chairman, General Science

Dean of Graduate School

Date thesis is presented: August 29, 1967

Typed by Dianne Neketin for Robert Allen Meck
ACKNOWLEDGEMENT

The author wishes to thank his major professor, Dr. John C. Ringle, for his helpful suggestions and criticisms. A sincere, "Thank you," to all of the friends who have generously shared their time, experience, and equipment. A special thanks is extended to the typist, Dianne Neketin, who has been so patient.
# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>INTRODUCTION</td>
<td>1</td>
</tr>
<tr>
<td>EXPERIMENTAL APPARATUS</td>
<td>3</td>
</tr>
<tr>
<td>CALIBRATIONS</td>
<td>7</td>
</tr>
<tr>
<td>STEADY STATE $^{41}$Ar PRODUCTION RATES</td>
<td>11</td>
</tr>
<tr>
<td>$^{41}$Ar PRODUCTION BY PULSES</td>
<td>17</td>
</tr>
<tr>
<td>DISCUSSION</td>
<td>21</td>
</tr>
<tr>
<td>CONCLUSION</td>
<td>25</td>
</tr>
<tr>
<td>BIBLIOGRAPHY</td>
<td>27</td>
</tr>
<tr>
<td>APPENDICES</td>
<td></td>
</tr>
<tr>
<td>APPENDIX I. CONSTANTS USED IN CALCULATIONS</td>
<td>28</td>
</tr>
<tr>
<td>REFERENCES</td>
<td>29</td>
</tr>
<tr>
<td>APPENDIX II. THERMAL NEUTRON FLUX CALIBRATION</td>
<td>30</td>
</tr>
<tr>
<td>APPENDIX III. $^{41}$Ar SOURCE CALIBRATION</td>
<td>34</td>
</tr>
<tr>
<td>APPENDIX IV. FLOW MEASUREMENTS</td>
<td>36</td>
</tr>
</tbody>
</table>
LIST OF FIGURES

<table>
<thead>
<tr>
<th>FIGURE</th>
<th>DESCRIPTION</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>FIGURE 1</td>
<td>SCHEMATIC: ARGON DUCT EXPERIMENTAL CONFIGURATION.</td>
<td>4</td>
</tr>
<tr>
<td>FIGURE 2</td>
<td>SCHEMATIC: PNEUMATIC TRANSFER (RABBIT) EXPERIMENTAL CONFIGURATION.</td>
<td>5</td>
</tr>
<tr>
<td>FIGURE 3</td>
<td>SCHEMATIC: FLOW MEASUREMENTS.</td>
<td>6</td>
</tr>
<tr>
<td>FIGURE 4</td>
<td>GAMMA RAY SPECTRUM OF ARGON DUCT EFFLUENT AT 250 KW REACTOR POWER.</td>
<td>12</td>
</tr>
<tr>
<td>FIGURE 5</td>
<td>PRODUCTION OF $^{41}$Ar AT EQUILIBRIUM.</td>
<td>16</td>
</tr>
<tr>
<td>FIGURE 6</td>
<td>ARGON DUCT EFFLUENT ACTIVITY FOLLOWING A $3.00$ PULSE.</td>
<td>18</td>
</tr>
<tr>
<td>FIGURE 7</td>
<td>COMPARISON: RABBIT EXHAUST ACTIVITY FOLLOWING A $3.00$ PULSE.</td>
<td>18</td>
</tr>
<tr>
<td>FIGURE 8</td>
<td>PRODUCTION OF ARGON-$^{41}$ DUE TO PULSES.</td>
<td>20</td>
</tr>
<tr>
<td>TABLE</td>
<td>CONTENT</td>
<td></td>
</tr>
<tr>
<td>-------------</td>
<td>-------------------------------------------------------------------------</td>
<td></td>
</tr>
<tr>
<td>TABLE I.</td>
<td>FLOW RATES</td>
<td></td>
</tr>
<tr>
<td>TABLE II.</td>
<td>SATURATION 41Ar CONCENTRATION IN THE ARGON DUCT</td>
<td></td>
</tr>
<tr>
<td>TABLE III.</td>
<td>SATURATION 41Ar CONCENTRATION IN THE RABBIT EXHAUST</td>
<td></td>
</tr>
<tr>
<td>TABLE IV.</td>
<td>41Ar PRODUCTION RATES AT EQUILIBRIUM</td>
<td></td>
</tr>
<tr>
<td>TABLE V.</td>
<td>41Ar PRODUCTION BY PULSES</td>
<td></td>
</tr>
<tr>
<td>TABLE VI.</td>
<td>A COMPARISON OF ARGON MONITORS</td>
<td></td>
</tr>
</tbody>
</table>
ARGON-41 PRODUCTION BY THE OSU TRIGA III REACTOR

INTRODUCTION

The quantitative determination of radioactive materials released into the environment is important to all nuclear reactor operations. Assays of these materials are used to maintain safe working environments and to assure the general population near reactor facilities that the concentrations of radionuclides in air and water do not exceed those recommended by the National Committee on Radiation Protection (5).

The only significant radioisotope release to the atmosphere by the Oregon State University TRIGA III reactor is argon-41. The $^{41}\text{Ar}$ is produced by the $^{40}\text{Ar} (n, \gamma)^{41}\text{Ar}$ reaction during the irradiation of air within the reactor system by thermal neutrons. Hence, the production and consequently the release of $^{41}\text{Ar}$ from this and similar terrestrial reactors is usually unavoidable. Although the maximum permissible concentration (MPC) for the unrestricted release of $^{41}\text{Ar}$ in gaseous effluents ($4 \times 10^{-8} \mu\text{Ci/cc}$) is not low in comparison to that of other isotopes, measurements of $^{41}\text{Ar}$ releases are complex. Since $^{41}\text{Ar}$ is a noble gas, scrubbing systems and filters have little effect upon its concentration in air. Calibrated sources of $^{41}\text{Ar}$ are commercially unavailable in small quantities of activity, since the half-life is 110 minutes, making the calibration of an instrument which can
detect this isotope at or near the MPC more complicated than routine calibrations.

The object of this work was to obtain an accurate measurement of the $^{41}\text{Ar}$ produced by the OSU TRIGA III reactor as a function of irradiation conditions and power levels. The value of such a measurement lies in verifying calculations for reactor license applications and evaluating the design and health physics requirements for proposed and existing TRIGA reactors. The results herein enable one to predict the relative and absolute production of $^{41}\text{Ar}$ in facilities such as beam tubes, thermal columns, and pneumatic transfer tubes.
EXPERIMENTAL APPARATUS

Air which is unavoidably exposed to thermal neutrons from the reactor is contained in the following facilities: Four beam tubes, thermal column, thermalizing column, pneumatic transfer tube, lazy susan access tube, central thimble tube, reactor pool and irradiation pool waters (dissolved), and outside the shielding in the reactor bay. The first four facilities listed are the only significant sources of the $^{41}$Ar released to the atmosphere. The air in the other locations is either trapped, in low concentrations, or exposed to a low thermal flux. For these reasons, the only measurements of $^{41}$Ar were from the air exhausting from the beam tubes, thermal column, thermalizing column, and pneumatic transfer system (rabbit).

The measurement of $^{41}$Ar was made by passing the entire effluent of the argon duct, comprised of the exhausts from the four beam tubes plus those of the thermal and thermalizing columns (Figure 1.), through a Cary Instruments Model 3810, ten liter ion chamber-electrometer tritium monitor. The exhaust of the rabbit facility was similarly passed through the Cary electrometer system (Figure 2.).

The volume flow rate of air passing through the chamber was determined by using a sharp edged orifice, a water micro-manometer, a mercury barometer, American Society of Mechanical Engineers (ASME) standard pressure taps (7), and a calibrated thermometer (Figure 3.).
FIGURE 1. SCHEMATIC: ARGON DUCT EXPERIMENTAL CONFIGURATION.
FIGURE 2. SCHEMATIC: PNEUMATIC TRANSFER (RABBIT) EXPERIMENTAL CONFIGURATION.
FIGURE 3. SCHEMATIC: FLOW MEASUREMENTS.
CALIBRATIONS

The calibrations involved in the measurement made are, in broad terms, two-fold: One major section of calibration involves the detection of $^{41}$Ar; the other deals with the measurement of the air volume containing the argon. The measurement of radioactivity will be discussed first.

Many installations have found that the detection of the MPC values of $^{41}$Ar is difficult, if not impossible, with Ge-M or scintillation detectors (1, 2, 3, 4, 6). Primarily for this reason, it was decided that a sensitive ionization chamber-electrometer detector calibrated with a known amount of $^{41}$Ar would be an efficient detection system.

The argon source was manufactured by irradiating research grade, stable argon (99.9995% argon) in the glory hole facility of the OSU AGN-201 reactor. The thermal neutron flux within the argon irradiation container was measured by using NBS calibrated gold foils (Appendix II). An average value of the flux was obtained by weighing the plot of flux measurements vs. reactor position. The argon was placed in the container at atmospheric pressure, and the temperature of the glory hole was measured during irradiation with an NBS calibrated thermometer. The volume of the container was measured by weighing the irradiation vessel filled with water of a known temperature. The amount of $^{41}$Ar produced was calculated by using the equations derived
in Appendix II in conjunction with the ideal gas law (Appendix III).

The ionization chamber, a Cary Instruments Model 3810 tritium monitor was sealed and partially evacuated. After irradiation, one end of the source container was connected to the chamber, the other end was then opened to the atmosphere; thus purging the argon into the ion chamber. The amount of $^{41}$Ar released into the 14,393 cc ion chamber was

$$0.0842 \pm 7\% \mu\text{Ci}$$

and resulted in a reading of 298 mV above background. The $^{41}$Ar was allowed to decay in the chamber, and the activity was observed to have the 1.83 hour half-life characteristic of $^{41}$Ar. The instrument response was found to be linear on all ranges with a slope of $1.875 \times 10^{-8} \mu\text{Ci}^{41}\text{Ar/cc-mV}$. This calibration indicated that the MPC of $^{41}$Ar would correspond to a reading of 2.13 mV above background. Under stable conditions (natural radon concentrations relatively invariant) background was measured to within $\pm 1.0$ mV. Since in-line measurements of background vary to $\pm 1.0$ mV, the minimum detectable concentration of $^{41}$Ar, $A_m$, is determined by:

$$A = 1.875 \times 10^{-8} (m_r - m_b) \mu\text{Ci/cc},$$

where:

- $A$ is the concentration of $^{41}$Ar,
- $m_r$ is the mean of reading taken, and
- $m_b$ is the mean background reading.

$A_m$ is given when $m_r = m_b$. The associated error, $E$, is given by:

$$E = \pm \left[ (1 \text{ mV})^2 + (1 \text{ mV})^2 \right]^{1/2} = \pm 1.414 \text{ mV}$$

$$< 0.67 \times \text{MPC } ^{41}\text{Ar}.$$
Hence:

\[
A = 0.0 (\pm 2.65 \times 10^{-8}; -0.0) \mu \text{Ci}^{41}\text{Ar/cc}, \text{ or } \\
A_m = 0.67 \times \text{MPC}^{41}\text{Ar} = 2.65 \times 10^{-8} \mu \text{Ci}^{41}\text{Ar/cc}.
\]

The air flow through the ion chamber was measured by utilizing a sharp edged orifice, ASME standard pressure taps at one pipe diameter upstream from the orifice and one-half pipe diameter downstream, a mercury barometer, a water micro-manometer, and a calibrated thermometer. Calculations of air flow were obtained by using ASME standard curves (7), and the equations found in Appendix IV.

The volume of exhaust from each beam port depends upon the configuration of the beam access door, the beam tube ventilation valve (open or closed), and the volume of material in the beam tube. Each beam tube contained its standard beam plug during the quoted flow rate measurements. The volume-rate of effluent from each facility was observed to be somewhat variable with time. The flow rate measurements are estimated to be within ± 5% standard error, based upon the tolerances cited in the ASME flow measurement codes (7) and the observed fluctuations in the data. Flow rate values for several reactor facility configurations are shown in Table I.
TABLE I.  FLOW RATES

<table>
<thead>
<tr>
<th>FACILITY</th>
<th>CONFIGURATION</th>
<th>NET FLOW RATE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Argon Duct</td>
<td>Manifold - all open</td>
<td>7.7 cfm</td>
</tr>
<tr>
<td></td>
<td>Beam doors - closed</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Beam valves - closed</td>
<td></td>
</tr>
<tr>
<td>Argon Duct</td>
<td>Manifold - all open</td>
<td>12.3 cfm</td>
</tr>
<tr>
<td></td>
<td>Beam doors - open</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Beam valves - open</td>
<td></td>
</tr>
<tr>
<td>Rabbit</td>
<td>Blower on</td>
<td>55.6 cfm</td>
</tr>
<tr>
<td>Rabbit</td>
<td>Blower off</td>
<td>7.9 cfm</td>
</tr>
</tbody>
</table>
STEADY STATE $^{41}\text{Ar}$ PRODUCTION RATES

For each irradiation the reactor was brought to the desired
power in a square wave. Gamma ray spectra of the effluent from the
argon duct and rabbit system were the same. The spectra obtained are
characteristic of $^{41}\text{Ar}$ (Figure 4). Neither nitrogen-16 nor any other
isotope was detected when the spectra were corrected for background.

It was necessary for the reactor to remain at the same power
for several hours in order to approach the $^{41}\text{Ar}$ equilibrium production
rate of the argon duct effluent. The concentration, $N$, of $^{41}\text{Ar}$ in the
argon duct was observed to vary with time as given by:

$$N = N_E (1 - e^{-\lambda t}),$$  \hspace{1cm} (Eq. 1)

where $\lambda$ is the effective production constant:

$$\lambda = \frac{\ln 2}{T_{1/2 \text{ (Production)}}}.$$  

The equilibrium concentration, $N_E$, was determined by first finding
and then using $\lambda$ in (Eq. 1) as follows:

Pick two points $N_1(t_1)$ and $N_2(t_2)$, then

$$\begin{align*}
N_1 &= \frac{1 - e^{-\lambda t_1}}{1 - e^{-\lambda t_2}}, \\
N_2 &= \frac{1}{1 - e^{-\lambda t_2}}.
\end{align*}$$

$$e^{-\lambda t_2} = 1 - \frac{N_2}{N_1} (1 - e^{-\lambda t_1}).$$  \hspace{1cm} (Eq. 2)

Since (Eq. 2) is a transcendental equation, $\lambda$ was found by trial and
error. Having found $\lambda$, the value of $N_E$ is given by:

$$N_E = \frac{N_3}{1 - e^{-\lambda t_3}}.$$
FIGURE 4. GAMMA RAY SPECTRUM OF ARGON DUCT EFFLUENT AT 250 KW REACTOR POWER.
The calculated saturation concentration is compared to the extrapolated saturation concentration in Table II. Other measurements taken indicate that with the beam doors and valves closed at least 90% of the activity in the argon duct effluent is produced in the thermal column.

**TABLE II. SATURATION $^{41}$Ar CONCENTRATION IN THE ARGON DUCT**

<table>
<thead>
<tr>
<th>Reactor Power</th>
<th>$T_{1/2}$ (Production)</th>
<th>Calculated Concentration ($\mu$Ci/cc)</th>
<th>Extrapolated Concentration ($\mu$Ci/cc)</th>
<th>Relative Error</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 KW*</td>
<td>41.26 min.</td>
<td>$3.23 \times 10^{-7}$</td>
<td>$3.08 \times 10^{-7}$</td>
<td>4.6%</td>
</tr>
<tr>
<td>10 KW*</td>
<td>44.72 min.</td>
<td>$4.67 \times 10^{-6}$</td>
<td>$4.67 \times 10^{-6}$</td>
<td>0.0%</td>
</tr>
<tr>
<td>100 KW*</td>
<td>35.13 min.</td>
<td>$6.23 \times 10^{-5}$</td>
<td>$5.96 \times 10^{-5}$</td>
<td>4.3%</td>
</tr>
<tr>
<td>250 KW*</td>
<td>56.82 min.</td>
<td>$1.58 \times 10^{-4}$</td>
<td>$1.58 \times 10^{-4}$</td>
<td>0.0%</td>
</tr>
<tr>
<td>250 KW**</td>
<td>58.74 min.</td>
<td>$1.06 \times 10^{-4}$</td>
<td>$1.06 \times 10^{-4}$</td>
<td>0.0%</td>
</tr>
</tbody>
</table>

* Beam doors and valves closed.  
** Beam doors and valves open.

Presumably, due to the relatively rapid exchange of air the activity in the rabbit exhaust was observed to reach an equilibrium value almost immediately. Even with the blower off equilibrium was reached within a few minutes. Results of the equilibrium concentration measurements of $^{41}$Ar in the rabbit exhaust are displayed in Table III.
### TABLE III. SATURATION $^{41}\text{Ar}$ CONCENTRATION IN THE RABBIT EXHAUST

<table>
<thead>
<tr>
<th>Reactor Power (KW)</th>
<th>Blower Condition</th>
<th>Concentration ($\mu$Ci/cc)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 KW</td>
<td>ON</td>
<td>Not detectable</td>
</tr>
<tr>
<td>10 KW</td>
<td>ON</td>
<td>$1.35 \times 10^{-6}$</td>
</tr>
<tr>
<td>50 KW</td>
<td>ON</td>
<td>$6.98 \times 10^{-6}$</td>
</tr>
<tr>
<td>100 KW</td>
<td>ON</td>
<td>$1.49 \times 10^{-5}$</td>
</tr>
<tr>
<td>250 KW</td>
<td>ON</td>
<td>$3.34 \times 10^{-5}$</td>
</tr>
<tr>
<td>250 KW</td>
<td>OFF</td>
<td>$4.07 \times 10^{-5}$</td>
</tr>
</tbody>
</table>

A summary of the $^{41}\text{Ar}$ equilibrium production rates, given by the product of the gross flow rates and the $^{41}\text{Ar}$ concentrations, is presented in Table IV. Figure 5 shows the relative $^{41}\text{Ar}$ production rate vs. reactor power with the reactor in a "normal" operating configuration.
<table>
<thead>
<tr>
<th>Facility</th>
<th>Reactor Power</th>
<th>Production Rate* (µCi/min.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Argon Duct (Beam doors and valves closed)</td>
<td>0.1 KW</td>
<td>≤ 0.013</td>
</tr>
<tr>
<td></td>
<td>1 KW</td>
<td>0.16</td>
</tr>
<tr>
<td></td>
<td>10 KW</td>
<td>2.3</td>
</tr>
<tr>
<td></td>
<td>100 KW</td>
<td>31</td>
</tr>
<tr>
<td></td>
<td>250 KW</td>
<td>79</td>
</tr>
<tr>
<td>Argon Duct (Beam doors and valves open)</td>
<td>250 KW</td>
<td>67</td>
</tr>
<tr>
<td>Rabbit (Blower on)</td>
<td>1 KW</td>
<td>≤ 0.042</td>
</tr>
<tr>
<td></td>
<td>10 KW</td>
<td>2.1</td>
</tr>
<tr>
<td></td>
<td>50 KW</td>
<td>11</td>
</tr>
<tr>
<td></td>
<td>100 KW</td>
<td>23</td>
</tr>
<tr>
<td></td>
<td>250 KW</td>
<td>53</td>
</tr>
<tr>
<td>Rabbit (Blower off)</td>
<td>250 KW</td>
<td>9</td>
</tr>
</tbody>
</table>

* Since the standard errors associated with the $^{41}$Ar concentration and the flow rates are 7% and 5% respectively, the standard error calculated for the values listed is approximately 9%.
FIGURE 5. PRODUCTION OF $^{41}$Ar AT EQUILIBRIUM.
The resolution of the pulse-produced $^{41}\text{Ar}$ was generally good as shown in Figure 6 and Figure 7. The time delay from the pulse to the initial response due to $^{41}\text{Ar}$ was 1.6 minutes for the argon duct, 0.15 minutes for the rabbit with the blower on and 1.06 minutes for the rabbit with the blower off. The elapsed time from the pulse to the peak concentration ranged from 2.67 to 3.21 minutes in the argon duct, was 0.2 minutes in the rabbit exhaust with the blower on, and was 1.48 minutes with the blower off.

All pulses were initiated from a reactor power of 1 watt. The activity produced by the pulses was quantitized by cutting out the entire pulse minus background and weighing the paper. The weight of the paper was then compared to a standard. The values thus determined are contained in Table V. No explanation is offered for the apparent anomaly in $^{41}\text{Ar}$ production in the argon duct for a $2.50$ pulse. Figure 8 displays the data in Table V.
FIGURE 6. ARGON DUCT EFFLUENT ACTIVITY FOLLOWING A $3.00 PULSE.

FIGURE 7. COMPARISON: RABBIT EXHAUST ACTIVITY FOLLOWING A $3.00 PULSE.
TABLE V. $^{41}$Ar PRODUCTION BY PULSES

<table>
<thead>
<tr>
<th>Facility</th>
<th>Pulse</th>
<th>Production* ($\mu$Ci $^{41}$Ar/pulse)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Worth</td>
<td></td>
</tr>
<tr>
<td>Argon Duct</td>
<td>$0.80$</td>
<td>Not detectable</td>
</tr>
<tr>
<td>(Beam doors and valves closed)</td>
<td>$1.00$</td>
<td>Not detectable</td>
</tr>
<tr>
<td></td>
<td>$1.50$</td>
<td>16</td>
</tr>
<tr>
<td></td>
<td>$2.00$</td>
<td>38</td>
</tr>
<tr>
<td></td>
<td>$2.50$</td>
<td>36</td>
</tr>
<tr>
<td></td>
<td>$3.00$</td>
<td>65</td>
</tr>
<tr>
<td>Rabbit</td>
<td>$0.80$</td>
<td>Not detectable</td>
</tr>
<tr>
<td>(Blower on)</td>
<td>$1.00$</td>
<td>Not detectable</td>
</tr>
<tr>
<td></td>
<td>$1.50$</td>
<td>3.9</td>
</tr>
<tr>
<td></td>
<td>$2.00$</td>
<td>5.4</td>
</tr>
<tr>
<td></td>
<td>$2.50$</td>
<td>6.5</td>
</tr>
<tr>
<td></td>
<td>$3.00$</td>
<td>9.4</td>
</tr>
<tr>
<td>Rabbit</td>
<td>$3.00$</td>
<td>19</td>
</tr>
<tr>
<td>(Blower off)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Since the standard errors associated with the $^{41}$Ar concentration and the flow rates are 7% and 5% respectively, the standard error calculated for the values listed is approximately 9%. 
FIGURE 8. PRODUCTION OF ARGON-41 DUE TO PULSES.
DISCUSSION

The $^{41}$Ar production rates reported here are comparable to those reported at similar facilities. Due to the counting statistics of other detectors, the sensitivity of the ion chamber-electrometer was found to be more precise near the MPC for $^{41}$Ar. A comparison of $^{41}$Ar monitors and equilibrium production rates at similar reactors is contained in Table VI.

### TABLE VI. A COMPARISON OF ARGON MONITORS

<table>
<thead>
<tr>
<th>Location</th>
<th>Detector</th>
<th>Sensitivity $^{41}$Ar/cc-cpm</th>
<th>$^{41}$Ar Equilibrium Production at 250 KW</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cornell Univ. (1)</td>
<td>G-M</td>
<td>$2.1 \times 10^{-9}$ *</td>
<td>----</td>
</tr>
<tr>
<td>Oregon St. Univ.</td>
<td>ion</td>
<td>$1.875 \times 10^{-8}$ µCi/cc-mV (20 mV bkgrd)</td>
<td>88 µCi/min.</td>
</tr>
<tr>
<td>Univ. (experimental)</td>
<td>chamber</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Oregon St. Univ. (6)</td>
<td>G-M</td>
<td>$1.8 \times 10^{-8}$ (20 cpm bkgrd)</td>
<td>68 µCi/min.</td>
</tr>
<tr>
<td>Univ. of Cal. Berkeley (4)</td>
<td>NaI</td>
<td>$2.4 \times 10^{-9}$ * (100 cpm bkgrd)</td>
<td>204 µCi/min.*</td>
</tr>
<tr>
<td>Univ. of Ill. (3)</td>
<td>G-M</td>
<td>$1.2 \times 10^{-9}$ (100 cpm bkgrd)</td>
<td>13.6 µCi/min.**</td>
</tr>
<tr>
<td>Wash. St. Univ. (2)</td>
<td>G-M</td>
<td>$4 \times 10^{-9}$ * (67 cpm bkgrd)</td>
<td>85 µCi/min.*</td>
</tr>
</tbody>
</table>

* Calculated from reported data.

** Of the reactor facilities listed, the one at the Univ. of Ill. is most similar to that of Oregon State University. The volume of effluent is 1/5 that of Oregon State University's, while the $^{41}$Ar concentration released to the atmosphere is reportedly the same at both facilities.
Calculations in the Oregon State University's hazards analysis for the TRIGA reactor (8) predict that 48 μCi/minute leaves the beam tubes with the reactor power at 250 KW and the beam valves open. In this reactor configuration the $^{41}$Ar production from the beam ports, thermal column and thermalizing column was measured to be 66.7 μCi/minute. With the beam doors and valves closed, data indicates that at least 80% of the $^{41}$Ar in the argon duct is produced in the thermal column. It is postulated that the decrease in the $^{41}$Ar production rate measured while the beam doors and valves were open is primarily the result of a decreased flow rate of air through the thermal column. With the beam doors and valves open, air entering a beam tube has a somewhat direct route to the exhaust port, thus making the residence time in the neutron flux relatively short. In contrast, air entering the thermal column is likely to travel a longer and rather indirect route to the exhaust port.

Thus, with the beam doors and valves open, most of the air entering the beam tubes would have otherwise entered the thermal column. The air passing through the beam tubes is exposed to a lower average thermal neutron flux than that in the thermal column. Hence, the portion of air which otherwise would have entered the thermal column is exposed to a lower average thermal neutron flux for a shorter time. Apparently, the increased flow rate, and thus the number of
$^{40}$Ar nuclei exposed per unit time, is not great enough to override the effects of a lower nvt (time integral of flux). The flow rate through the thermal column could probably be reduced further by attaching a weather stripping-type of seal around the thermal column door.

Linear extrapolation to 1 MW gives an $^{41}$Ar equilibrium production rate exhausted in the argon duct of 316 $\mu$Ci/minute with the beam doors and valves closed and 267 $\mu$Ci/minute with the beam doors and valves open. One megawatt production rates for the rabbit are calculated to be 210 $\mu$Ci/minute with the blower on and 36 $\mu$Ci/minute with the blower off. The effective production half-life in the argon duct ranged from 35.13 to 58.74 minutes and was generally near 47 minutes.

Table V. shows that the $^{41}$Ar produced by a $3.00$ pulse with the rabbit blower off is approximately 2.02 times as much as with the blower on. In theory, the $^{41}$Ar produced by a pulse should be independent of flow rate except for small corrections for the compressability of air. Over the ranges considered, the volume flow rate is directly proportional to the rate of flow of the number of nuclei of interest per unit volume. The reactivity worth of a pulse is directly proportional to the energy released, which in turn is proportional to the nvt. Hence, if half the number of $^{40}$Ar nuclei per unit volume per unit time were exposed to $3.00$ Pulse A for twice the time as for $3.00$ Pulse B, the
amount of $^{41}$Ar produced in each case would be the same. Since the reactivity worth of the pulse is proportional to the energy release, the $^{41}$Ar produced by a pulse varies linearly with the energy released as illustrated in Figure 8.

The explanation of the observed production by pulses is as follows: Even though the response time of the detector used is very fast (1/6 second time constant), with the blower on the pulses of $^{41}$Ar activity filled the ion chamber and began to exhaust from the chamber faster than the instrument could completely record the events. The flow rates in the argon duct and in the rabbit exhaust with the blower off were low enough to neglect any losses due to the response time. By this reasoning the production values listed in Table V. for the rabbit with the blower on should be multiplied by 2.02 to obtain the true production values. Similarly, values read from the RABBIT EXHAUST (BLOWER ON) curve in Figure 8, should be multiplied by 2.02 to obtain the true value. The undetectable production of $^{41}$Ar by pulses of less than or equal to $1.00 is consistent with Fuchs-Nordheim model for reactor dynamics as discussed in the Oregon State University's hazard analysis for the TRIGA reactor (8).
CONCLUSION

Based on the data collected, the following conclusions can be made:

A. The only significant radioisotope released to the atmosphere during the normal operation of the Oregon State University TRIGA Mark III Reactor is $^{41}$Ar.

B. Under constant flow rate conditions the equilibrium production rate of $^{41}$Ar is a linear function of power.

C. The reactor configuration minimizing the steady state production of $^{41}$Ar is with the beam doors and beam valves open and the rabbit blower off.

D. The minimum $^{41}$Ar equilibrium production rate at 250 KW reactor power is 76 $\mu$Ci/minute. By linear extrapolation, at 1 MW reactor power the minimum $^{41}$Ar equilibrium production rate is predicted to be 303 $\mu$Ci/minute.

E. The $^{41}$Ar produced by a pulse due to a reactivity insertion greater than $1.00 is a linear function of the pulse worth.

F. The $^{41}$Ar produced by a pulse due to a reactivity insertion less than or equal to $1.00 is negligibly small.

G. A $3.00 pulse produces 85 $\mu$Ci $^{41}$Ar, under normal conditions, which is released to the atmosphere.
H. A sensitive ion chamber-electrometer system may be used as an efficient and reliable means of detecting $^{41}$Ar in air.
BIBLIOGRAPHY


7. The American Society of Mechanical Engineers. Flow measurement by means of thin plate orifices, flow nozzles and venturi tubes. 1959. 91p. (ASME Power Test Codes, Supplement on Instruments and Apparatus, Part 5., Chapter 4. PTC 19.5; 4-1959)

APPENDICES
APPENDIX I

CONSTANTS USED IN CALCULATIONS

1. Avogadro constant (A4): \( N_A = 6.02252 \pm 28 \times 10^{23} \text{ atoms per gm at. wt.} \)

2. Gas constant (A8): \( R = 8.31432 \pm 0.00034 \times 10^7 \text{ erg per g mole deg} \)

3. Half life of \(^{41}\text{Ar}\): \( T_{1/2} (^{41}\text{Ar}) = 110 \text{ min} \)

4. Half life of \(^{198}\text{Au}\) (A5): \( T_{1/2} (^{198}\text{Au}) = 64.63 \pm .11 \text{ hr} \)

5. Natural logarithm of 2: \( \ln 2 = .693147 \)

6. NBS standard flux (A7): \( \Phi_{\text{NBS}} = 4193 \pm 2\% \text{ neutrons/cm}^2\text{-second} \)

7. Nuclidian mass of \(^{197}\text{Au}\) = 196.96654 (A1)

8. Relative isotopic abundance of \(^{40}\text{Ar}\) (A3): \( f_{40} = .9959899 \)

9. Thermal activation cross section for \(^{40}\text{Ar}\) (A2): \( \sigma_{40} = 0.723 \pm 0.025 \text{b} \)

10. Thermal activation cross section for \(^{197}\text{Au}\) (A6): \( \sigma_a = 98.4 \pm 0.5 \text{b} \)
REFERENCES


APPENDIX II

THERMAL NEUTRON FLUX CALIBRATION

If a stable gold foil containing N nuclei/cm$^3$, each with a thermal activation cross section, $\sigma_a$, is exposed to a known neutron flux, $\Phi$, for a time $t_r$, the resulting activity is given by:

$$ A = N\sigma_a \Phi (1-e^{-\lambda t_r}), \text{ where } \lambda = \frac{\ln 2}{T_{1/2}(^{198}\text{Au})}. $$

If the activity decays for a time, $t_w$, and produces $C^{th}$ counts due to thermal activation during a counting time, $t_C$, then the counting efficiency, $k$, is:

$$ k = \frac{C^{th} \lambda}{N\sigma_a \Phi (1-e^{-\lambda t_r}) e^{-\lambda t_w} (1-e^{-\lambda t_C})}, \hspace{1cm} (\text{Eq. A-1}) $$

where:

$$ C^{th} = C \left( \frac{R-1}{R} \right), \text{ if } $$

$$ C = \text{total counts obtained and} $$

$$ R = \text{cadmium ratio} = \frac{\text{Activity due to resonance plus thermal neutrons}}{\text{Activity due to resonance neutrons}} $$

When identical counting conditions are used, an unknown flux may be compared with a reference flux by writing (Eq. A-1) for each case and equating.

The flux inside the argon irradiation container was compared to the NBS standard flux in the manner stated above. Solving for $\Phi_{\text{AGN}}^{th}$:
\[ \phi_{th}^{AGN} = (B - D)K, \text{ where:} \]

\[ B = C_B^{AGN} \left[ m (1 - e^{-\lambda t_r}) e^{-\lambda t_w} (1 - e^{-\lambda t_c}) \right]^{-1}_{BARE} \]

\[ D = C_D^{AGN} \left[ m (1 - e^{-\lambda t_r}) e^{-\lambda t_w} (1 - e^{-\lambda t_c}) \right]^{-1}_{Cd} \]

\[ K = \left\{ \phi_m (1 - e^{-\lambda t_r}) e^{-\lambda t_w} (1 - e^{-\lambda t_c}) \left[ C_{NBS} \left( \frac{R - 1}{R} \right) \right]^{-1} \right\}_{NBS} \]

Again \( C \) represents the total number of counts obtained from the indicated foil. The foil mass is given by \( m \).

The error of the flux measurements was conservatively determined in the following way:

A. Errors in time measurements were considered negligibly small.

B. Since multiplying each of \( N \) values by the same constant multiplies the mean by this constant and the variance by this constant squared, the variance, \( \sigma^2 \), of each net count was found by:

\[ \sigma^2 = t g^2 \left( \frac{C_g}{t g^2} + \frac{C_b}{t b^2} \right), \]

where \( C \) indicates the count,

\( t \) indicates the count time,

\( g \) denotes the gross assay (i.e. sample plus background),

\( b \) denotes the background assay.
C. The variance of each quotient was determined by using the same reasoning found in B, in conjunction with the mean values of the constants incorporated. These variances were added together to obtain the variance of the difference.

D. The mass values were estimated to be within ± 50 µg m (less than 0.008% relative error).

E. The error associated with the product of saturation, decay, and counting time corrections was estimated by comparing the product using λ plus one standard error with the value obtained by using the mean λ. The relative error of the two products was less than 0.33%.

F. The largest credible deviation from (B - D) using the mean values of the constants was estimated to be less than (1.0034) (B - D) ± (1.0033) (1.00008) (B - D). Assuming that the difference using the means was in error by a factor of 1.0034, the maximum variance for the difference may be found by again using the same reasoning as in B.

G. The values of $\Phi_{th}^{AGN}$ were maximized by adding one standard error to both the difference (B - D) and $\Phi_{NBS}^N$ and subtracting one standard error from $C_{NBS}$. The errors of other factors were considered negligible.

H. The product of the factors in G was found to lie within
2.3% of the product obtained using the mean values, i.e. the calculated values of $\phi_{\text{th}}^\text{AGN}$ lie within 2.3% of the calculated values.

I. The confidence level of the error determined in $H$ was taken to be $0.318 \pm (0.6826)^3$, where 0.6826 is the confidence level of the standard error. From normal tables, one finds that a confidence level of 0.318 corresponds to $0.41 \sigma$.

The standard error of the flux calculation was then found to be $\sigma_\phi = \frac{2.3\%}{0.41} \approx 6.3\%$

Since the same foils and cadmium covers which were used to measure the AGN-201 flux were calibrated at the NBS standard pile, no correction was needed for self absorption. The average values of the cadmium ratios were:

\[
\begin{align*}
R_{\text{AGN}} &= 3.76 \\
R_{\text{NBS}} &= 8.01.
\end{align*}
\]
Let $f_i$ be the relative abundance of the $i$th isotope and $d$ be the atomic density ($\text{atoms/cm}^3$) of argon. Then from the ideal gas law

$$f_i d = \frac{f_i N_A P}{RT}$$

is the isotopic density of the $i$th isotope.

If one ignores the contribution by successive captures of $^{36}$Ar, the rate of change of the number of $^{41}$Ar nuclei per cc, $N_{41}$ is given by:

$$\frac{dN_{41}}{dt} = (a - f_{40} d \sigma_{40}^2 \Phi^2) t - \frac{a}{\lambda_{39}} (1 - e^{-\lambda_{39}t}) + f_{40} d \sigma_{40} \Phi - E N_{41}.$$

Where:

$$a = \frac{f_{38} d \sigma_{38} \sigma_{39} \alpha_{40} \Phi^3}{\lambda_{39}}, \text{ and}$$

$$E = \lambda_{41} + \alpha_{41} \Phi.$$

Solving for $N_{41}$ gives:

$$N_{41} = \left( \frac{a}{E^2} - \frac{f_{40} d \sigma_{40}^2 \Phi^2}{E^2} \right) (e^{-Et} + Et - 1)$$

$$+ \frac{a}{\lambda_{39}^2 - \lambda_{39} E} \left( e^{-Et} e^{-\lambda_{39}t} \right)$$

$$+ \left( \frac{f_{40} d \sigma_{40} \Phi}{E} - \frac{a}{\lambda_{39} E} \right) (1 - e^{-Et}).$$
Real values of the constants reveal that

\[ N_{41} \approx \frac{f_{40} d\sigma_{40}}{E} (1 - e^{-Et}). \]

The activity of $^{41}$Ar produced is then

\[ ^{41}\text{Ar} = \lambda_{41} N_{41} V \text{ (dis/sec)} \]

Since no error was observable in the measurements of $P$ and $V$, and since $T$ was measured with an NBS calibrated thermometer, the error in the above calculation lies mostly in the uncertainty of the thermal flux, viz. 6.3% (standard error). Therefore, it is felt that the calibration of the $^{41}$Ar source has a standard error of less than 7%.
FLOW MEASUREMENTS

The flow measurements were calculated as follows:

A. The absolute static pressure of the air, \( p \), was determined by summing the upstream static pressure and the barometric pressure (in lbs/in\(^2\)).

B. The specific weight, \( \gamma \), of the air at the measured temperature and pressure was found by:
\[
\gamma = \frac{\gamma_{air} \rho}{14.6967},
\]
where \( \gamma_{air} \) is the specific weight of air at 29.92 in. Hg.

C. The ratio of the orifice diameter to the pipe diameter, \( \beta = \frac{d}{D} \), was determined. By finding the differential pressure, \( h \) (in. H\(_2\)O), divided by the absolute static pressure, \( p \), the coefficient of expansion, \( Y \), was found. For the measurements made, \( Y = 1 \).

D. The flow coefficient, \( K \), was found by iteration. An arbitrary Reynolds number, \( R_D \), was picked and from tables an associated flow coefficient was found which corresponds to \( \beta \) and \( R_D \). The weight rate of flow is given by:
\[
w = 359 KYd^2 (h\gamma)^{1/2} \text{ lbs./hr},
\]
where \( d \) is the orifice diameter in inches.
The viscosity of air at the measured temperature, $g_{\mu}$, was found, and a new Reynolds number was computed from the equation:

$$R_D = \frac{0.004244 w}{\pi D g_{\mu}}$$  \hspace{1cm} (Eq. A-2)

where $D$ is the pipe diameter. Using the computed value of $R_D$, a new value of $K$, and hence $w$, was found. The new value of $w$ was substituted into (Eq. A-2) and another $R_D$ and hence $K$ was found, and similarly until $K$ no longer changed.

E. The volume rate of flow, $Q$, was found using the final $w$ obtained in D. and the equation

$$Q = \frac{w}{60\gamma} \text{ cfm.}$$

Note that $Q$ was not found in standard cfm. This is due to the fact that the MPC is defined in terms of cubic centimeters and not standard cubic centimeters of air.