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SPECACT, A FORTRAN PROGRAM FOR THE ROUTINE CALCULATION OF THE SPECIFIC ACTIVITY OF FISSION-PRODUCED MOLYBDENUM-99

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ABSTRACT

The specific activity of ^{99}Mo is an important factor in the production of ^{99}Mo/^{99m}Tc generators, used in nuclear medicine. Molybdenum-99 formed *via* the fission route is not carrier free but is contaminated by a number of stable molybdenum isotopes formed concurrently during neutron irradiation of the uranium target. The specific activity of fission-based ^{99}Mo is therefore a function of irradiation time and post-irradiation decay. A computer program, written in FORTRAN, is presented for defining routinely the specific activity of fission-based ^{99}Mo.

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FORTRAN; S CODES; MOLYBDENUM 99; RADIOACTIVITY; NUCLEAR DECAY; IRRADIATION; MASS; NEUTRON FLUX; URANIUM 235 TARGET; FISSION YIELD

CONTENTS

1. INTRODUCTION

Since 1970, ^{99m}Tc generators based upon fission-produced ^{99}Mo have gained a greater acceptability in nuclear medicine than those from (n,γ) derived ^{99}Mo. This increase in popularity may be attributed partly to the very high specific activities of ^{99}Mo attainable using the fission pathway.

Technetium-99m generators prepared from fission-based ^{99}Mo having very high specific activities have demonstrated significantly improved elution characteristics when compared with generators prepared from low specific activity ^{99}Mo, produced from (n,γ) reactions [Boyd 1973]. Additionally, in the production of high activity ^{99m}Tc generators it is important to ensure that the maximum sorption capacity of the alumina bed employed in the generator is not approached, as this may lead to high ^{99}Mo breakthrough during the elution cycle of the generator and therefore compromise the radionuclidic purity of acquired ^{99m}Tc. Hence a knowledge of the specific activity of ^{99}Mo is an important factor in the preparation of ^{99}Mo/^{99m}Tc generators for use in nuclear medicine.

The computer program SPECACT was developed, as a part of the quality control regime, for assessing routinely the specific activity of fission-produced ^{99}Mo, prepared at Lucas Heights. This program computes the specific activity of ^{99}Mo based upon neutron-irradiation and post-irradiation decay parameters of the uranium target

2. SPECIFIC ACTIVITY OF FISSION PRODUCED MOLYBDENUM-99

Molybdenum-99 is formed directly from ^{235}U and has a cumulative fission yield of 6.1 per cent [Rose and Burrows 1976] in a flux of thermal neutrons; however, when formed by this pathway it is not 'carrier free' but contains a number of stable molybdenum isotopes which are formed concurrently, during the irradiation process, by the following reactions:

(a)
$$\begin{tikzcd} {}^{235}\text{U(n,f)}\ \ldots\ldots & \underset{(64.02\text{ d})}{{}^{95}\text{Zr}} \arrow[r, "1.2\%"] \arrow[dr, "98.8\%"'] & \underset{(86.6\text{ h})}{{}^{95}\text{Nb}} \arrow[d] & \\ & & \underset{(34.97\text{ d})}{{}^{95}\text{Nb}} & {}^{95}\text{Mo; fission yield 6.5\% (stable)} \end{tikzcd}$$

(b)
$$\begin{tikzcd} {}^{235}\text{U(n,f)}\ \ldots\ldots & \underset{(17.0\text{ h})}{{}^{97}\text{Zr}} \arrow[r, "86.2\%"] \arrow[dr, "13.8\%"'] & \underset{(60\text{ s})}{{}^{97m}\text{Nb}} \arrow[d] \\ & & \underset{(72.1\text{ min})}{{}^{97}\text{Nb}} \arrow[r] & {}^{97}\text{Mo; fission yield 5.9\% (stable)} \end{tikzcd}$$

(c)
$$\begin{tikzcd} {}^{235}\text{U(n,f)}\ \ldots\ldots\ \underset{(51.3\text{ min})}{{}^{98m}\text{Nb}} \arrow[r] & {}^{98}\text{Mo; fission yield 5.7\% (stable)} \end{tikzcd}$$

(d) ^{235}U(n,f) .... ^{100}Mo; fission yield 6.3% (stable)

In addition to being formed directly in the fission process, ^{99}Mo is also produced by neutron activation of the stable ^{98}Mo formed in reaction (c); however, it was computed that the activity of ^{99}Mo derived from this source is less than one per cent of the total ^{99}Mo activity. Short-lived fission-produced molybdenum, such as ^{102}Mo-^{107}Mo with half-lives of less than 12 minutes, makes insignificant contributions to the specific activity of ^{99}Mo and was therefore not considered in the program. Reactions (a) to (d) effectively reduce the specific activity from its carrier-free value of 4.79×10^5 Ci g^{-1} ^{99}Mo by factors of up to 20, depending on the level of irradiation and the decay times.

3. METHOD OF CALCULATION

3.1 Activation Equations

During the neutron irradiation of a uranium target, the build-up of fission fragments in a fission chain of the type $^{235}U(n,f) \cdots N_1 \longrightarrow N_2 \longrightarrow N_3$ in which N_1, N_2 and N_3 are radioactive fission fragments, is given by the general activation equations (refer to section 7 for definition of notation):

$$N_1 = \frac{K_1}{\lambda_1}(1 - e^{-\lambda_1 t})$$

$$N_2 = \frac{K_1 + K_2}{\lambda_2}(1 - e^{-\lambda_2 t}) - \frac{K_1}{(\lambda_2 - \lambda_1)}(e^{-\lambda_1 t} - e^{-\lambda_2 t})$$

$$N_3 = \frac{K_1 + K_2 + K_3}{\lambda_3}(1 - e^{-\lambda_3 t}) \qquad - \frac{(K_1 + K_2)}{(\lambda_3 - \lambda_2)}(e^{-\lambda_2 t} - e^{-\lambda_3 t})$$

$$- K_1\lambda_2 \left\{ \frac{e^{-\lambda_1 t}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)} \right.$$

$$\left. + \frac{e^{-\lambda_3 t}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)} \right\}$$

The production of the niobium intermediates in reactions (a) and (b), arise almost solely from direct decay of the respective zirconium precursors. The direct fission yield of these intermediates is so low that the values of K_2 and K_3, in the activation equation (B) and (C), can be considered to be zero.

The application of equations (A), (B) and (C) assumes that N_μ remains essentially constant throughout the irradiation period. For short irradiation periods of less than ten days, as is used at Lucas Heights, and at neutron fluxes of between 8×10^{13} to 1.0×10^{14} n cm^{-2} s^{-1}, there is little target burn-up and the value of N_μ remains essentially constant throughout the period of irradiation.

3.2 Post-irradiation Decay Equations

During the post-irradiation phase, stable molybdenum nuclides are still being generated from decay of the parent and daughter fission fragments, as indicated in reactions (a), (b) and (c). The decay of these fragments has the effect of reducing the specific activity of ^{99}Mo.

The amount of the radioactive species, N_1, N_2 and N_3 remaining after time T in a multiple step decay chain is given by $N_1 = N_1^0 \, e^{-\lambda_1 T}$

$$N_2 = \frac{N_1^0 \, \lambda_1}{\lambda_2 - \lambda_1}(e^{-\lambda_1 T} - e^{-\lambda_2 T}) + N_2^0 \, e^{-\lambda_2 T}$$

$$N_3 = N_3^0 \, e^{-\lambda_3 T} + N_2^0 \, \frac{\lambda_2}{\lambda_3 - \lambda_2}(e^{-\lambda_2 T} - e^{-\lambda_3 T})$$

$$+ N_1^0 \left\{ \frac{\lambda_1}{(\lambda_2-\lambda_1)} \frac{\lambda_2}{(\lambda_3-\lambda_1)} e^{-\lambda_1 T} + \frac{\lambda_1}{(\lambda_1-\lambda_2)} \frac{\lambda_2}{(\lambda_3-\lambda_2)} e^{-\lambda_2 T} \right.$$

$$\left. + \frac{\lambda_1}{(\lambda_1-\lambda_3)} \frac{\lambda_2}{(\lambda_2-\lambda_3)} e^{-\lambda_3 T} \right\}$$

In this case T is the time lapse between reactor discharge and generator loading.

4. PROGRAM

The program computes the activity of ^{99}Mo and thc total mass of each of the molybdenum isotopes formed during neutron irradiation, taking into account the branching ratios in reactions (a) and (b). Similarly, during the post-irradiation phase, the specific activity of ^{99}Mo is affected by the decay of ^{99}Mo and the ingrowth of stable molybdenum species from the parent-daughter fission fragments.

The input parameters for the program include the neutron flux and irradiation time followed by the post-irradiation decay time (Appendix A). The computer output gives the individual masses for each of the molybdenum isotopes produced, in addition to the total activity and specific activity for ^{99}Mo (see Appendix B for typical output).

5. CONCLUSIONS

The program SPECACT was written to monitor routinely the specific activity of fission-based ^{99}Mo produced at Lucas Heights as part of its quality assurance regime for the preparation of ^{99}Mo/^{99m}Tc generators. The program input simply requires the values of neutron flux, irradiation time and post-irradiation decay time to be entered to complete the specific activity calculations.

6. REFERENCES

Boyd, R. [1973] - In *Radiopharmaceuticals and Labelled Compounds*, Vol.1, p.3. IAEA, Vienna.

Rose, P.F., Burrows, T.W. [1976] - Fission Product Decay Data, Vol.1, BNL-NCS-50545.

7. NOTATION

K_1 = $Y_1 \sigma_f \phi N_\mu$.

K_2 = $Y_2 \sigma_f \phi N_\mu$.

K_3 = $Y_3 \sigma_f \phi N_\mu$.

t = time of irradiation.

T = post-irradiation decay time.

λ_1 = decay constant of the parent fission fragment with N_1 atoms at time t.

λ_2 = decay constant of the fission product with N_2 atoms at time t.

λ_3 = decay constant of the fission product with N_3 atoms at time t.

σ_f = fission cross section of uranium (580 barns for ^{235}U).

N_μ = number of atoms of uranium in target.

N_1^0 = amount of species N_1 at $T = 0$.

N_2^0 = amount of species N_2 at $T = 0$.

N_3^0 = amount of species N_3 at $T = 0$.

ϕ = neutron flux.

$\left.\begin{matrix}Y_1\\Y_2\\Y_3\end{matrix}\right\}$ = the fission yields of N_1, N_2 and N_3 respectively.

APPENDIX A
LISTING OF THE PROGRAM SPECACT

```
      IMPLICIT REAL (K-N)
      DOUBLE PRECISION DT,TC,DI,TI,L1,L2,L3,L4,L5,L6,L7,L8
      NU=6.0248E23/235.
      SIG=560.*1.0E-24
      FLUX=1.0E14
      L1=.693147/(64.02*8.64D4)
      L2=.693147/(86.6*3.6D3)
      L3=.693147/(34.97*8.64D4)
      L4=.693147/(17.0*3.6D3)
      L5=.693147/60.0
      L6=.693147/(72.1*60.)
      L7=.693147/(51.3*60.)
      L8=.693147/(66.02*3.6D3)
      Y1=0.0646
      Y2=0.0595
      Y3=0.0573
      Y4=0.0610
      Y5=0.0630
      K1=Y1*NU*SIG*FLUX
      K2=Y2*NU*SIG*FLUX
      K3=Y3*NU*SIG*FLUX
      K4=Y4*NU*SIG*FLUX
      DI=0.0
      DO 99 J=1,21
      DI=DI+1.0
      IF(MOD(J,2).EQ.0) WRITE(3,3)
      IF(MOD(J,2).NE.0) WRITE(3,6)
    6 FORMAT('1  ')
    3 FORMAT(////)
      WRITE(3,1)DI
    1 FORMAT(' IRRADIATION TIME = ',F4.1,'  DAYS')
      TI=DI*8.64D4
      TC=-1.0
      WRITE(3,2)
      WRITE(3,8)
      WRITE(3,9)
    2 FORMAT('  DECAY   ACTIVITY     MASS        MASS        MASS
     1 MASS        MASS        MASS      SPECIFIC')
    8 FORMAT('  TIME      MO99       MO95        MO97        MO98
     1 MO99        MO100    MOLYBDENUM   ACTIVITY')
    9 FORMAT('  DAYS      CURIE      GRAM        GRAM        GRAM
     1 GRAM        GRAM        GRAM     CURIE/GRAM')
      DO 88 JJ=1,21
      TC=TC+1.
      DT=TC*8.64D4
      ATM5=K1*TI
      N1=K1/L1*(1.0-DEXP(-L1*TI))
      N2=K1/L2*(1.-DEXP(-L2*TI))-K1/(L2-L1)*(DEXP(-L1*TI)-DEXP(-L2*TI))
      N2=N2*0.012
      N3=K1/L3*(1.-DEXP(-L3*TI))-K1/(L3-L2)*(DEXP(-L2*TI)-DEXP(-L3*TI))
      A=DEXP(-L1*TI)/(L2-L1)/(L3-L1)
      B=DEXP(-L2*TI)/(L1-L2)/(L3-L2)
      C=DEXP(-L3*TI)/(L1-L3)/(L2-L3)
```

```
      N3=N3-K1*L2*(A+B+C)
      N3=N3*0.012
      N3B=K1/L3*(1.-DEXP(-L3*TI))-K1/(L3-L1)*(DEXP(-L1*TI)-DEXP(-L3*TI))
      N3B=N3B*0.988
      N3=N3+N3B
      NN1=N1*DEXP(-L1*DT)
      NN2=0.012*L1*N1/(L2-L1)*(DEXP(-L1*DT)-DEXP(-L2*DT))+N2*DEXP(-L2*DT
     1)
      P=L1*L2*DEXP(-L1*DT)/(L2-L1)/(L3-L1)
      Q=L1*L2*DEXP(-L2*DT)/(L1-L2)/(L3-L2)
      R=L1*L2*DEXP(-L3*DT)/(L1-L3)/(L2-L3)
      S=N2*L2/(L3-L2)*(DEXP(-L2*DT)-DEXP(-L3*DT))
      T=0.988*L1*N1/(L3-L1)*(DEXP(-L1*DT)-DEXP(-L3*DT))+N3*DEXP(-L3*DT)
      NN3=S+0.012*N1*(P+Q+R)+T
      MO95=ATM5-(NN1+NN2+NN3)
      MAS5=95./6.0248E23*MO95
      ATM7=K2*TI
      N4=K2/L4*(1.-DEXP(-L4*TI))
      N5=K2/L5*(1.-DEXP(-L5*TI))-K2/(L5-L4)*(DEXP(-L4*TI)-DEXP(-L5*TI))
      N5=N5*0.862
      N6=K2/L6*(1.-DEXP(-L6*TI))-K2/(L6-L5)*(DEXP(-L5*TI)-DEXP(-L6*TI))
      D=DEXP(-L4*TI)/(L5-L4)/(L6-L4)
      E=DEXP(-L5*TI)/(L4-L5)/(L6-L5)
      F=DEXP(-L6*TI)/(L4-L6)/(L5-L6)
      N6=N6-K2*L5*(D+E+F)
      N6=0.862*N6
      N6B=K2/L6*(1.-DEXP(-L6*TI))-K2/(L6-L4)*(DEXP(-L4*TI)-DEXP(-L6*TI))
      N6B=N6B*0.138
      N6=N6+N6B
      NN4=N4*DEXP(-L4*DT)
      NN5=0.862*L4*N4/(L5-L4)*(DEXP(-L4*DT)-DEXP(-L5*DT))+N5*DEXP(-L5*DT
     1)
      P1=L4*L5*DEXP(-L4*DT)/(L5-L4)/(L6-L4)
      Q1=L4*L5*DEXP(-L5*DT)/(L4-L5)/(L6-L5)
      R1=L4*L5*DEXP(-L6*DT)/(L4-L6)/(L5-L6)
      S1=N5*L5/(L6-L5)*(DEXP(-L5*DT)-DEXP(-L6*DT))
      T1=0.138*L4*N4/(L6-L4)*(DEXP(-L4*DT)-DEXP(-L6*DT))+N6*DEXP(-L6*DT)
      NN6=S1+0.862*N4*(P1+Q1+R1)+T1
      MO97=ATM7-(NN4+NN5+NN6)
      MAS7=97./6.0248E23*MO97
      ATM8=K3*TI
      N7=K3/L7*(1.-DEXP(-L7*TI))
      NN7=N7*DEXP(-L7*DT)
    7 MO98=ATM8-NN7
      MAS8=98./6.0248E23*MO98
      N8=K4/L8*(1.-DEXP(-L8*TI))
      NN8=N8*DEXP(-L8*DT)
      ACT9=NN8*L8/3.7E10
      MAS9=99./6.0248E23*NN8
      ATM1=Y5*NU*SIG*FLUX*TI
      MAS1=100./6.0248E23*ATM1
      MASS=MAS5+MAS7+MAS8+MAS9+MAS1
      SACT=ACT9/MASS
      WRITE(3,4)TC,ACT9,MAS5,MAS7,MAS8,MAS9,MAS1,MASS,SACT
    4 FORMAT(F6.1,F12.3,1P7E12.3)
   88 CONTINUE
   99 CONTINUE
      STOP
      END
```

APPENDIX B
TYPICAL OUTPUT

```
IRRADIATION TIME =   7.0  DAYS
 DECAY   ACTIVITY      MASS          MASS          MASS          MASS          MASS          MASS        SPECIFIC
 TIME      MO99        MO95          MO97          MO98          MO99          MO100      MOLYBDENUM    ACTIVITY
 DAYS      CURIE       GRAM          GRAM          GRAM          GRAM          GRAM          GRAM       CURIE/GRAM
  0.0     203.134    1.509E-06     7.269E-04     8.321E-04     4.235E-04     9.404E-04     2.924E-03    6.946E+04
  1.0     157.888    2.232E-06     8.107E-04     8.382E-04     3.292E-04     9.404E-04     2.921E-03    5.406E+04
  2.0     122.721    3.125E-06     8.424E-04     8.382E-04     2.558E-04     9.404E-04     2.880E-03    4.261E+04
  3.0      95.386    4.182E-06     8.543E-04     8.382E-04     1.989E-04     9.404E-04     2.836E-03    3.363E+04
  4.0      74.140    5.401E-06     8.588E-04     8.382E-04     1.546E-04     9.404E-04     2.797E-03    2.650E+04
  5.0      57.626    6.774E-06     8.605E-04     8.382E-04     1.201E-04     9.404E-04     2.766E-03    2.083E+04
  6.0      44.791    8.297E-06     8.611E-04     8.382E-04     9.338E-05     9.404E-04     2.741E-03    1.634E+04
  7.0      34.814    9.966E-06     8.614E-04     8.382E-04     7.258E-05     9.404E-04     2.723E-03    1.279E+04
  8.0      27.060    1.178E-05     8.615E-04     8.382E-04     5.641E-05     9.404E-04     2.708E-03    9.992E+03
  9.0      21.033    1.372E-05     8.615E-04     8.382E-04     4.385E-05     9.404E-04     2.698E-03    7.797E+03
 10.0      16.348    1.580E-05     8.615E-04     8.382E-04     3.408E-05     9.404E-04     2.690E-03    6.077E+03
 11.0      12.707    1.800E-05     8.615E-04     8.332E-04     2.649E-05     9.404E-04     2.685E-03    4.733E+03
 12.0       9.876    2.033E-05     8.615E-04     8.382E-04     2.059E-05     9.404E-04     2.681E-03    3.634E+03
 13.0       7.676    2.278E-05     8.615E-04     8.332E-04     1.600E-05     9.404E-04     2.679E-03    2.866E+03
 14.0       5.967    2.534E-05     8.615E-04     8.382E-04     1.244E-05     9.404E-04     2.678E-03    2.228E+03
 15.0       4.638    2.801E-05     8.615E-04     8.382E-04     9.668E-06     9.404E-04     2.678E-03    1.732E+03
 16.0       3.605    3.079E-05     8.615E-04     8.382E-04     7.515E-06     9.404E-04     2.678E-03    1.346E+03
 17.0       2.802    3.368E-05     8.615E-04     8.332E-04     5.841E-06     9.404E-04     2.680E-03    1.046E+03
 18.0       2.178    3.666E-05     8.615E-04     8.382E-04     4.540E-06     9.404E-04     2.681E-03    8.122E+02
 19.0       1.693    3.974E-05     8.615E-04     8.332E-04     3.529E-06     9.404E-04     2.683E-03    6.303E+02
 20.0       1.316    4.291E-05     8.615E-04     8.382E-04     2.743E-06     9.404E-04     2.680E-03    4.899E+02
```

```
IRRADIATION TIME =   8.0  DAYS
 DECAY   ACTIVITY      MASS          MASS          MASS          MASS          MASS          MASS        SPECIFIC
 TIME      MO99        MO95          MO97          MO98          MO99          MO100      MOLYBDENUM    ACTIVITY
 DAYS      CURIE       GRAM          GRAM          GRAM          GRAM          GRAM          GRAM       CURIE/GRAM
  0.0     212.492    2.236E-06     8.499E-04     9.518E-04     4.430E-04     1.075E-03     3.322E-03    6.397E+04
  1.0     165.162    3.156E-06     9.337E-04     9.580E-04     3.443E-04     1.075E-03     3.314E-03    4.984E+04
  2.0     128.374    4.268E-06     9.655E-04     9.580E-04     2.676E-04     1.075E-03     3.270E-03    3.926E+04
  3.0      99.731    5.566E-06     9.774E-04     9.580E-04     2.080E-04     1.075E-03     3.224E-03    3.095E+04
  4.0      77.556    7.044E-06     9.819E-04     9.580E-04     1.617E-04     1.075E-03     3.183E-03    2.436E+04
  5.0      60.281    8.676E-06     9.836E-04     9.580E-04     1.257E-04     1.075E-03     3.151E-03    1.913E+04
  6.0      46.854    1.052E-05     9.842E-04     9.580E-04     9.768E-05     1.075E-03     3.125E-03    1.499E+04
  7.0      36.413    1.250E-05     9.844E-04     9.580E-04     7.592E-05     1.075E-03     3.106E-03    1.173E+04
  8.0      28.306    1.455E-05     9.845E-04     9.580E-04     5.901E-05     1.075E-03     3.091E-03    9.158E+03
  9.0      22.002    1.694E-05     9.846E-04     9.580E-04     4.587E-05     1.075E-03     3.080E-03    7.143E+03
 10.0      17.101    1.939E-05     9.846E-04     9.580E-04     3.565E-05     1.075E-03     3.072E-03    5.566E+03
 11.0      13.292    2.193E-05     9.846E-04     9.580E-04     2.771E-05     1.075E-03     3.067E-03    4.334E+03
 12.0      10.331    2.470E-05     9.846E-04     9.580E-04     2.154E-05     1.075E-03     3.064E-03    3.372E+03
 13.0       8.030    2.756E-05     9.846E-04     9.580E-04     1.674E-05     1.075E-03     3.062E-03    2.623E+03
 14.0       6.242    3.055E-05     9.846E-04     9.580E-04     1.301E-05     1.075E-03     3.061E-03    2.039E+03
 15.0       4.851    3.366E-05     9.846E-04     9.580E-04     1.011E-05     1.075E-03     3.061E-03    1.585E+03
 16.0       3.771    3.690E-05     9.846E-04     9.580E-04     7.861E-06     1.075E-03     3.062E-03    1.231E+03
 17.0       2.931    4.025E-05     9.846E-04     9.580E-04     6.110E-06     1.075E-03     3.064E-03    9.567E+02
 18.0       2.278    4.371E-05     9.846E-04     9.580E-04     4.749E-06     1.075E-03     3.066E-03    7.431E+02
 19.0       1.771    4.728E-05     9.846E-04     9.580E-04     3.691E-06     1.075E-03     3.068E-03    5.771E+02
 20.0       1.376    5.095E-05     9.846E-04     9.580E-04     2.369E-06     1.075E-03     3.071E-03    4.481E+02
```