Analysis of the Gas-Stack Monitor Calibration Data for the Diamond Ordnance Radiation Facility

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In order to effectively monitor the release of radioactive argon-41 gas from the DORF site, it is necessary to periodically calibrate the monitoring equipment. This report reviews the analysis of the calibration case.
CONTENTS

1. INTRODUCTION .................................................. 5
2. BACKGROUND .................................................... 5
3. DATA PROCESSING PROCEDURE ................................. 9
   3.1 Handling the Raw Data ................................. 9
   3.2 Details of Data Processing .......................... 11
      3.2.1 Input ........................................... 11
      3.2.2 Output ........................................ 12
APPENDIX A. Argon Program ................................. 13
APPENDIX B. Argon User's Guide ............................. 17
APPENDIX C. Argon Output ................................. 19
DISTRIBUTION .................................................. 21

FIGURES

1   Diamond Ordnance Radiation Facility, main building with
    a view of the gas stack .................................. 6
2   Vertical section of the DORF reactor ..................... 6
3   Perspective view of DORF reactor ....................... 7
4   Gas-stack monitor .......................................... 8
5   Diagram of the argon-41 flow system during calibration .. 9
6   Trace of the gas-stack monitor data ...................... 10
1. INTRODUCTION

Radioactive argon gas (A-41) is produced when naturally occurring A-40 captures a neutron. Since air contains approximately 1 percent A-40, neutron irradiation of air produces a measurable quantity of A-41, which decays with a half-life of 110 min.

At the Diamond Ordnance Radiation Facility (DORF), a research reactor is operated adjacent to a large air-filled volume called the exposure room. The irradiated air, when exhausted from the room through a gas stack, contains a measurable quantity of A-41. To be certain that this radioactivity does not represent a radiation hazard to the surrounding environment, the release of A-41 must be measured and reported annually as required by Army Regulations 385-80.

To assist in the analysis of the amount of argon released, a computer program has been written to take the data recorded on the gas-stack monitor and calculate the conversion constant between counts per minute on the monitor and the number of curies of A-41 released to the environment. The procedure for obtaining the experimental input data is given in a memorandum by T. Chichester. This report documents the procedure used to process and record the data for the gas-stack monitor calibration.

2. BACKGROUND

The DORF reactor is a TRIGA-type reactor capable of operating in a pulsed or steady-state mode. The reactor is movable and can operate either completely shielded by water or immediately adjacent to an exposure room. During operation next to the exposure room, air (which is flowing through the room) is irradiated and vented to the outside through the gas stack (fig. 1, 2, 3). Several radioactive effluents are produced; however, A-41 is the only important one, because the others have sufficiently short half-lives that they decay before leaving the stack.

Located near the lower end of the air stack is the gas-stack monitor. This system (fig. 4) continuously samples the air in the stack. The sampled air is pumped into a chamber containing a Geiger tube. A logarithmic circuit accepts this output voltage and converts it

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1Procedure for Calibrating the Gas-Stack Monitor, Health Physics Section, Walter Reed Army Medical Center.
Figure 1. Diamond Ordnance Radiation Facility, main building with a view of the gas stack.

Figure 2. Vertical section of the DORF reactor.
to a logarithm output that drives a count rate meter and a strip-chart recorder. It is necessary to relate the trace of the recorder, in units of counts per minute, to the number of curies per cubic centimeter in the air sampling chamber. When this relationship is known, the number of curies per minute released is found by multiplication with the stack flow rate.

The process for finding the conversion constant between counts per minute and curies per cubic centimeter begins by irradiation of a 210-cc flask of 99.97 percent pure argon. This irradiation produces a high concentration of A-41. A second, unirradiated flask of A-40 is placed in the gas-stack monitor system in series with the irradiated flask by means of tubing (fig. 5). The contents of both flasks are pumped through the gas-stack monitor by a varistaltic pump to allow
Figure 4. Gas-stack monitor.

equalization of the A-41 concentration throughout the system. When equilibrium is attained, the irradiated flask is sent to the Health Physics Office, Walter Reed Army Medical Center, for radio-analysis. The other flask is left connected to the gas-stack monitor.
The sample is analyzed at the Health Physics Office. A report sent to DORF gives the concentration of A-41 in the flask in microcuries per cubic centimenter at a given time. After this concentration is corrected for decay to a predetermined time, it is possible to correlate the count rate on the gas-stack monitor with the concentration of the A-41 in the sample chamber.

3. DATA PROCESSING PROCEDURE

3.1 Handling the Raw Data

When the unirradiated flask is sent to the Health Physics Office, the gas-stack monitor, still connected to the irradiated flask, is left running for about 18 hr. In this time, the logarithmic trace of the recorder comes on scale at 100,000 cpm and goes through a number of half-lives before it blends with the background activity (fig. 6). A straight line is drawn from the point at which the trace comes on scale to the low count rate end where the trace starts to become nonlinear (because of the effects of background). If the line has the correct
DISTANCE BETWEEN COUNT RATES IS 110 min, T 1/2 of $^{41}$Ar

Figure 6. Trace of the gas-stack monitor data.
slope (i.e., exhibits the 110-min decay of A-41), after the data have been fitted to a straight line by the method of least squares, there is no indication of a leak in the system, and the calibration is assumed to be valid.

If the slope is not correct, the cause must be found, and the procedure must be repeated. Possible causes are leaks in the system and inadequate background compensation. The time at which the trace comes on scale is defined as time zero. The sample from the Health Physics Office is decay corrected to time zero. Then starting at time zero, the count rate is taken at 110-min intervals (i.e., every half-life), and the ratios of A-41 concentration to the count rate are calculated. These data are taken until nonlinearity becomes a problem. Both the average value of the ratios and their RMS deviations are determined for the first four (i.e., most statistically significant) data points.

3.2 Details of Data Processing

The computer program ARGON that performs the analysis is currently stored in the DORF library on time-share computer at the Applied Physics Laboratories in Columbia, MD. The program (app A) is written in the Conversational Programming System (CPS).

To execute the program, the user must first call the remote-terminal port of the computer and log on with a valid account number. When the computer answers with "READY," the directive "CPS" is given, telling the computer what working language is being used. Then, the user can begin processing data through the teletype in accordance with the following paragraphs. An example is given in appendix B.

3.2.1 Input

To begin, the program is transferred from the user library with the LOAD (ARGON) command. A question mark follows, and the user types "XEQ." Execution of ARGON begins at this time.

The first input, to be entered after "DATE(1):" is typed, is the date on which the calibration was made, entered as day, month, and year. Next (after "NUM:" is printed), the number of half-lives measured is entered. In the example shown in appendix A, eight half-lives were measured. The concentration of A-41 in the assayed flask is the next input parameter. The concentration in microcuries per cubic centimeter, decay corrected to time zero, is entered following "MICRO:". The remaining values to be entered are the count rate from zero time, "COUNT (0)," to the last half-life value, which is "COUNT (8)" in the example. The count rate at zero time is always 100,000 cpm with the present system, and the remaining count rates are found by measurement of 110-min intervals from zero time (fig. 6).
3.2.2 Output

The analysis (app C) follows the input of count rates. All input data are listed, as well as the following information:

a. A determination of the half-life and count rate at zero time by the method of least squares

b. The error in the half-life

c. The concentration of A-41 at each half-life

d. The calculated ratio of microcuries per cubic centimeter per counts per minute at every half-life

e. The average ratio of the first four values given in d

f. The RMS variation of the first four ratios.

The output shown in appendix C is from the calibration run of 27 August 1974. Results show that there were no leaks in the system; however, results show also inadequate background compensation. This is indicated by an increase in the calculated half-life by 3.5 percent after five half-lives. The low RMS error shown indicates the value of the better statistics of the first four points.
APPENDIX A.—ARGON PROGRAM

/* ARGON */
/* AN ARGON-41 ANALYSIS PROGRAM */
DECLARE DATE(3), COUNT(0:10);
DECLARE DD(0:10), DR(0:10);
SUMX, SUMXY, SUMX2 = 0;
SUMDEL, SUMRAT = 0;
PUT LIST('ENTER THE DAY/MONTH/YEAR');
GET LIST(DATE);
PUT LIST('ENTER THE NUMBER OF HALF LIVES MEASURED');
GET LIST(NUM);
PUT LIST('ENTER THE ASSAYED CONCENTRATION OF ARGON-41 I N');
PUT LIST('MICRO-CURIES/CC CORRECTED FOR THE 110 MIN. HA E');
PUT LIST('LIFE TO TIME=0 OF THE SERIES OF COUNTS');
GET LIST(MICRO);
PUT LIST('ENTER THE INITIAL COUNTS AT ZERO HALF LIVES');
GET LIST(COUNT(0));
PUT LIST('FINALLY, ENTER THE COUNTS AT EACH FOLLOWING
ALF LIFE');
T = 110;
TIME = 0;
N = NUM + 1;
SUMY = Log(COUNT(0));
SUMY2 = SUMY*SUMY;
DO I = 1 TO NUM;
GET LIST(COUNT(I));
TIME = TIME*T;
Y = Log(COUNT(I));
SUMA = SUMA*TIME;
SUMY = SUMY*Y;
SUMAY = SUMAY + TIME*Y;
SUMA2 = SUMA2 + TIME*TIME;
SUMY2 = SUMY2 + Y*Y;
END;
DEN = SUMA**2 - N*SUMA2;
AI = (SUMA*SUMY - N*SUMXY)/DEN;
TEMP = -1*(SUMX2*SUMY - SUMX*SUMXY)/DEN;
AO = EXP(TEMP);
DO I = 0 TO NUM;
D = MICRO/2**I;
RATIO = D/COUNT(I);
? SUMRAT = SUMRAT + RATIO;
DD(I) = D*10000;
DR(I) = RATIO*.1E09;
END;
XBAR = SUMRAT*.1E09/N;
PUT EDIT('') (SKIP(8), F(0));
PUT LIST('LEAST SQUARES ANALYSES OF ARGON-41 CALIBRATION N');
PUT LIST('- - - - - - - - - - - - - - - - - - - - - - - - - - - - - - - - - -')
13
APPENDIX A

- ');

245. PUT LIST(' ');

250. PUT LIST('DATE=',DATE(1),'/'DATE(2),'/'DATE(3));

255. PUT LIST(' ');

260. PUT LIST('HP VALUE OF CONCENTRATION=',MICRO,'MICROCI./C PER CPM');

265. PUT LIST(' ');

270. PUT LIST('INPUT DATA ARE AS FOLLOWS: ');

275. PUT LIST('NO. TIME(MIN) COUNTS/MIN. ');

280. DO I=0 TO NUM;

285. PUT IMAGE(I,T*I,COUNT(I))(IME);

290. END;

295. PUT LIST(' ');

300. PUT LIST('SLOPE AND INTERCEPT OF ARGON-41 DECAY CURVE A ');

305. PUT LIST(' ');

310. PUT IMAGE(-LOG(2)/A1)(IMB);

315. PUT LIST(' ');

320. PUT IMAGE(A0)(IMC);

325. PUT LIST(' ');

330. ER=ABS((LOG(2)/A1+T)/T)*100;

335. PUT IMAGE(ER)(IMD);

340. PUT LIST(' ');

345. PUT LIST(' ');

350. PUT LIST('CALIBRATION CONSTANTS FOR THIS RUN ARE AS FOL');

355. PUT LIST(' ');

360. PUT LIST(' ');

365. PUT LIST(' ');

370. PUT LIST(' OBSERVED CALCULATED ACTIVITIES RA IDS ');

375. PUT LIST(' COUNTS (X 10^-4) (X 10^-8) ');

380. PUT LIST(' ');

385. DO I=0 TO NUM;

390. PUT IMAGE(COUNT(I),DD(I),DR(I))(IMA);

395. END;

400. XBAR=(DR(0)+DR(1)+DR(2)+DR(3))/4;

405. DO I=0 TO 3;

410. SUMDEL=SUMDEL+(XBAR-DR(I))*2;

415. END;

420. RMS=SQRT(SUMDEL/3)/XBAR;

425. PUT LIST(' ');

430. PUT LIST(' FOR THE MOST SIGNIFICANT DATA POINTS ');

435. PUT LIST(' ');

440. PUT IMAGE(XBAR)(IMF);

445. PUT LIST(' ');

450. PUT IMAGE(RMS*100)(IMG);

455. PUT EDIT('')(SKIP(8),F(0));

460. IMA: IMAGE;

465. IMB: IMAGE;
CALCULATED HALF LIFE = ---- MIN
INTERCEPT = --------- COUNTS
ERROR IN HALF LIFE = ------ PERCENT
AVERAGE RATIO = ------- MICROCURIES/CC/CPM
RMS VAR. IN RATIO = ------- PERCENT
STOP ;
APPENDIX B.—ARGON USER'S GUIDE

NEW
ENTER THE DAY, MONTH, YEAR
DATE(1)
-27 8 74
ENTER THE NUMBER OF HALF LIVES MEASURED
NUM
-8
ENTER THE ASSAYED CONCENTRATION OF ARGON-41 IN MICRO-CURIES/CC CORRECTED FOR THE 110 MIN. HALF LIFE TO TIME=0 OF THE SERIES OF COUNTS
MICRO
-0.00265
ENTER THE INITIAL COUNTS AT ZERO HALF LIVES
COUNT(0)
-100000
FINALLY, ENTER THE COUNTS AT EACH FOLLOWING HALF LIFE
COUNT(1)
-50000
COUNT(2)
-26000
COUNT(3)
-12500
COUNT(4)
-6500
COUNT(5)
-3500
COUNT(6)
-1900
COUNT(7)
-900
COUNT(8)
-450
APPENDIX C.—ARGON OUTPUT

LEAST SQUARES ANALYSES OF ARGON-41 CALIBRATION

DATE= 27 / 8 / 74

HP VALUE OF CONCENTRATION= .00268 MICROCI./CC PER CPM

INPUT DATA ARE AS FOLLOWS:

<table>
<thead>
<tr>
<th>NO.</th>
<th>TIME(MIN)</th>
<th>COUNTS/MIN.</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
<td>100000</td>
</tr>
<tr>
<td>1</td>
<td>110</td>
<td>50000</td>
</tr>
<tr>
<td>2</td>
<td>220</td>
<td>26000</td>
</tr>
<tr>
<td>3</td>
<td>330</td>
<td>12500</td>
</tr>
<tr>
<td>4</td>
<td>440</td>
<td>6500</td>
</tr>
<tr>
<td>5</td>
<td>550</td>
<td>3500</td>
</tr>
<tr>
<td>6</td>
<td>660</td>
<td>1900</td>
</tr>
<tr>
<td>7</td>
<td>770</td>
<td>900</td>
</tr>
<tr>
<td>8</td>
<td>880</td>
<td>450</td>
</tr>
</tbody>
</table>

SLOPE AND INTERCEPT OF ARGON-41 DECAY CURVE ARE

CALCULATED HALF LIFE= 113.9 MIN

INTERCEPT= 98025 COUNTS

ERROR IN HALF LIFE= 3.526 PERCENT

CALIBRATION CONSTANTS FOR THIS RUN ARE AS FOLLOWS:

<table>
<thead>
<tr>
<th>OBSERVED COUNTS</th>
<th>CALCULATED ACTIVITIES (X 10^-4)</th>
<th>RATIOS (X 10^-8)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100000</td>
<td>26.8000</td>
<td>2.6800</td>
</tr>
<tr>
<td>50000</td>
<td>13.4000</td>
<td>2.6800</td>
</tr>
<tr>
<td>26000</td>
<td>6.7000</td>
<td>2.5769</td>
</tr>
<tr>
<td>12500</td>
<td>3.3500</td>
<td>2.6800</td>
</tr>
<tr>
<td>6500</td>
<td>1.6750</td>
<td>2.5769</td>
</tr>
<tr>
<td>3500</td>
<td>0.8375</td>
<td>2.3929</td>
</tr>
<tr>
<td>1900</td>
<td>0.4187</td>
<td>2.2039</td>
</tr>
<tr>
<td>900</td>
<td>0.2094</td>
<td>2.3264</td>
</tr>
<tr>
<td>450</td>
<td>0.1047</td>
<td>2.3264</td>
</tr>
</tbody>
</table>

FOR THE MOST SIGNIFICANT DATA POINTS

AVERAGE RATIO= 2.654 MICROCURIES/CC/CPM

RMS VAR. IN RATIO= 1.942 PERCENT