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# AUSTRALIAN ATOMIC ENERGY COMMISSION
## RESEARCH ESTABLISHMENT
## LUCAS HEIGHTS

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# A FORTRAN PROGRAM FOR THE ANALYSIS OF THE GAMMA SPECTRUM OF TECHNETIUM-99m ELUTED FROM FISSION PRODUCED MOLYBDENUM-99

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## ABSTRACT

The FORTRAN program IMPURE is used to process the gamma spectrum of $^{99m}Tc$ eluted from fission produced $^{99}Mo$ to determine the levels of $^{99}Mo$, $^{131}I$, $^{132}I$, $^{103}Ru$, and $^{140}La$ impurities. During counting with a Ge(Li) detector and 512 channel analyser, the sample is screened in a 4.0 mm lead pot to reduce the $^{99m}Tc$ contributions in comparison with the impurities.

The program sums the counts accumulated in appropriate spectrum peaks, and calculates the standard deviation. After correction for decay, lead attenuation, detector efficiency and the interference by overlapping peaks, the activity of each nuclide and its percentage contribution are calculated. The results are displayed in an easily read format.

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FISSION PRODUCTS; FORTRAN; GAMMA SPECTRA; I CODES; IBM COMPUTERS; IMPURITIES; IODINE 131; IODINE 132; ISOTOPE RATIO; LANTHANUM 140; MOLYBDENUM 99; RADIOACTIVITY; RUTHENIUM 103; TECHNETIUM 99

CONTENTS

## 1. INTRODUCTION

Gamma ray analysis of $^{99m}Tc$ eluted from fission produced $^{99}Mo$ generators shows that in addition to $^{99m}Tc$, small quantities of $^{99}Mo$, $^{131}I$, $^{132}I$, $^{103}Ru$ and $^{140}La$ may be present. A FORTRAN program 'IMPURE' for the IBM360/50 computer is used to calculate the activities of these nuclides from the spectral data. The use of this program in the analysis of A.A.E.C. produced $^{99m}Tc$ was outlined in another report (Boyd et al. 1971). Details of the program itself are given here.

## 2. THE GAMMA SPECTRUM

The gamma spectrum analysed is obtained from a 10 ml eluted $^{99m}Tc$ sample shielded in a 4.0 mm thick lead pot to reduce the 140 keV contribution to manageable levels in comparison with the impurities (Figure 1). The sample is counted in a fixed geometry using a lithium drifted germanium detector with a known energy response and a 512 channel pulse height analyser. Counting times for millicurie levels of $^{99m}Tc$ are of the order of 100 minutes. The analyser gain corresponds to approximately 5 keV per channel. At the end of counting the accumulated data are read out in digital form and transcribed to punched cards to serve as input data for the program. Only the first 400 channels are used since no relevant peaks occur beyond that point. The counting time and the decay time before analysis are also read in as data. The program is used to calculate the relevant activities from the counts accumulated in selected spectrum peaks.

## 3. THE PROGRAM 'IMPURE'

For a detailed flow diagram and a listing of the program see Appendix A and Appendix B.

Initially three 400 element arrays of physical data used in the calculations are established. These arrays are EN, P and EF. EN is the photon energy from 0.005 MeV to 2.000 MeV in steps of 0.005 MeV corresponding to each analyser channel. P in its final form is a table of linear attenuation coefficients for each energy multiplied by the lead screen thickness. This array is derived from a table of mass attenuation coefficients read in as data. To avoid overflow in the calculations the first 16 values of P, which are not used in the calculations, are arbitrarily set to zero. EF is the detector efficiency for each energy step, calculated from an empirical formula fitted to experimental data (Boyd et al. 1971). The calculation of EF values (Statement 3,

Appendix B) includes a correction to the absolute efficiencies to allow for the sample geometry.

A Do loop to Statement 56 contains the remainder of the program to permit any number of spectra (M) to be analysed in a run on the computer. The operations on the spectral data commence at Statement 309. The counts accumulated in the 400 analyser channels used are read in as an array KH and immediately converted from integer to real numbers (array CH). The counting time, CT, the decay time before counting DT and a sample reference KODE are read in according to FORMAT Statement 8.

The activities are calculated using photopeaks nominated in the program. Since it was difficult to set the gain to exactly 5 keV per channel, the positions nominated were determined from a number of calibration runs and not calculated from the instrument gain. Exact steps of 5 keV are used in the above data arrays and the values used in the calculations are selected on this basis.

Where possible a photopeak unique to the nuclide involved is used. Where two peaks overlap, the order of calculation is chosen so that appropriate corrections can be made. COMMENT statements head the program steps for each nuclide. The activity of $^{99}$Mo is calculated first using the 0.74 MeV peak. The exact position of the peak (channel K) is determined using a simple searching routine around the approximate location between channel 143 and channel 150. The counts in this channel and in three channels on each side are summed by using a Do loop (to Statement 6). The background continuum of the spectrum (BG) is taken as the mean of the counts in the extreme channels of the peak multiplied by the number of channels. This is subtracted from the total counts in the selected channels to give the molybdenum peak height.

The standard deviation (DEVM) of the peak height is calculated at Statement 22 as an indication of the statistical significance of the peak. If no $^{99}$Mo is present or if the peak is very small it is possible for the calculated value to be negative after subtraction of background. When this occurs the program arbitrarily sets both the peak height and the standard deviation to zero.

Two correction terms COR and CORT corresponding to the $^{99}$Mo contribution at the 0.140 MeV peak of $^{99m}$Tc and the 0.36 MeV peak of $^{131}$I are calculated at this stage. The peak total is corrected for lead screen attenuation, detector efficiency and photon yield (10%) per disintegration at Statement 23.

Using the formula

$$Co = \frac{\lambda N}{(1-e^{-\lambda CT})}$$

where Co = the count rate at commencement of counting

CT = counting time

N = total counts in time CT

$\lambda$ = decay constant of nuclide

a correction is made for decay during counting. This is followed by a correction back to the time of separation. The final step calculates the $^{99}$Mo activity (ACTM) in microcuries at the time of preparation.

The program then calculates the activities of the other nuclides in the following order. $^{99m}$Tc (ACTT), $^{140}$La (ALA), $^{132}$I (ACTIB), $^{131}$I(ACTIA), and $^{103}$Ru (ACTRU). The $^{103}$Ru calculation includes a correction CORTL for the contribution of $^{140}$La to the 0.498 MeV peak.

The $^{99m}$Tc and impurity activities are summed by the statement

TOT = ACTM + ACTT + ALA + ACTIB + ACTIA + ACTRU

and the contributions of each as a percentage of the total calculated. A series of WRITE and FORMAT statements following the statement

WRITE (3,99) KODE

is used to display the results of the calculations for each nuclide. The activity, the percentage of the total activity and the standard deviation of the selected peak are written out. The number of figures displayed is not intended to imply the degree of accuracy. It has been shown (Boyd et al. 1971) that the accuracy of results obtained is generally of the order of ± 20 per cent at the 90 per cent confidence level.

A typical set of results is given in Appendix C. The number following the heading ANALYSIS OF TECHNETIUM 99M is the identification KODE. As a check on the stability of the analyser system the position of the $^{99}$Mo peak is also noted.

## 4. ROUTINE USE OF PROGRAM

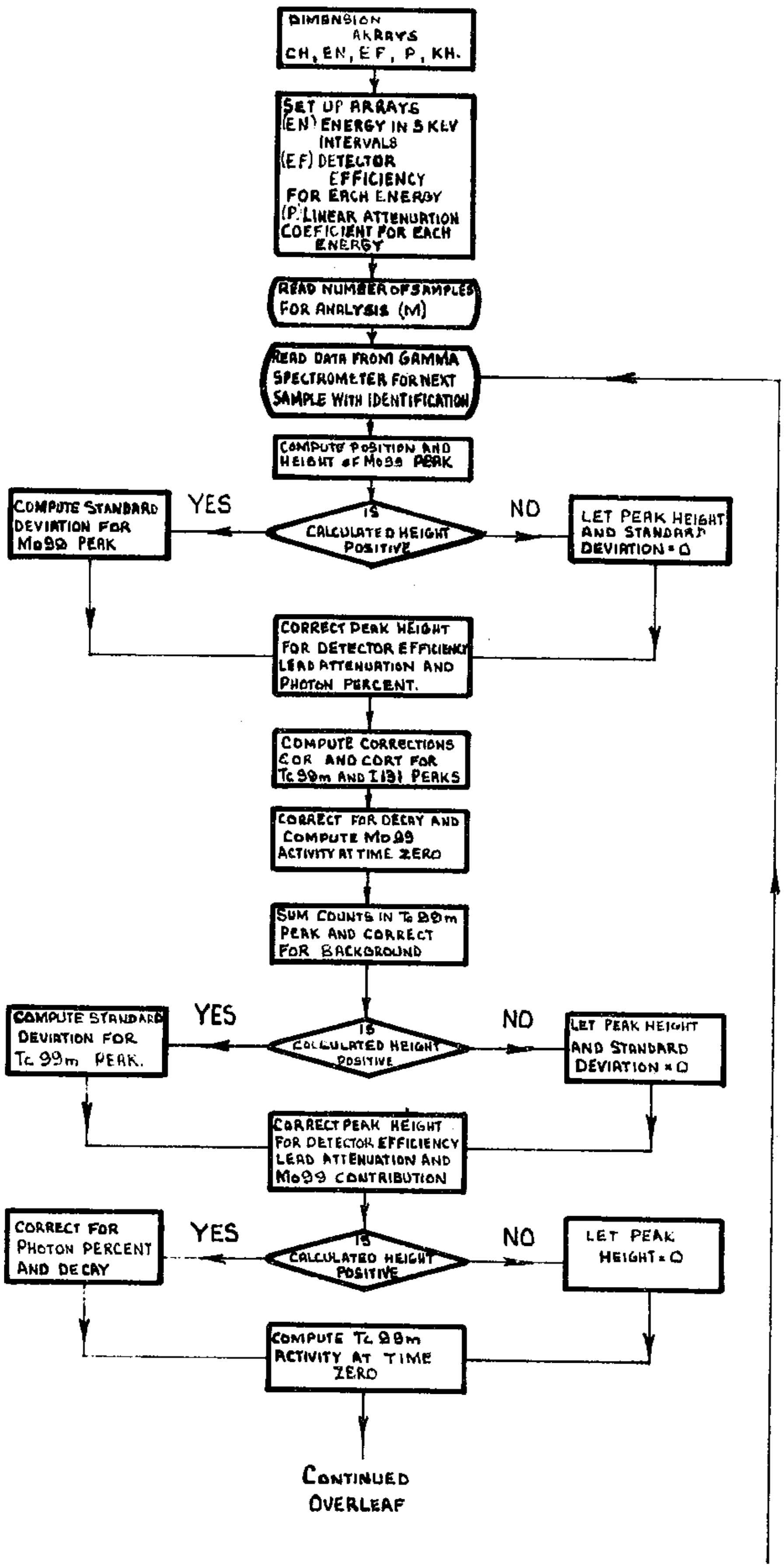
The program may be run as a Class A job under the MVT system of operation used with the AAEC IBM360/50 computer. For convenience the program has been stored on disc but may also be run with the FORTRAN source deck. The appropriate control cards for disc and card operation under this system and some additional information on data formats etc. are given in Appendix D.

## 5. REFERENCE

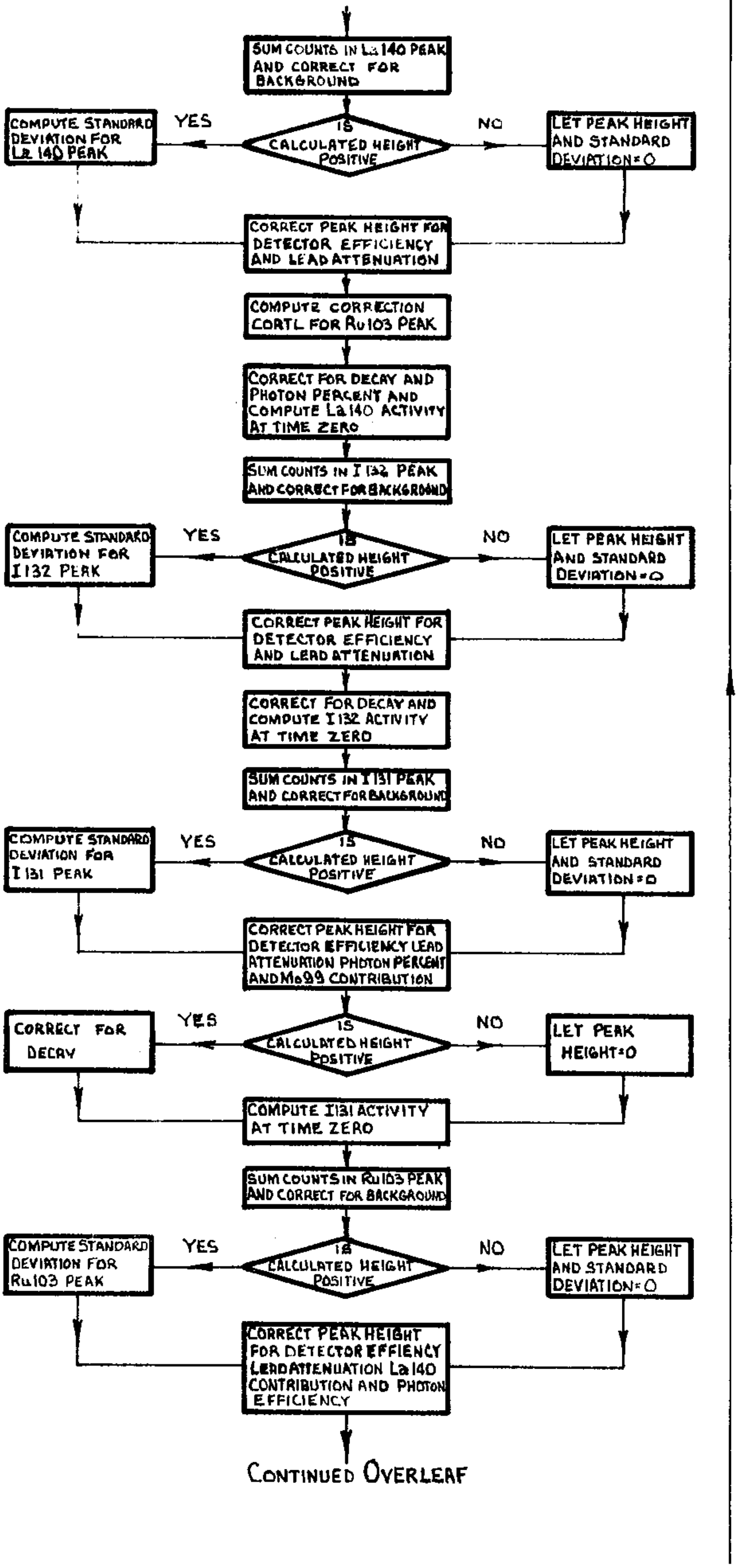
Boyd, R.E., Hetherington, E.L.R. and Wood, N.R. (1971). - 'Technetium-99m generators prepared from fission produced molybdenum-99. Quality control and performance aspects.' AAEC/E224.

APPENDIX A

## FLOW SHEET FOR IMPURE

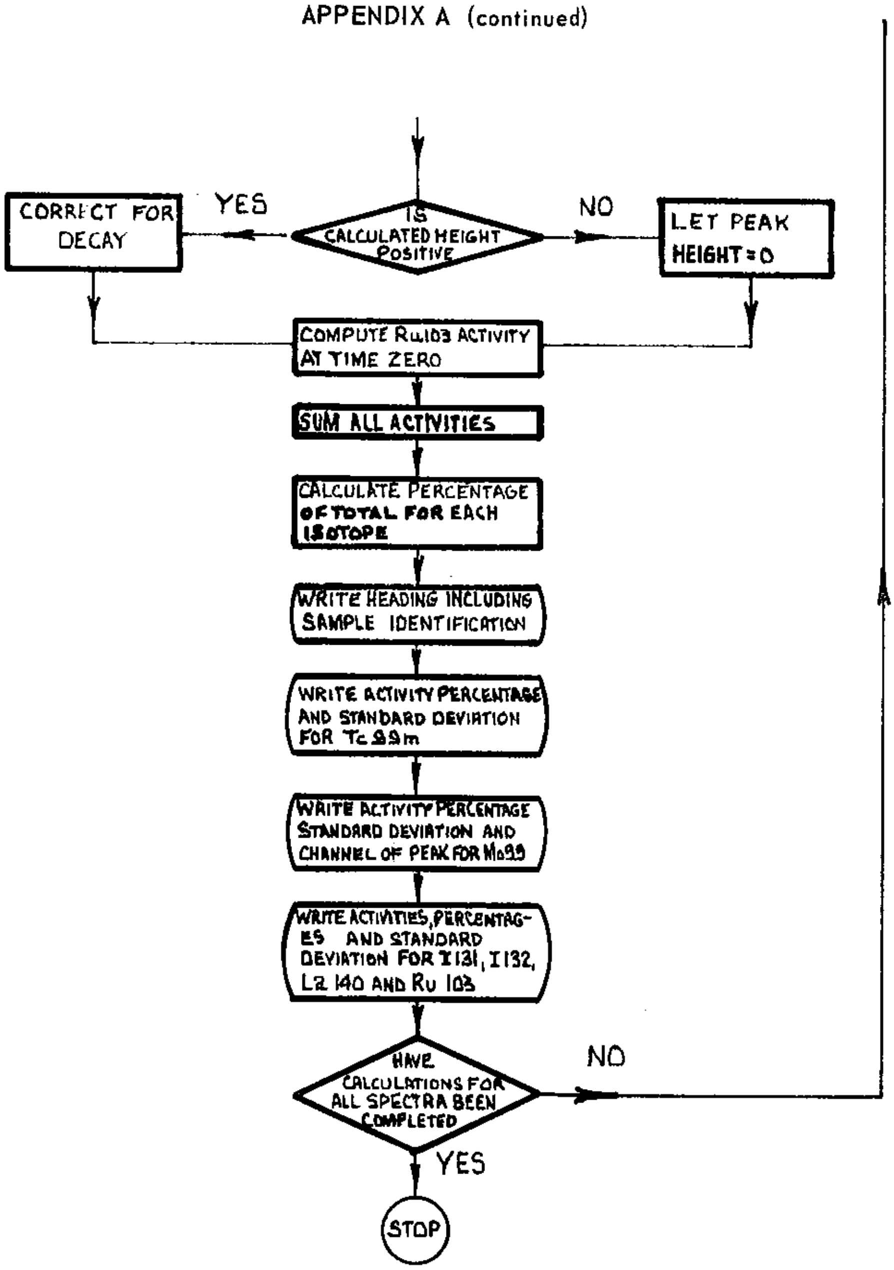



CONTINUED OVERLEAF




CONTINUED OVERLEAF

APPENDIX A (continued)



# APPENDIX B

Listing of program IMPURE

```
      DIMENSION CH(4ØØ),EN(4ØØ),EF(4ØØ),P(4ØØ),KH(4ØØ)
      X=Ø.Ø
      DO 2 I=1,4ØØ
      X=X+1.Ø
      EN(I)=Ø.Ø
      EN(I)=EN(I)+X
    2 EN(I)=EN(I)/2ØØ.Ø
      CONTINUE
      DO 3 I=1,4ØØ
    3 EF(I)=Ø.ØØØØ49/(EN(I)**1.95)
      CONTINUE
      READ(1,4Ø)P
   4Ø FORMAT(8F8.3)
      DO 5 I=1,4ØØ
    5 P(I)=P(I)*11.3437*Ø.4ØØØ
      READ(1,2ØØ)M
  2ØØ FORMAT(I5)
      DO 56 JJ=1,M
  3Ø9 READ(1,1)KH
    1 FORMAT(8I7)
      DO 55 I=1,4ØØ
   55 CH(I)=KH(I)
  2Ø8 CONTINUE
      READ(1,8)CT,DT,KODE
    8 FORMAT(F6.3,F6.3,I6)
C     MOLYBDENUM CALCULATION
      T=Ø.Ø
      DO 2Ø1 I=143,15Ø
      Q=CH(I)-T
      IF(Q)122,123,123
  122 CONTINUE
      GO TO 2Ø1
  123 T=CH(I)
      K=I
  2Ø1 CONTINUE
      CM=Ø.Ø
      J=K-3
      L=K+3
      DO 6 I=J,L
    6 CM=CM+CH(I)
    7 BG=3.5*(CH(K-3)+CH(K+3))
      CM=CM-BG
      IF(CM-Ø.Ø)21,24,22
   21 CM=Ø.Ø
   24 DEVM=Ø.Ø
      GO TO 23
   22 DEVM=1ØØ.Ø*(CM**Ø.5)/CM
   23 CM=CM*EXP(P(148))/EF(148)
      CM=CM/Ø.1Ø
      CMCO=CM
      COR=CMCO*Ø.Ø46
      CORT=CMCO*Ø.Ø1
      CM=Ø.693*CM/(1.Ø-1.Ø/EXP(Ø.693*CT/67.Ø))/67.Ø
      CM=CM*EXP(Ø.693*DT/67.Ø)
      ACTM=CM/(3.7E+4*36ØØ.Ø)
C     TECHNETIUM CALCULATION
      CTM=Ø.Ø
      DO 1Ø I=25,31
   1Ø CTM=CTM+CH(I)
      CONTINUE
```

```
      BG=(CH(25)+CH(31))*3.5
      CTM=CTM-BG
      IF(CTM-Ø.Ø)25,26,27
   25 CTM=Ø.3
   26 DEVTM=Ø.Ø
      GO TO 28
   27 DEVTM=1ØØ.Ø*(CTM**Ø.5)/CTM
   28 CTM=CTM*EXP(P(28))/EF(28)
C     MOLYBDENUM CORRECTION
      CTM=CTM-COR
      IF(CTM-Ø.Ø)29,3Ø,3Ø
   29 CTM=Ø.Ø
   3Ø CTM=Ø.693*CTM/(1.Ø-1.Ø/EXP(Ø.693*CT/6.Ø))/6.Ø
      CTM=CTM*EXP(Ø.693*DT/6.Ø)
      CTM=CTM/Ø.9Ø1
      ACTT=CTM/(3.7E+4*36ØØ.Ø)
C     LANTHANUM CALCULATION
      CLA=Ø.Ø
      DO 13 I=311,317
   13 CLA=CLA+CH(I)
      CONTINUE
      BG=(CH(311)+CH(317))*3.5
      CLA=CLA-BG
      IF(CLA-Ø.Ø)31,32,33
   31 CLA=Ø.Ø
   32 DEVLA=Ø.Ø
      GO TO 38
   33 DEVLA=1ØØ.Ø*(CLA**Ø.5)/CLA
   38 CLA=CLA*EXP(P(32Ø))/EF(32Ø)
      CORTL=CLA*Ø.41
      CLA=Ø.693*CLA/(1.Ø-1.Ø/EXP(Ø.693*CT/4Ø.2))/4Ø.2
      CLA=CLA*EXP(Ø.693*DT/4Ø.2)
      CLA=CLA/Ø.95
      ALA=CLA/(36ØØ.Ø*3.7E+4)
C     IODINE 132 CALCULATION
      CIB=Ø.Ø
      DO 18 I=129,135
   18 CIB=CIB+CH(I)
      BG=CH(129)+CH(135)
      BG=3.5*BG
      CIB=CIB-BG
      IF(CIB-Ø.Ø)39,41,42
   39 CIB=Ø.Ø
   41 DEVIB=Ø.Ø
      GO TO 43
   42 DEVIB=1ØØ.Ø*(CIB**Ø.5)/CIB
   43 CIB=CIB*EXP(P(134))/EF(134)
      CIB=Ø.693*CIB/(1.Ø-1.Ø/EXP(Ø.693*CT/2.3))/2.3
      CIB=CIB*EXP(Ø.693*DT/2.3)
      ACTIB=CIB/(36ØØ.Ø*3.7E+4)
C     IODINE 131 CALCULATION
      CIA=Ø.Ø
      DO 16 I=7Ø,75
   16 CIA=CIA+CH(I)
      BG=CH(7Ø)+CH(75)
      BG=3.Ø*BG
      CIA=CIA-BG
      IF(CIA-Ø.Ø)44,45,46
   44 CIA=Ø.Ø
   45 DEVIA=Ø.Ø
```

```
      GO TO 47
   46 DEVIA=100.0*(CIA**0.5)/CIA
   47 CIA=CIA*EXP(P(72))/EF(72)
      CIA=CIA-CORT
      CIA=CIA/0.79
      IF(CIA-0.0)60,61,61
   60 CIA=0.0
   61 CIA=0.693*CIA/(1.0-1.0/EXP(0.693*CT/192.96))/192.96
      CIA=CIA*EXP(0.693*DT/192.96)
      ACTIA=CIA/(3600.0*3.7E+4)
C     RUTHENIUM 103 CALCULATION
      CR=0.0
      DO 12 I=97,101
   12 CR=CR+CH(I)
      BG=(CH(97)+CH(101))*2.5
      CR=CR-BG
      IF(CR-0.0)34,50,51
   34 CR=0.0
   50 DEVRU=0.0
      GO TO 35
   51 DEVRU=100.0*(CR**0.5)/CR
   35 CR=CR*EXP(P(100))/EF(100)
      CR=CR-CORTL
      CR=CR/0.88
      IF(CR-0.0)36,37,37
   36 CR=0.0
   37 CR=0.693*CR/(1.0-1.0/EXP(0.693*CT/960.0))/960.0
      CR=CR*EXP(0.693*DT/960.0)
      ACTRU=CR/(3600.0*3.7E+4)
      TOT=ACTM+ACTT+ALA+ACTIB+ACTIA+ACTRU
      PT=ACTT*100.0/TOT
      PM=ACTM*100.0/TOT
      PLA=ALA*100.0/TOT
      PIA=ACTIA*100.0/TOT
      PIB=ACTIB*100.0/TOT
      PRU=ACTRU*100.0/TOT
      WRITE(3,99)KODE
   99 FORMAT(1H1,26HANALYSIS OF TECHNETIUM 99M,I6)
      WRITE(3,11)ACTT,PT
   11 FORMAT(1H ,14HTECHNETIUM 99M,F9.2,11HMICROCURIES,F9.5,1H%)
      WRITE(3,4)DEVTM
    4 FORMAT(1H ,18HSTANDARD DEVIATION,F9.5,1H%)
      WRITE(3,9)ACTM,PM
    9 FORMAT(1H ,13HMOLYBDENUM 99,F9.4,11HMICROCURIES,F9.5,1H%)
      WRITE(3,100)K
  100 FORMAT(1H ,36HMOLYBDENUM 99 PEAK OCCURS IN CHANNEL,I4)
      WRITE(3,4)DEVM
      WRITE(3,17)ACTIA,PIA
   17 FORMAT(1H ,10HIODINE 131,F9.4,11HMICROCURIES,F9.5,1H%)
      WRITE(3,4)DEVIA
      WRITE(3,19)ACTIB,PIB
   19 FORMAT(1H ,10HIODINE 132,F9.4,11HMICROCURIES,F9.5,1H%)
      WRITE(3,4)DEVIB
      WRITE(3,14)ALA,PLA
   14 FORMAT(1H ,13HLANTHANUM 140,F9.4,11HMICROCURIES,F9.5,1H%)
      WRITE(3,4)DEVLA
      WRITE(3,20)ACTRU,PRU
   20 FORMAT(1H ,13HRUTHENIUM 103,F9.4,11HMICROCURIES,F9.5,1H%)
      WRITE(3,4)DEVRU
   56 CONTINUE
      CONTINUE
      STOP
      END
```

# APPENDIX C

<u>Samples of results as read out by the program IMPURE</u>

```
ANALYSIS OF TECHNETIUM 99M157371
TECHNETIUM 99M 37414.47MICROCURIES 99.96417%
STANDARD DEVIATION  0.31832%
MOLYBDENUM 99  13.3262MICROCURIES  0.03561%
MOLYBDENUM 99 PEAK OCCURS IN CHANNEL 146
STANDARD DEVIATION  0.78584%
IODINE 131    0.0    MICROCURIES  0.0    %
STANDARD DEVIATION  1.90174%
IODINE 132    0.0    MICROCURIES  0.0    %
STANDARD DEVIATION  0.0    %
LANTHANUM 140   0.0047MICROCURIES  0.00001%
STANDARD DEVIATION 26.72610%
RUTHENIUM 103   0.0767MICROCURIES  0.00020%
STANDARD DEVIATION  2.61176%
```

```
ANALYSIS OF TECHNETIUM 99M470473
TECHNETIUM 99M 15339.97MICROCURIES 99.94594%
STANDARD DEVIATION  0.37065%
MOLYBDENUM 99   7.8748MICROCURIES  0.05131%
MOLYBDENUM 99 PEAK OCCURS IN CHANNEL 146
STANDARD DEVIATION  0.99575%
IODINE 131   0.0120MICROCURIES  0.00008%
STANDARD DEVIATION  2.21079%
IODINE 132    0.0    MICROCURIES  0.0    %
STANDARD DEVIATION  0.0    %
LANTHANUM 140   0.0025MICROCURIES  0.00002%
STANDARD DEVIATION 35.35532%
RUTHENIUM 103   0.4119MICROCURIES  0.00268%
STANDARD DEVIATION  1.13728%
```

# APPENDIX D

<u>CONTROL CARDS REQUIRED TO RUN IMPURE</u>

<u>(1) WITH PROGRAM STORED ON DISC</u>

```
   //
#  //ELHIMP    JOB I1CØ5673,E.L.R.HETHERINGTON
   //  EXEC  FORTHG,REF=PACK6,DSN='ISOTOPE.PROGLIB',PRG=IMPURE
   //GO.SYSIN    DD     *


     5Ø DATA CARDS GIVING MASS ATTENUATION COEFFICIENTS IN FORMAT 8F8.3 THE
     FIRST 16 VALUES TO BE SET TO ZERO


     DATA CARD GIVING THE NUMBER OF SPECTRA ANALYSED (FORMAT I5)


                                 FOR EACH SPECTRUM ANALYSED
     5Ø DATA CARDS GIVING COUNTS ACCUMULATED IN 4ØØ ANALYSER CHANNELS IN FORMAT
     8I7 (6 DIGIT NUMBERS WITH THE FIRST POSITION BLANK)
                                        FOLLOWED BY
     DATA CARD GIVING COUNT TIME (FORMAT F6.3) DECAY TIME (FORMAT F6.3) AND
     IDENTIFICATION KODE (FORMAT I6)


   /*
   //
```

<u>(2) WITH FORTRAN SOURCE DECK</u>

```
   //
#  //ELHIMP    JOB I1CØ5673,E.L.R.HETHERINGTON
   // EXEC FORTGCLG
   //FORT.SYSIN        DD        *


     FORTRAN SOURCE PROGRAM CARDS


   /*
   //GO.SYSIN    DD     *


     5Ø DATA CARDS GIVING MASS ATTENUATION COEFFICIENTS IN FORMAT 8F8.3 THE
     FIRST 16 VALUES TO BE SET TO ZERO


     DATA CARD GIVING THE NUMBER OF SPECTRA ANALYSED (FORMAT I5)


                                 FOR EACH SPECTRUM ANALYSED
     5Ø DATA CARDS GIVING COUNTS ACCUMULATED IN 4ØØ ANALYSER CHANNELS IN FORMAT
     8I7 (6 DIGIT NUMBERS WITH THE FIRST POSITION BLANK)
                                        FOLLOWED BY
     DATA CARD GIVING COUNT TIME (FORMAT F6.3) DECAY TIME (FORMAT F6.3) AND
     IDENTIFICATION KODE (FORMAT I6)


   /*
   //
```

# User's own JOB card required.

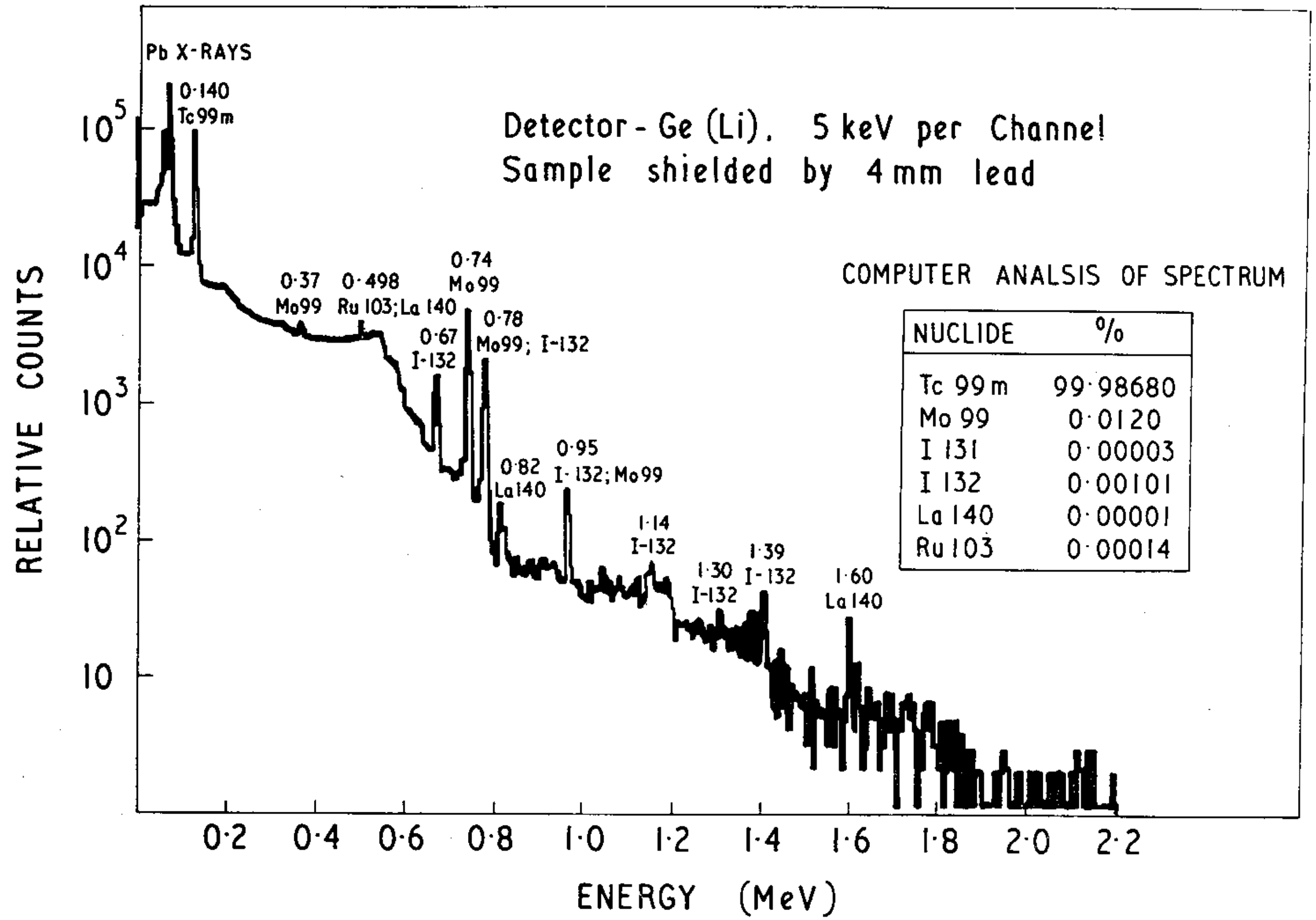



**FIGURE 1. GAMMA SPECTRUM OF SAMPLE OF 2nd 19 ml OF ELUATE FROM A FISSION PRODUCED Mo99 GENERATOR**