MASTER

FORTRAN II DATA REDUCTION PROGRAM
FOR GAS PHASE RADIOLYSIS STUDIES

by

G. J. Werkema

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

THE DOW CHEMICAL COMPANY
ROCKY FLATS DIVISION
P. O. BOX 888
GOLDEN, COLORADO 80402
U. S. ATOMIC ENERGY COMMISSION
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Printed in USA. Price \$2. Available from the Clearinghouse for Federal
Scientific and Technical Information, National Bureau of Standards,
U. S. Department of Commerce, Springfield, Virginia

3 November 1965

RFP-624

UC-32 MATHEMATICS AND
COMPUTERS

TID-4500 (43rd Ed.)

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ACKNOWLEDGMENTS

The need for a program such as this was brought to our attention by J. R. Seed, who has been of assistance in the design and testing of this program. We are grateful to G. G. Risley and G. VanTrump for programming advice.

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ABSTRACT

A computer program is described whose purpose is to convert analytical data obtained at several times during a gas phase radiolysis experiment into the molar and molecular quantities more useful to the chemist. These quantities are presented in tabular form for each gas, along with the computed radiation dose, as a function of time.

INTRODUCTION

The RADF* computer code was written to perform the reduction of analytical data arising from gas phase radiolysis experiments. It is assumed that the radiolytic reaction takes place in a fixed volume and that small samples of the gas mixture are periodically withdrawn for analysis. The results of the chemical analysis must be converted to molecular quantities which are subsequently plotted to determine a "G-value" for the reaction.

The calculations that are required to convert gas sample analysis data, usually presented in volume percents for each gas, to the molar and molecular quantities (that are of interest to research chemists) are neither difficult nor complex. They are, however, tedious, time-consuming, and subject to error since the quantities involved are very small. In addition, for accurate work it is necessary to account for small amounts of gas that are removed with each sampling. Although the sample quantities are very small, they do accumulate, and during the course of a reaction may amount to several percent of the remaining reactants. For each sampling it is then necessary to know the total quantity of each gas present, which is equal to the measured quantity at that sample-time plus the amount of that gas removed in all previous samplings. One usually introduces a quantity of a noble gas to the reaction mixture as an indicator for the accounting procedures and for system leakage. If the data are treated properly, and

* RADF - Abbreviation for RADiOLYSIS Fortran

if the system is not leaking, the computed number of molecules of the noble gas should remain constant within analytical error for the duration of the experiment.

It is also useful to know the amount of radiation to which the reaction system has been exposed at each sampling period. This program computes radiation dose as a function of time for each sampling in addition to performing the data reduction calculations.

MATHEMATICAL METHODS

The computations are carried out in floating point arithmetic. The total number of moles of all gases in the reaction vessel at any time (t_j) is computed using the equation of state of an ideal gas: $n_j = \frac{P_j V}{RT_j}$. The number of moles of each gas ($n_{j,i}$) is computed from the

expression $n_{j,i} = n_j \cdot \left(\frac{V_i}{V}\right)_j$, where $\left(\frac{V_i}{V}\right)_j$ is the volume fraction of gas (i) in the reaction at t_j . The number of moles of each gas ($S_{j,i}$) in the sample volume (V_s) is given by $S_{j,i} = n_{j,i} \cdot \frac{V_s}{V}$.

Finally, the total number of moles of each gas ($N_{j,i}$) is $N_{j,i} = n_{j,i} + \sum_1^j S_{j,i}$.

The number of molecules of each gas ($\mathcal{N}_{j,i}$) is obtained from the expression $\mathcal{N}_{j,i} = N_{j,i} \times 6.023 \times 10^{23}$. The change in molecular population from zero time is given by the expression $\Delta \mathcal{N}_{j,i} = \mathcal{N}_{j,i} - \mathcal{N}_{1,i}$ where $\mathcal{N}_{1,i}$ is the initial population of gas i.

The computation of radiation dose is likewise direct. $(\text{Dose})_j = \frac{A_0 \cdot \bar{E} \cdot C [1 - \exp(-\lambda t_j)]}{\lambda}$, where

A_0 = activity of source at the start of the experiment,

$\bar{E}$ = average energy of emission from source,

λ = decay constant for source = $\log_e 2/t_{1/2}$, and

t_j = elapsed time from the beginning of the experiment to the jth sampling.

$C = 1.332 \times 10^{20}$, and converts Dose to ev. C is a product of sec/hr.d/curie , and ev/Mev .

In this program A_0 is dimensioned in curies, $\bar{E}$ in Mev, λ in (hours) $^{-1}$, and t_j in hours.

The program calls one special library subroutine, EXPF(a). This is normally available in most FORTRAN II libraries.

FORTRAN NOTATION FOR MATHEMATICAL SYMBOLS

<u>Symbol</u>	<u>Fortran Label</u>
n_j	ALL
P_j	P
V	V
T_j	T
$n_{j,i}, N_{j,i}$	GASMOL (J,I)
$\left(\frac{V_i}{V}\right)_j \cdot 100$	GASVP (I)
$\sum_{i=1}^j S_{j,i}$	SUMSAM (I)
t_j	Time (J)
V_s	SV
$(Dose)_j$	DOSE (J)
A_o	AZRO
$\bar{E}$	EBAR
λ	FLAM
number of gases, $1 \leq i \leq k$	I, K (I is running)
number of samples, $1 \leq j \leq l$	J, L (J is running)
$x_{j,i}$	AVOGAS (J,I)
$\Delta x_{j,i}$	DELMCL (J,I)

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RUNNING TIME

The time required to process a set of data is dependent on the amount of data and on the computer configuration. Data for 10 gases and 12 sample-times (120 items) required about 3 minutes on an ASI-210 machine with paper tape I/O.

INPUT

The traditional approach to an explanation of the program input consists of a statement of the contents of each line (or card) and the necessary format. We are attempting an improvement on this formal procedure. Included with this write-up are two data coding forms, with decimal points and field breaks in place. One should be able to fill out this form directly, hand it to a keypunch operator, and have the input material in a minimum time.

RADD* COVER

SHEET ONE, APPENDIX I

This sheet contains the data which remain constant during the entire experiment, and which belong at the beginning of a data set. The field format specification is shown in parentheses after each datum description.

* RADD - Abbreviation for RADiolysis Data

Line 1. Title - may be any alphabetic and/or numeric information, but no more than 70 characters (10A7).

Line 2. Reaction Volume (cc.) - the volume of the reaction vessel in cubic centimeters. (F8.2).

Number of Samples, L - the total number of times that samples were withdrawn for analysis, $1 \leq L \leq 12$. L is equal to the number of data sets entered on the sheets labeled RADD, Sheet 2, Appendix I. (I3).

Number of Gases, K - the total number of different gases for which analyses were made, $1 \leq K \leq 15$. The value of K is equal to the number of formulae entered in Lines 4-18. (I3).

Sample Volume (cc.) - the volume of the sample vessel in cubic centimeters (F5.2).

Line 3. A_0 - the activity of the source at the start of the experiment, in curies. (F6.2).

$\bar{E}$ - the average energy of emission from the source, in Mev. (F5.3).

λ - the decay constant for the source, in hr^{-1} . $\lambda = \log_e 2/t_{1/2}$. Because of the wide range of values λ may assume, the program calls for the exponential format (E-conversion) for this constant. If $\lambda = 2.1 \times 10^{-4}$, the proper format is .210E-03. (E9.3).

Lines 4-18. Gas Formulae, in order of data presentation - exactly K lines of alphabetic and/or numeric information, no more than five characters per line. Each line should contain the formula or some identifying label for one gas. For example, if there are eight gases involved in the experiment, then only eight formulae may be entered. These will occupy lines 4-11, ignoring 12-18. (A5).

RADD

SHEET TWO, APPENDIX I

This sheet contains the data which derive from the analysis of each sampling. The program will accept data from exactly J analyses, in order of the time of sample taking.

Line 1. Pressure (mm. Hg) - the pressure reading at time of sampling, in millimeters of mercury. (F7.2).

Temperature (deg. C.) - the temperature reading at the time of sampling, in degrees centigrade. The program converts the centigrade value to absolute for use in the ideal gas law. (F6.2).

Time (hrs.) - the hours elapsed from the beginning of the experiment to the time of the sampling. The values entered for Time increase sequentially through successive samplings. (F7.1).

Line 2. Volume Percent Data in order of list on cover -

these are the analytical results in percentages by volume for the gases in the order of the list on RADD COVER. The sequence of volume percent data must agree with the sequence of gas names on RADD COVER. That is, if the first gas name listed is CH₄ (CH₄), the volume percent figure in Line 2 must be the value for CH₄, etc. Correspondence between the two lists must be maintained for valid identification. (F5.2). The input for each computation consists of one RADD COVER sheet followed by J RADD sets. Each RADD sheet contains spaces for two sets. A sample input is presented on pages 13 and 14.

OUTPUT

The program generates three pages of output. The first line of output on each page is the title, reproduced exactly as entered. This feature is useful for identification of the experiment with the computed results. Page one (p.15*) contains a table of the micromolar quantities of each gas. Page two (p.16*) contains a table of the molecular populations of each gas. Page three (p.17*) contains a table of the changes in molecular populations compared with the initial populations. A positive value for a population change indicates an increase in the number of molecules compared with the initial population.

* Page no. of this report.

Each column is headed by the elapsed time, t_j , and the computed dose, $(\text{Dose})_j$, appropriate to the columns of answers. Each row of answers is preceded by the gas formula appropriate to that row. Each table may contain not more than 15 rows (K) and 6 columns (L) of answers. If L is greater than 6 the output will consist of two tables each for the molar and molecular quantities. Output is terminated by the comment END OF JOB which appears at the end of the last table. A page control feature has been included with the program which automatically places the three sets of tables on separate pages. A sample output is presented on pages 15-17.

OPERATION

The program in its present form is written in ASI-210 FORTRAN II, with input-output device specifications for paper tape.

The ASI-210 FORTRAN II compiler permits "load-and-go" operation. It is a one-pass system; the FORTRAN source tape is loaded and read, after which the data may be loaded.

The program tests sense switch 2 at the end of each job. If sense switch 2 is OFF, the program branches to its beginning to reinitialize and read data for a new job. If sense switch 2 is ON, the program execution pauses. Therefore, if data for several experiments are ready for reduction, the operator may run these in uninterrupted sequence by turning sense switch 2 OFF.

If the operator permits execution pause by leaving sense switch 2 ON, depression of the RUN button will return control to the beginning of the program, to start a new job.

The FORTRAN code is presented on pages 18-20. The program flow chart is presented on page 21.

SAMPLE INPUT

RAD IOLYSIS DATA REDUCTION PROGRAM TEST

02740.0000601015.00
 4.4 3.3 .210E-03

GAS 1
 GAS 2
 GAS 3
 GAS 4
 GAS 5
 GAS 6
 GAS 7
 GAS 8
 GAS 9
 GAS10

0956.00022.90 0.0
 000.02
 006.01
 000.02
 000.00
 000.00
 000.05
 000.01
 093.30
 000.57
 000.01

0956.00022.40 2.0
 000.02
 006.01
 000.03
 000.10
 000.00
 000.00
 000.00
 000.00
 093.31
 000.48
 000.04

0949.50022.80 4.0
 000.03
 006.02
 000.02
 000.12
 000.00
 000.00
 000.00
 000.00
 093.32
 000.45
 000.04

SAMPLE INPUT, CONCLUDED

0942.50022.70 6.0

000.02

6.05

000.02

0.14

000.00

000.00

000.00

93.28

000.42

000.05

0936.50022.80 8.0

000.03

006.04

000.02

000.12

000.00

000.00

000.00

093.33

000.40

000.06

0931.00023.00 10.5

000.02

006.07

000.03

000.11

000.00

000.07

000.02

093.23

000.38

000.06

SAMPLE OUTPUT

RADIOLYSIS DATA REDUCTION PROGRAM TEST

MICROMOLES OF GASES IN REACTION MIXTURE

TIME	0.0	2.0	4.0	6.0	8.0	10.5
DOSE	0.00E+00	3.87E+21	7.73E+21	1.16E+22	1.55E+22	2.03E+22
GAS 1	2.84E+01	2.86E+01	4.27E+01	2.85E+01	4.25E+01	2.85E+01
GAS 2	8.53E+03	8.59E+03	8.58E+03	8.61E+03	8.58E+03	8.62E+03
GAS 3	2.84E+01	4.29E+01	2.86E+01	2.85E+01	2.85E+01	4.24E+01
GAS 4	0.00E+00	1.43E+02	1.71E+02	1.99E+02	1.71E+02	1.56E+02
GAS 5	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
GAS 6	7.09E+01	0.00E+00	0.00E+00	0.00E+00	0.00E+00	9.72E+01
GAS 7	1.42E+01	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.78E+01
GAS 8	1.32E+05	1.33E+05	1.33E+05	1.33E+05	1.33E+05	1.32E+05
GAS 9	8.09E+02	6.86E+02	6.42E+02	5.98E+02	5.70E+02	5.41E+02
GAS 10	1.42E+01	5.72E+01	5.70E+01	7.10E+01	8.49E+01	8.48E+01

SAMPLE OUTPUT, CONTINUED

RADIOLYSIS DATA REDUCTION PROGRAM TEST

MOLECULES OF GASES IN REACTION MIXTURE

TIME	0.0	2.0	4.0	6.0	8.0	10.5
DOSE	0.00E+00	3.87E+21	7.73E+21	1.16E+22	1.55E+22	2.03E+22
GAS 1	1.71E+19	1.72E+19	2.57E+19	1.72E+19	2.56E+19	1.72E+19
GAS 2	5.14E+21	5.17E+21	5.17E+21	5.18E+21	5.17E+21	5.19E+21
GAS 3	1.71E+19	2.58E+19	1.72E+19	1.72E+19	1.72E+19	2.55E+19
GAS 4	0.00E+00	8.61E+19	1.03E+20	1.20E+20	1.03E+20	9.42E+19
GAS 5	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
GAS 6	4.27E+19	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.86E+19
GAS 7	8.55E+18	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.67E+19
GAS 8	7.97E+22	8.03E+22	8.01E+22	7.99E+22	7.99E+22	7.97E+22
GAS 9	4.87E+20	4.13E+20	3.86E+20	3.60E+20	3.43E+20	3.26E+20
GAS10	8.55E+18	3.44E+19	3.43E+19	4.28E+19	5.11E+19	5.11E+19

SAMPLE OUTPUT, CONCLUDED

RADIOLYSIS DATA REDUCTION PROGRAM TEST

CHANGE IN MOLECULE POPULATION FROM ZERO TIME

TIME	0.0	2.0	4.0	6.0	8.0	10.5
DOSE	0.00E+00	3.87E+21	7.73E+21	1.16E+22	1.55E+22	2.03E+22
GAS 1	0.00E+00	1.23E+17	8.61E+18	9.55E+16	8.49E+18	1.02E+17
GAS 2	0.00E+00	3.69E+19	3.14E+19	4.87E+19	3.38E+19	5.33E+19
GAS 3	0.00E+00	8.73E+18	1.23E+17	9.59E+16	7.46E+16	8.42E+18
GAS 4	0.00E+00	8.61E+19	1.03E+20	1.20E+20	1.03E+20	9.42E+19
GAS 5	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
GAS 6	0.00E+00	-4.27E+19	-4.27E+19	-4.27E+19	-4.27E+19	1.58E+19
GAS 7	0.00E+00	-8.55E+18	-8.55E+18	-8.55E+18	-8.55E+18	8.18E+18
GAS 8	0.00E+00	5.81E+20	3.73E+20	2.12E+20	1.55E+20	-1.55E+19
GAS 9	0.00E+00	-7.40E+19	-1.01E+20	-1.27E+20	-1.44E+20	-1.61E+20
GAS10	0.00E+00	2.59E+19	2.58E+19	3.42E+19	4.26E+19	4.25E+19

FORTRAN LISTING

```

C  RADF  17MY65
      DIMENSION EXPTID(10),FORMLA(15),DOSE(12)
      DIMENSION GASMOL(12,15),AVOGAS(12,15),DELMCL(12,15)
      DIMENSION SUMSAM(15),TIME(12),GASVP(15)
010   FORMAT(10A7)
011   FORMAT(F8.2,2I3,F5.2)
012   FORMAT(A5)
013   FORMAT(F7.2,F6.2,F7.1)
014   FORMAT(F6.2)
015   FORMAT(40H MICROMOLES OF GASES IN REACTION MIXTURE/)
016   FORMAT(5H TIME 6F10.1)
116   FORMAT(5H DOSE 1P6E10.2)
017   FORMAT(39H MOLECULES OF GASES IN REACTION MIXTURE/)
018   FORMAT(2H /)
019   FORMAT(A5,1P6E10.2)
020   FORMAT(15H END OF JOB      //)
120   FORMAT(F6.2,F5.3,E9.3)
121   FORMAT(1H )
122   FORMAT(45H CHANGE IN MOLECULE POPULATION FROM ZERO TIME/)
021   NSP=66
      READ 10,EXPTID
      PUNCH 10,EXPTID
      PUNCH 18
      NSP=NSP-3
      READ 11,V,L,K,SV
      READ 120,AZRO,EBAR,FLAM
      DO 22 I=1,K
      READ 12,FORMLA(I)
      SUMSAM(I)=0.0
022   CONTINUE
      SBYV=SV/V
      TEN17=(10.0**17)
      AVNR=6.023*TEN17
      AMPL=(1332.0*TEN17*AZRO*EBAR)/FLAM
      DO 45 J=1,L
      READ 13,P,T,TIME(J)
      ALL=(P*V)/(0.08205*76.0*(T+273.16))
      DOSE(J)=AMPL*(1-EXP(-FLAM*TIME(J)))
      DO 44 I=1,K
      READ 14,GASVP(I)
      GASMOL(J,I)=ALL*GASVP(I)
      IF(J-1)43,43,42
042   SUMSAM(I)=SUMSAM(I)+SBYV*GASMOL(J,I)
      GASMOL(J,I)=GASMOL(J,I)+SUMSAM(I)
043   AVOGAS(J,I)=AVNR*GASMOL(J,I)
      DELMCL(J,I)=AVOGAS(J,I)-AVOGAS(1,I)
044   CONTINUE
045   CONTINUE
      IF(L-6)50,50,55
050   MARK=1

```


FORTRAN LISTING, CONTINUED

```

      N=L
      M=1
      GO TO 60
055  MARK=3
      N=6
      M=1
060  PUNCH 15
      PUNCH 16,(TIME(J),J=M,N)
      PUNCH 18
      PUNCH 116,(DOSE(J),J=M,N)
      PUNCH 18
      NSP=NSP-8
      DO 62 I=1,K
      PUNCH 19,FORMLA(I),(GASMOL(J,I),J=M,N)
062  CONTINUE
      PUNCH 18
      NSP=NSP-K-2
      GO TO(67,68,65),MARK
065  MARK=2
      N=L
      M=7
      GO TO 60
067  N=L
      M=1
      GO TO 69
068  N=6
      M=1
      MARK=3
069  DO 169 NL=1,NSP
      PUNCH 121
169  CONTINUE
      PUNCH 10,EXPTID
      PUNCH 18
070  PUNCH 17
      PUNCH 16,(TIME(J),J=M,N)
      PUNCH 18
      PUNCH 116,(DOSE(J),J=M,N)
      PUNCH 18
      DO 72 I=1,K
      PUNCH 19,FORMLA(I),(AVOGAS(J,I),J=M,N)
072  CONTINUE
      PUNCH 18
      GO TO(77,78,75),MARK
075  MARK=2
      N=L
      M=7
      GO TO 70
077  N=L
      M=1
      GO TO 80
078  N=6
      M=1

```

FORTRAN LISTING, CONCLUDED

```
080    DO 180 NL=1,NSP
      PUNCH 121
180    CONTINUE
      PUNCH 10,EXPTID
      PUNCH 18
081    PUNCH 122
      PUNCH 16,(TIME(J),J=M,N)
      PUNCH 18
      PUNCH 116,(DOSE(J),J=M,N)
      PUNCH 18
      DO 82 I=1,K
      PUNCH 19,FORMLA(I),(DELMCL(J,I),J=M,N)
082    CONTINUE
      PUNCH 18
      GO TO(84,83),MARK
083    MARK=1
      N=L
      M=7
      GO TO 81
084    DO 184 NL=1,NSP
      PUNCH 121
184    CONTINUE
      PUNCH 20
      IF(SENSE SWITCH 2)85,21
085    PAUSE
      GO TO 21
      END
      EOF
```

PROGRAM FLOW CHART

APPENDIX A

RADD COVER

1. Title

2. Reaction volume[cc.], number of samples, number of gasses, sample volume[cc.]

3. A_0 [curies], $\bar{E}$ [MeV], λ [hr^{-1}]

4. Gas formulae, in order of data presentation

[illegible]

<u>Line</u>	<u>Format</u>
1.	10A7
2.	F8.2,2I3,F5.2
3.	F6.2,F5.3,E9.3
4.-18.	A5

RADD

1. Pressure[mm.Hg], Temperature[deg.C], Time[hrs.]

2. Volume % data in order of list on cover

1. Pressure[mm.Hg], Temperature[deg.C], Time[hrs.]

2. Volume % data in order of list on cover

<u>Line</u>	<u>Format</u>
1.	F7.2,F6.2,F7.1
2:-16.	F6.2

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