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DAUGHTER GROWTH IN FRESHLY SEPARATED
 ^{226}Ra , ^{227}Ac , AND ^{232}U

by

D. C. Stewart, E. S. Macias,
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Chemistry Division

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INTRODUCTION

Radium-226, actinium-227, and uranium-232 each act as the parent nuclide of a decay series of predominantly alpha-emitting daughters, making them of considerable interest for various isotopic source applications. Radium-226 and actinium-227 are each also the longest-lived isotopes of their respective elements. They thus are of prime importance to the chemist, anxious to work with the least radioactive material available when doing research on elemental properties.

For either type of use, applied or in research, it is often desirable to know the rate at which the radioactive daughters are regenerated into a freshly purified parent, either because of their effects on the projected use of the material or as a basis for estimating handling hazards and shielding requirements. The decay chains are complex for all three nuclides, however, so in the past it has scarcely been practicable to obtain the desired information by hand calculation. This report accordingly presents the daughter buildup curves for the three nuclides on a relative yield basis as calculated by computer techniques. The curves are also given in the possibly more useful form of radioactivity buildup with time. Since this last information is usually of most interest to the radiochemist during the time immediately after parent purification, representative values for buildup of the daughters in this period are also presented in tabular form.

CALCULATION METHODS

The programs employed were modified from those previously used to determine transplutonium-element yields from heavy-element targets exposed to high neutron fluxes in a reactor.¹ An IBM-1620(II) computer was employed exclusively for this earlier problem, the first programs used being based on the equations developed by Bateman.² This particular approach breaks down, however, in certain situations. In the reactor case, the Bateman method cannot be used if the daughter of an intermediate product itself captures enough neutrons to recycle back into the buildup chain. The presence of branching in decay chains has the same effect. To overcome these difficulties, additional programs based on numerical integration were

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developed.¹ Both types of programs were adapted so that the yield data can be graphed directly by accessory equipment associated with IBM-1620(II).

Since extensive branching occurs in the decay chains of all the nuclides considered in the present study, the numerical integration approach was used for the calculations. To shorten calculation time, the original program was modified to allow the yield data to be accumulated on a CDC-3600 computer. The data were then put into a form usable by the IBM-1620 with a special "converter" program, and that machine was used to prepare the final data in graphical as well as tabular form. The data-producing programs are presented in the appendix to this report.

The numerical-integration program cannot consider nuclides having half-lives shorter than the time increment used for the integration. Since lowering the time-increment value causes a proportionate lengthening of computer time, a compromise usually must be made. In the present study, two different time increments and time scales were used for each situation considered. The first scale was for 5 months with a 50-sec increment to obtain information on short-time changes; the second, a 50-yr scale with a 2×10^4 -sec (5.56-hr) increment. Since all the shorter-lived nuclides are in equilibrium well within 5 months, combination of the two scales gives an accurate picture of the yield situation over a 50-yr period for any daughters having a half-life greater than 50 sec.

The yield data were calculated in terms of the atoms of daughter relative to the original number of atoms of the parent heading the chain. The following relation was used to convert the yield data to radioactivity units:

$$\frac{\text{Dis/min of daughter per original gram of chain parent}}{(T_{1/2})_D(M)} = \frac{(0.693)(6.02 \times 10^{23})(\text{R.Y.})(60)}{(T_{1/2})_D(M)},$$

where

$(T_{1/2})_D$ = Half-life of daughter in seconds,

(M) = Mass number of chain parent,

and

(R.Y.) = Relative yield of daughter.

Since relative yield values were not available for any chain members having half-lives shorter than 50 sec, it was assumed in calculating total radioactivity that these very short-lived nuclides would come into essentially immediate equilibrium. They were accordingly assigned the same disintegration rate value as their immediate parent at any given time.

The following was assumed where branching in the decay chains occurred:

Formation half-life of A = $(100/X\%) \times$ gross-destruction half-life of C. In certain cases, where the value of X was extremely low (^{206}Hg from ^{210}Pb as an example), yield data are not shown on the graphs, since it would not be possible to do so on any reasonable set of ordinates. In all these cases, however, the low-yield daughter is short-lived and can be assumed to come to rapid equilibrium with the immediate parent. It will thus have a buildup curve of the same shape as that parent but at a proportionately lower level as set by the branching ratio.

RESULTS; DISCUSSION, AND CONCLUSIONS

Radium-226

Figure 1(a) shows the complete decay chain of ^{226}Ra ; Figs. 1(b) and 1(c) indicate the adaptations made for the computer programs for the 5-month and 50-yr time scales, respectively. Figures 2 and 3 show the daughter buildup in fresh ^{226}Ra for the two time scales. Table I gives numerical values for the corresponding radioactivity changes immediately after purification, and Figs. 4 and 5 give the radioactivity buildup for the longer intervals.

The four alpha emitters (^{222}Rn , ^{218}Po , ^{218}At , and ^{214}Po) and three beta emitters (^{214}Bi , ^{214}Pb , and ^{210}Tl) of the upper part of the chain come to equilibrium in about a month's time and, along with the parent ^{226}Ra , dominate the radioactivity picture for at least a decade. The activity buildup in the lower part of the chain is controlled by 22-yr ^{210}Pb . After the 5-day ^{210}Bi comes to equilibrium with its ^{210}Pb parent, all the buildup curves of the lower chain follow the ^{210}Pb curve at levels determined by the branching ratios. The overall activity is still increasing at the end of the 50-yr time span considered in the calculations.

Actinium-227

The complete decay chain for ^{227}Ac is presented in Fig. 6(a); Figs. 6(b) and 6(c) show the simplifications made for the 5-month and 50-yr calculations, respectively. Figure 7 shows the relative yield curves for the 5-month period, and Fig. 8 shows the corresponding activity buildups. Table II gives the radioactivity change for the period immediately after parent purification.

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Fig. 1. Radium-226 Decay

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Fig. 2. Early Daughter Growth in ^{226}Ra

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Fig. 3. Long-time Daughter Growth in ^{226}Ra TABLE I. Short-time Activity Growth (dis/min) into One gram of Purified ^{226}Ra *

Elapsed Time, sec x 10 ⁵ **	^{222}Rn	^{218}Po	^{218}At	^{214}Pb	^{214}Bi	^{210}Tl	^{210}Pb	^{210}Bi	^{210}Po	Total Activity
0.001	4.50×10^8	1.20×10^8	2.40×10^4	3.50×10^6	1.30×10^5	2.54×10^1	-	-	-	2.16×10^{12}
0.005	2.26×10^9	1.41×10^9	2.82×10^5	1.31×10^8	1.28×10^7	3.71×10^3	-	-	-	2.16
0.01	4.50	3.57	7.14	6.16	1.08×10^8	3.60×10^4	2.93×10^1	-	-	2.17
0.02	9.04	8.03	1.61×10^6	2.55×10^9	7.92	2.92×10^5	4.32×10^2	-	-	2.18
0.05	2.25×10^{10}	2.16×10^{10}	4.32	1.26×10^{10}	7.42×10^9	2.89×10^6	1.13×10^4	2.10×10^1	-	2.22
0.1	4.46	4.36	8.72	3.38	2.69×10^{10}	1.06×10^7	9.50	3.82×10^2	-	2.31
0.2	8.86	8.75	1.75×10^7	7.78	7.06	8.24×10^8	5.80×10^5	5.22×10^3	-	2.48
0.5	2.15×10^{11}	2.14×10^{11}	4.28	2.05×10^{11}	1.98×10^{11}	8.50	4.61×10^6	1.13×10^5	7.67×10^1	2.99
1	4.07	4.07	8.14	3.96	3.92	8.60	1.94×10^7	9.76	1.39×10^3	3.74
2	7.38	7.38	1.48×10^8	7.27	7.27	8.61	7.60	7.60×10^6	2.23×10^4	5.08
5	1.40×10^{12}	1.40×10^{12}	2.80×10^8	1.40×10^{12}	1.39×10^{12}	8.64×10^8	4.03×10^8	9.50×10^7	7.42×10^5	7.74×10^{12}

*1 gram pure ^{226}Ra = 2.16×10^{12} dis/min.**One day = 8.64×10^4 sec.

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Fig. 4. Early Radioactivity Growth in ^{226}Ra

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Fig. 5. Long-time Radioactivity in ^{226}Ra

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Fig. 6. Actinium-227 Decay³¹

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Fig. 7. Early Daughter Growth in ^{227}Ac

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Fig. 8. Early Radioactivity Growth in ^{227}Ac TABLE II. Short-time Activity Growth (dis/min) into One gram of Purified ^{227}Ac *

Elapsed Time, sec x 10 ³ **	^{223}Fr	^{219}At	^{215}Bi	^{227}Th	^{223}Ra & ^{219}Rn	^{215}Po	^{211}Pb	^{211}Bi	^{211}Po	^{207}Tl	Total
0.001	9.83×10^{10}	2.98×10^6	2.92×10^5	6.91×10^9	5.44×10^6	5.72×10^6	1.24×10^5	3.78×10^4	1.21×10^3	5.40×10^3	1.58×10^{14}
0.005	4.43×10^{11}	1.68×10^7	5.26×10^6	3.46×10^{10}	9.36×10^7	9.90×10^7	6.23×10^6	3.55×10^6	1.14×10^4	1.07×10^6	1.59
0.01	7.85	3.06	1.55×10^7	6.91	3.32×10^8	3.47×10^8	3.92×10^7	2.83×10^7	9.1×10^4	1.28×10^7	1.59
0.02	1.24×10^{12}	4.93	3.60	1.38×10^{11}	1.12×10^9	1.16×10^9	2.40×10^8	2.02×10^8	6.5×10^5	1.30×10^8	1.60
0.05	1.77	7.02	6.55	3.46	4.86	4.93	2.12×10^9	2.00×10^9	6.4×10^6	1.68×10^9	1.61
0.1	1.89	7.52	7.27	6.91	1.30×10^{10}	1.30×10^{10}	8.39	8.17	2.61×10^7	7.60	1.61
0.2	1.90	7.56	7.34	1.38×10^{12}	3.30	3.31	2.66×10^{10}	2.64×10^{10}	8.45×10^7	2.55×10^{10}	1.62
0.5	1.90	7.56	7.34	3.43	1.20×10^{11}	1.20×10^{11}	1.10×10^{11}	1.09×10^{11}	3.49×10^8	1.07×10^{11}	1.65
1	1.90	7.56	7.34	6.77	3.54	3.54	3.36	3.35	1.06×10^9	3.31	1.70
2	1.90	7.56	7.34	1.32×10^{13}	1.12×10^{12}	1.12×10^{12}	1.09×10^{12}	1.09×10^{12}	3.49×10^9	1.08×10^{12}	1.81
5	1.90×10^{12}	7.56×10^7	7.34×10^7	3.10×10^{13}	5.47×10^{12}	5.47×10^{12}	5.44×10^{12}	5.40×10^{12}	1.73×10^{10}	5.40×10^{12}	2.30×10^{14}

*1 gram pure ^{227}Ac = 1.58×10^{14} dis/min.**One day = 8.64×10^4 sec.

In spite of its complexity, the ^{227}Ac decay chain comes to relatively rapid equilibrium since the longest-lived daughter in the chain is the 18.17-day ^{227}Th . Yield and activity curves for the 50-yr period are not given for this reason. All the curves have essentially reached equilibrium on the far right side of Figs. 7 and 8 (complete equilibrium occurs at about 1.3×10^7 sec or 0.41 yr). At this point, all the curves for the daughters in the main decay branch have reached their maximum values and subsequently follow the straight-line decay of the ^{227}Ac parent.

The ^{233}Fr - ^{219}Ac - ^{215}Bi side chain reaches equilibrium at about 2×10^4 sec (5.55 hr), then similarly follows the parent ^{227}Ac decay.

Of the nine products dominating the activity situation in Fig. 7, six are basically alpha emitters, while only three decay primarily or completely by beta emission.

Uranium-232

The complete and simplified decay chains of ^{232}U and its daughters are shown in Fig. 9. Relative yield data for the 5-month calculation are shown in Fig. 10; radioactivity buildup for both the 5-month and 50-yr intervals are presented in Fig. 11. The very short-term radioactivity growth of the system is given in Table III.

The relative yield curve of the ^{232}U daughters is not presented for the 50-yr period. The fact that the longest-lived member of the chain (1.9-yr ^{228}Th) is the first daughter formed means that all of the shorter, more "junior" daughters follow the shape of the ^{228}Th curve. This can be seen in the insert of Fig. 11. The daughters below ^{228}Th come into effective equilibrium with that nuclide within a few months, and the entire system comes to equilibrium with the ^{232}U parent in about a decade. Subsequent change of all components is then by the straight-line decay of ^{232}U .

At the point of maximum relative yield (about 3×10^8 sec), the values are:

$$^{228}\text{Th} \quad 2.36 \times 10^{-2}$$

$$^{224}\text{Ra} \quad 1.24 \times 10^{-4}$$

$$^{212}\text{Pb} \quad 1.50 \times 10^{-5}$$

$$^{208}\text{Pb} \quad 6.26 \times 10^{-2}$$

The decay chain effectively contains six equally active alpha-emitting sources (^{232}U , ^{228}Th , ^{224}Ra , ^{230}Rn , ^{216}Po , and the ^{212}Bi plus ^{212}Po combination) at equilibrium. There is the equivalent of two equally active

beta systems (^{212}Po and the ^{212}Bi plus ^{208}Tl combination). The 72-yr half-life of the ^{232}U parent makes this nuclide a particularly attractive one for many isotopic power applications.

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Fig. 9. Uranium-232 Decay³

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Fig. 10. Early Daughter Growth in ^{232}U

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Fig. 11. Radioactivity in Purified ^{232}U TABLE III. Short-time Activity Growth (dis/min) into One gram of Purified $^{232}\text{U}^*$

Elapsed Time, sec x 10 ⁵ **	^{228}Th	^{224}Ra	^{220}Rn & ^{216}Po	^{212}Pb	^{212}Bi	^{212}Po	^{208}Tl	Total
0.001	5.40×10^7	8.93×10^3	6.34×10^3	7.52×10^0	-	-	-	4.64×10^{13}
0.005	2.69×10^8	1.64×10^4	1.48×10^4	5.15×10^2	1.53×10^1	1.01×10^1	2.05×10^0	4.64
0.01	5.40	6.23×10^5	5.90×10^5	3.82×10^3	1.99×10^2	1.32×10^2	3.64×10^1	4.65
0.02	1.08×10^9	2.44×10^6	2.37×10^6	2.97×10^4	2.79×10^3	1.84×10^3	6.52×10^2	4.65
0.05	2.69	1.50×10^7	1.48×10^7	4.46×10^5	9.14×10^4	6.06×10^4	2.64×10^4	4.66
0.1	5.40	5.90	5.90	3.45×10^6	1.20×10^6	7.96×10^5	3.71×10^5	4.66
0.2	1.08×10^{10}	2.35×10^8	2.35×10^8	2.62×10^7	1.39×10^7	9.22×10^6	4.50×10^6	4.67
0.5	2.69	1.44×10^9	1.44×10^9	3.56×10^8	2.70×10^8	1.79×10^8	8.96×10^7	4.67
1	5.40	5.54	5.51	2.29×10^9	2.00×10^9	1.33×10^9	6.70×10^8	4.68
2	1.08×10^{11}	2.07×10^{10}	2.08×10^{10}	1.26×10^{10}	1.19×10^{10}	7.89×10^9	3.96×10^9	4.71
5	2.69×10^{11}	1.06×10^{11}	1.06×10^{11}	8.78×10^{10}	8.60×10^{10}	5.70×10^{10}	2.90×10^{10}	4.75×10^{13}

*1 gram pure ^{232}U = 4.71×10^{13} dis/min.**One day = 8.64×10^4 sec.

APPENDIX

Data-producing Programs

THE TWO PROGRAMS BELOW ARE THOSE USED FOR THE ACTUAL DATA CALCULATIONS ON THE CDC 3600 AND IBM 1620 (II) COMPUTERS. THE PROGRAM FOR CONVERTING THE DATA OUTPUT BETWEEN THE TWO COMPUTERS IS NOT GIVEN, NOR IS THE GRAPHING PROGRAM WITH ITS EXTENSIVE SERIES OF SUBROUTINES. ANY OF THE NON-INCLUDED MATERIAL CAN BE FURNISHED UPON REQUEST.

CDC 3600 PROGRAM FOR CALCULATING GROWTH AND DECAY TO OBTAIN YIELDS

INSTRUCTIONS

THIS PROGRAM CALCULATES YIELD DATA FOR ANY COMBINATION OF STARTING MATERIALS. THE INPUT DATA CONSISTS OF A CARD FOR EACH MEMBER OF THE BUILDUP AND DECAY SCHEME LISTING ITS INITIAL CONCENTRATION, ITS BUILDUP AND DECAY HALF-LIVES AND CROSS SECTIONS, AND ITS PARENT(S) AND DAUGHTER(S), AND CARDS LISTING THE NEUTRON FLUX AND TIME SCALE. FOR DAUGHTER GROWTH PROBLEMS THE NEUTRON FLUX SHOULD BE SET EQUAL TO ZERO. BUILDUP AND DECAY SCHEMES OF ANY COMPLEXITY MAY BE USED. THIS PROGRAM WILL HANDLE UP TO 45 GROWTH AND DECAY PATHS. SIZE CAN BE INCREASED BY CHANGING THE DIMENSION STATEMENT.

PROGRAM

```

DIMENSION SPEC(45),Y(45),A(45),B(45),T(6),YIELD(45,6),IA(45)
X,IB(45),SIGA(45),SIGB(45),HALFA(45),HALFB(45),IS(45),IP(45),IR(45)
X,YY(45)
1 FORMAT(2X,I2,4X,I2)
2 FORMAT(5X,E8,1)
5 FORMAT(A5,36X,E11,2,1X,F9,2,I2,F9,4)
6 FORMAT(A5,6X,A5,1X,E11,2,F10,2,I2,3X,E11,2,1X,F9,2,I2)
7 FORMAT(A5,6H FROM ,A5,1X,E11,2,F10,2,I2,1X,E11,2,1X,F9,2,I2,F9,4)
8 FORMAT(/5HTIME ,1X,6(F11,5))
9 FORMAT (A5, 2X, 6(E9,2, 2X))
10 FORMAT(6F11,6)
80 FORMAT(3X,F11,6)
160 FORMAT(F10,8,I2,3X,I2,3X,I1,4X,I1)
210 FORMAT(5HFLUX=,E8,1)
309 FORMAT(4(5X,F10,2))
712 FORMAT(74H
STION      INITIAL)
1713 FORMAT(60H      SECTION      HALFLIFE      XSECTION
1CONC)
4001 CONTINUE
READ1,NT
PUNCH1,NT
READ 80,DT
READ 1, N,NS
PUNCH1,N,NS
READ 2, FLUX
READ 309, TMAX,TL,TD,YMIN
PUNCH309, TMAX,TL,TD,YMIN
PUNCH712
PUNCH713
DOT = DT/2.
DO 150 K=1,N
READ 6,SPEC(K),PRED,SIGA(K),HALFA(K),IA(K),SIGB(K),HALFB(K),IB(K)
READ 160,Y(K),IS(K),IP(K),IR(K)
IF(IP(K))280,290,280
280 PUNCH7,SPEC(K),PRED,SIGA(K),HALFA(K),IA(K),SIGB(K),HALFB(K),IB(K),
XY(K)
PRINT7,SPEC(K),PRED,SIGA(K),HALFA(K),IA(K),SIGB(K),HALFB(K),IB(K),
XY(K)
GO TO 150
290 PUNCH 5,SPEC(1),SIGB(1),HALFB(1),IB(1),Y(1)
PRINT 5,SPEC(1),SIGB(1),HALFB(1),IB(1),Y(1)
150 CONTINUE
130 TIME=0.
DO 155 M=1,N
MS=IS(M)
Y(MS)=Y(M)
155 SPEC(MS)=SPEC(M)
DO 57 K=1,N
IF(IP(K))350,360,350
350 IAA=IA(K)
HALFAA=HALFA(K)

```

```

      SIGAA=SIGA(K)
      CALL TIMCO(IAA,HALFAA,SIGAA,FLUX,E)
      A(K)=E
      GO TO 370
360  A(K)=0.
370  IF(IR(K))102,101,102
101  IBB=IB(K)
      HALFBB=HALFB(K)
      SIGBB=SIGB(K)
      CALL TIMCO(IBB,HALFBB,SIGBB,FLUX,E)
      B(K)=E
      GO TO 57
102  B(K)=0.
57  CONTINUE
18  DO 77 K=1,NT
      READ 10,(T(L),L=1,6)
      PUNCH 8,(T(L), L=1,6)
      PRINT 8,(T(L), L=1,6)
      PUNCH 210,FLUX
      PRINT 210,FLUX
70  DO 79 L=1,6
74  DO 73 I=1,N
      MS=IS(I)
      MP=IP(I)
73  Y(MS)=Y(MS)+(A(I)*Y(MP)-B(I)*Y(MS))*DT
      TIME = TIME + DT
      EK=ABSF(TIME-T(L))
      IF (EK-DOT)72,72,4000
4000 IF(TIME-T(L))74,72,72
72  DO 76 J=1,NS
      YIELD(J,L)=Y(J)
76  CONTINUE
79  TX=T(L)
      DO 78 J = 1,NS
      PRINT 9, SPEC(J),(YIELD(J,L),L=1,6)
78  PUNCH 9, SPEC(J),(YIELD(J,L),L=1,6)
77  CONTINUE
      GO TO 4001
      END
      SUBROUTINE TIMCO(IC,HALFC,SIGC,FLUX,C)
      GO TO (30,31,32,33,34,36),IC
30  HALFC=HALFC*.000001
      GO TO 35
31  HALFC=HALFC*.00006
      GO TO 35
32  HALFC=HALFC*.0036
      GO TO 35
33  HALFC=HALFC*.0864
      GO TO 35
34  HALFC=HALFC*31.56
35  D=.69315/HALFC
      GO TO 37
36  D=0.
37  C=D+FLUX*SIGC*1000000.
      RETURN
      END

```

IBM 1620 (II) PROGRAM FOR CONVERTING YIELDS TO RADIOACTIVITY
(WORD LENGTH 1004)

INSTRUCTIONS

THIS PROGRAM GIVES ACTIVITY DATA IN DISINTEGRATIONS PER MINUTE PER GRAM OF STARTING MATERIAL USING CONVERTED YIELD DATA. TO CONSIDER SHORT-LIVED NUCLIDES NOT LISTED IN YIELD DATA ADD A CARD GIVING THE NUMBER OF EXTRA NUCLIDES TO BE CONSIDERED ACCORDING TO FORMAT(2X,I2) AT THE BEGINNING OF THE INPUT DATA. THEN ADD A SEPARATE CARD FOR EACH NEW NUCLIDE WITH THE NAME OF THE NUCLIDE, THE NAME OF ITS PREDECESSOR, ITS HALF-LIFE AND A NUMBER TELLING THE UNITS OF HALF-LIFE (1,SEC,2,MIN,3,HR,4,DAYS,5,YR,6,NOT APPLICABLE,7,MSEC) ACCORDING TO FORMAT (A5,1X,A5,1X,F10.6,I2). THE NEW NUCLIDE WILL BE GIVEN THE HALF-LIFE OF ITS PREDECESSOR. IF THE NEW NUCLIDE IS PART OF A BRANCH FROM ITS PREDECESSOR BE SURE ENOUGH OF THE NEW NUCLIDE IS FORMED TO JUSTIFY SETTING ITS ACTIVITY EQUAL TO ITS PREDECESSOR. IF NOT, DISREGARD THE NUCLIDE.

PROGRAM

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      DIMENSION ELMENT(30),T(6),Y(6),TOTAL(30),SPACT(30),HFLF(30)
      X, IA(30),D(30),SON(20),DAD(20),HHFLF(20),NIA(20)
      DIMENSION REMEM(196),MEMORY(30)

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1  FORMAT(2X,12,4X,12)
2  FORMAT(5X,E8,1)
3  FORMAT(5HFLUX=,E8,1)
9  FORMAT(A5,2X,6(E9,2,2X)) ,
11 FORMAT(A5,1X,A5,1X,F10,6,12)
20 FORMAT(25HSW1 ON FOR EXTRA NUCLIDES)
30 FORMAT(19HSW2 ON FOR NEW CASE)
93 FORMAT(22HELEMENT      HALF LIFE)
94 FORMAT(64HACTIVITY FROM YIELD DATA IN DIS./MIN. PER GRAM STARTING
  XMATERIAL)
121 FORMAT(A5,47X,F10,2,12)
122 FORMAT(A5,2X,6(E9,2,2X))
127 FORMAT(A5,2X,6(E9,2,2X))
131 FORMAT(A5,2X,F7,2)
304 FORMAT(1H )
305 FORMAT(5HTIME ,1X,6(F11,5))
309 FORMAT(4(5X,F10,2))
421 FORMAT(5HTOTAL,2X,6(E9,2,2X))
550 FORMAT(35HPRINT MASS NO. OF STARTING MATERIAL)
552 FORMAT(F9,3)
31  DO43 I=1,30
43  MEMORY(I)=-1
    DO186 I=1,196
186 REMEM(I)=0.
    PRINT 550
    ACCEPT552,AMN
    PRINT20
    PAUSE 21
    IF(SENSE SWITCH 1)22,23
22  READ1,NEXTRA
    READ11,(SON(I),DAD(I),HHFLF(I),NIA(I),I=1,NEXTRA)
    IAA=0
    SWITCH=NEXTRA-1
    IF(SENSE SWITCH12)44,45
45  J=NEXTRA+1
85  J=J-1
    K=0
    I=0
10  I=I+1
    SWITCH=SON(I)-SON(J)
    IF(SENSE SWITCH12)40,41
40  IAA=IAA+1
    IF(K)14,14,13
13  MEMORY(K)=0
14  MEMORY(I)=0
    MEMORY(J)=1
    K=J
41  SWITCH=I-J+1
    IF(SENSE SWITCH12)42,10
42  SWITCH=J-2
    IF(SENSE SWITCH12)44,85
44  GO TO 4001
23  NEXTRA=0
4001 CONTINUE
24  READ1,NT
    READ 1, N,NS
    PUNCH 1,NT
    NESS=N+NEXTRA+1-IAA
    NSS=NS+NEXTRA+1
    PUNCH1,NESS,NSS
    READ309,TMAX,TL,TD,YMIN
    PUNCH309,TMAX,TL,TD,YMIN
    PUNCH94
    PUNCH93
    READ304
    READ304
    READ121,(ELEMENT(K),HFLF(K),IA(K),K=1,N)
    DO 142 K=1,N
    IF(K-N)69,68,69
69  IK=K
66  IK=IK+1
    SWITCH=ELEMENT(K)-ELEMENT(IK)
    IF(SENSE SWITCH 12)142,67
67  SWITCH=IK-N
    IF(SENSE SWITCH12)68,66
68  IAA=IA(K)
    EE=ELEMENT(K)
    HL=HFLF(K)
    CALL LIFE(IAA,EE,HL)
142 CONTINUE
    IF(SENSE SWITCH 1)25,26

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25 PUNCH304
DO 12 I=1,NEXTRA
IF(I-NEXTRA)70,71,70
70 IK=I
72 IK=IK+1
SWITCH=SON(I)-SON(IK)
IF(SENSE SWITCH)12,74
74 SWITCH=IK-NEXTRA
IF(SENSE SWITCH 12)71,72
71 NI=NIA(I)
SO=SON(I)
HHFL=HHFLF(I)
CALL LIFE(NI,SO,HHFL)
12 CONTINUE
26 PUNCH 304
PUNCH304
DO160 NI=1,N
SPACT(NI)=0.
160 CONTINUE
DO308 I=1,NT
DO33 ICC=1,6
33 TOTAL(ICC)=0.
READ304
READ305,(T(L),L=1,6)
PUNCH305,(T(L),L=1,6)
READ2,FLUX
PUNCH3,FLUX
DO307 J=1,NS
MST=0
M=0
READ122,E,(Y(L),L=1,6)
II=1
125 IF(E-ELMENT(II))123,124,123
123 II=II+1
GO TO 125
124 IAI=IA(II)
CALL FREDDY(IAI,C)
136 DO126 IC=1,6
IF(HFLF(II))200,201,200
201 SPACT(IC)=0.
GO TO 126
200 SPACT(IC)=C*Y(IC)*60./(HFLF(II)*AMN)
TOTAL(IC)=SPACT(IC)+TOTAL(IC)
126 CONTINUE
PUNCH9,E,(SPACT(IZ),IZ=1,6)
IF(SENSE SWITCH 1)6,307
6 MST=MST+1
SWITCH=DAD(MST)-E
IF(SENSE SWITCH 12)4,5
4 IF(MEMORY(MST))51,52,53
51 PUNCH9,SON(MST),(SPACT(IZ),IZ=1,6)
56 DO7 IEJ=1,6
7 TOTAL(IEJ)=TOTAL(IEJ)+SPACT(IEJ)
5 CONTINUE
IF(MST-NEXTRA)6,307,6
52 MSM=MST*7
LL=0
MMM=MSM+6
DO 54 L=MSM,MMM
LL=LL+1
54 REMEM(L)=SPACT(LL)
GO TO 56
53 MSM=MEMORY(MST)*7
DO63 JI=1,6
SPACT(JI)=SPACT(JI)+REMEM(MSM)
63 MSM=MSM+1
GO TO 51
307 CONTINUE
PUNCH421,(TOTAL(IX),IX=1,6)
IF(I-NT)424,308,308
424 PUNCH 304
308 CONTINUE
PRINT30
PAUSE31
IF(SENSE SWITCH2)31,4001
END
SUBROUTINE FREDDY(IA,C)
SUBPROGRAM 'FREDDY II'
GO TO (40,41,42,43,44,45),IA
40 C=4.17E+23
GO TO 46

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41 C=6.95E+21
   GO TO 46
42 C=1.16E+20
   GO TO 46
43 C=4.83E+18
   GO TO 46
44 C=1.32E+16
   GO TO 46
45 C=0.
46 RETURN
   END
C  SUBROUTINE LIFE(IAA,EL,HF)
   SUBPROGRAM 'LIFE I'
149 FORMAT(A5,5X,F10.2,8H SECONDS)
150 FORMAT(A5,5X,F10.2,8H MINUTES)
151 FORMAT(A5,5X,F10.2,6H HOURS)
152 FORMAT(A5,5X,F10.2,5H DAYS)
153 FORMAT(A5,5X,F10.2,6H YEARS)
154 FORMAT(A5,5X,F10.2)
155 FORMAT(A5,5X,F10.4,12HMILLISECONDS)
   GO TO(143,144,145,146,147,148,160),IAA
143 PUNCH149,EL,HF
   GO TO 142
144 PUNCH150,EL,HF
   GO TO 142
145 PUNCH151,EL,HF
   GO TO 142
146 PUNCH152,EL,HF
   GO TO 142
147 PUNCH153,EL,HF
   GO TO 142
148 PUNCH154,EL,HF
   GO TO 142
160 PUNCH155,EL,HF
142 RETURN
   END

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REFERENCES

1. J. Milsted, P. R. Fields, and D. N. Metta, Nucl. Appl. 1, 136-144 (1965).
2. H. Bateman, Proc. Cambridge Phil. Soc. 15, 423 (1910).
3. G. Friedlander, J. W. Kennedy, and J. M. Miller, *Nuclear and Radiochemistry*, 2nd ed., 9-11 (John Wiley and Sons, New York, 1964).