ACTA CHEMICA SCANDINAVICA 12 (1958) 377—398

Composition of Products Formed by Thermal Neutron Fission of ^{235}U

II. Chemical Composition of Stable and Radioactive Fission Products

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The chemical composition of stable and radioactive fission products has been calculated for seven different irradiation times between 1 day and 2 years, and for three different neutron fluxes, 10^{12}, 10^{13} and 10^{14} n cm^{-2} sec^{-1}. The results are given in the form of curves showing the β-activity (Figs. 1—7) and mole fraction (Figs. 9—15) of each element as a function of cooling time from 1 day up to about 15 years. The calculations are based on nuclear data published before July 1957 [1].

For the processing of used nuclear fuels, the chemical composition of the fission products has to be known. This composition depends on a number of variables, such as the composition of the fuel "elements", the neutron energy, flux, and irradiation time in the reactor, cooling time outside the reactor, etc. If data for the fission yields, decay schemes, halflives and neutron cross sections of the fission product isotopes are known (see Part I of this series [1]), the chemical composition of the fission products formed in thermal uranium reactors may be calculated as a function of neutron flux (Φ), irradiation time (T) and cooling time (t).

Diagrams and tables of the β-activity of the fission isotopes of ^{235}U have been given by Hunter and Ballou [2], and Björnerstedt *et al.*[3] for instantaneous fission ($T = 0$), and, for various irradiation and cooling times, by Lock [4], and Moteff [5], and as an Appendix to *Progress in Nuclear Chemistry* [6]. The relative elemental activity of the most important fission products is given as a function of cooling time by Coryell *et al.*[7] and Rubinson *et al.*[8] for unpublished flux and irradiation time, and by Keneshea and Saul [10] for $\Phi = 10^{12}$ n cm^{-2} sec^{-1} and T = 1—60 d. Lock [4] gives tables of the elemental composition of the fission products obtained after three irradiation and cooling times at a very high flux (10^{15} n cm^{-2} sec^{-1}). Walton [11] has made a similar, though less comprehensive, calculation.

In this paper the β-activity and mole fraction of each fission element is calculated as a function of cooling time for various irradiation times between 1 day and 2 years and for neutron fluxes between 10^{12} and 10^{14} n cm^{-2} sec^{-1}. The calculations are based on nuclear data which have appeared in the literature up to July 1957 [1].

METHODS OF CALCULATION

A fission product isotope may be formed in several different ways: 1) as a primary fission product, 2) as a decay product in one or several chains, starting with primary fission products, or 3) through neutron capture. All these modes of formation have to be considered separately for each isotope. The elemental composition of the fission products is obtained by adding the corresponding isotopic values.

In the calculations, the following symbols are used:

A_n = activity (in disintegrations/time) of isotope in n:th generation of a primary ($n = 1$) fission product;
X_n = mole fraction (in percent) of an isotope in the n:th generation;
Φ = neutron flux (in neutrons per cm^2 per second);
T = irradiation time;
t = cooling time;
y_n = fission yield (in percent) of an isotope in a decay chain;
λ_n = disintegration constant (in time^{-1}); $\lambda_n = 0$ for stable isotopes;
τ_n = half-life; $\tau_n = \ln 2/\lambda_n$; in the calculations, τ_1 is used in a decay chain for the first isotope which has a half-life of at least one hour;
σ_n = thermal neutron capture cross section (in cm^2)
Λ_n = effective disintegration constant ($= \lambda_n + \sigma_n \cdot \Phi$).

The following equations give the relationships between activity and mole fraction of an isotope and their dependence on neutron flux, irradiation and cooling times.

$$A_n(\Phi, T, t) = R_A \cdot B(\Phi, T, t) \text{ curies per gram (gFP)} \tag{1}$$

where

$$R_A = 8.07 \cdot 10^3 \cdot y_n/T \text{ curies per gram (C/gFP); } T \text{ in days} \tag{2}$$

and

$$X_n(\Phi, T, t) = R_x \cdot B(\Phi, T, t) \text{ mole percent} \tag{3}$$

where

$$R_x = \frac{y_n}{2 \cdot \lambda_n \cdot T} \text{ mole percent} \tag{4}$$

As will be discussed in the paragraph on "burn up", R_A and R_x have this form only so long as the amount of ^{235}U is constant.

In order to convert mole percent for a certain element to weight percent (g of fission element per gram mixed fission products) the relation

$$X \text{ mole percent} = 8.6 \cdot 10^{-3} \cdot X \cdot M \text{ weight percent} \tag{5}$$

should be used, wehre M is the atomic weight of the element.

From eqns. (1—4) one obtains the simple relation

$$X_n \text{ mole percent} = \frac{6.20 \times 10^{-5}}{\lambda_n} A_n \text{ C/gFP; } \lambda_n \text{ in days}^{-1} \quad (6)$$

$B\ (\Phi,\ T,\ t)$ is calculated according to Rubinson [12].

$$\mathrm{B}\ (\Phi,\ T,\ t) = \prod_{i=1}^{n} \lambda_i \sum_{i=1}^{n} [(\prod_{j=1}^{i} \Lambda_j^{-1}) \sum_{j=1}^{i} b_j\ (1 - e^{-\Lambda_j T}) \sum_{k=1}^{n} a_k \cdot e^{-\lambda_k t}] \quad (7)$$

where $b_j = 1$ when $i = 1$,
and when $i > 1$

$$b_i = \prod_{m \neq j} \frac{\Lambda_m}{\Lambda_m - \Lambda_j},\ (m = 1,\ 2,\ \ldots\ldots\ i;\ \Lambda_m \neq \Lambda_j) \quad (7\,a)$$

and $a_k = 1$ when $i = n$,
and when $i < n$,

$$a_k = \prod_{p \neq k} (\lambda_p - \lambda_k)^{-1},\ k = i,\ i + 1,\ \ldots . n;\ \lambda_p \neq \lambda_k) \quad (7\,b)$$

In no practical cases (*i.e.* for $t > 1$ day) has it been necessary to consider $n > 5$. In the rare cases when $\Lambda_m = \Lambda_j$ and $\sigma_m = \sigma_j = 0$, eqn. 7 is no more valid. The following equation can then be used for calculating A_2

$$A_2\ (T,\ t) = R_A\ ((1 + \lambda t)e^{-\lambda t} - [1 + \lambda(T + t)]e^{-\lambda(T+t)}) \quad (8)$$

The complicated eqn. (7) can in most practical cases be considerably simplified. The following equations have been found very useful:

For $n = 1$, one obtains

$$A_1\ (\Phi,\ T,\ t) = R_A \frac{\lambda_1}{\Lambda_1} (1 - e^{-\Lambda_1 T})\ e^{-\lambda_1 t} \quad (9)$$

A similar equation is obtained for $X_1\ (\Phi,\ T,\ t)$.

For all $\sigma_i = 0$, where $i = 1,\ 2,\ \ldots .\ n$, one obtains

$$A_n\ (T,\ t) = R_A \sum_{i=1}^{n} (\prod_{j \neq i} \frac{\lambda_j}{\lambda_j - \lambda_i})\ (1 - e^{-\lambda_i T})\ e^{-\lambda_i t} \quad (10)$$

A similar equation is obtained for $X_n\ (T,\ t)$.

For $\Lambda_2 = 0$ and $\Lambda_1\ T \ll 1$ one obtains

$$X_2\ (\Phi,\ T,\ t) = \frac{y_1 \lambda_1}{2\Lambda_1} \left(\frac{\Lambda_1 T}{2} - \frac{\Lambda_1^2 T^2}{6} + \frac{\Lambda_1^3 T^3}{24} - \ldots . .\right) \quad (11)$$

For all $\sigma_i = 0$ and $\lambda_n = 0$ where i and $n > 1$, one obtains

$$X_n\ (T,\ t) = \frac{y_n}{2} - \sum_{i=1}^{n-1} X_i\ (T,\ t) \quad (12)$$

If $\sigma_1 = \lambda_1 = 0$, eqn. (11) reduces to $X_1\ (T,\ t) = y_1/2$.

For the exact calculation of the mole fractions, the factor 2, used in the denominators in the above eqns. (4), (11) and (12), should rather be 2.0025, because of the formation of ^4He through triple fission (yield 0.25 %).

DISTRIBUTION OF THE β-ACTIVITY OF THE ELEMENTS

The β-activity of the fission elements is given as a function of cooling time at various irradiation times and neutron fluxes in Figs. 1—7. Only nuclear β decays are considered here; the curves therefore do not include any "β-activity" due to internal gamma-beta conversion. In addition, very long-lived activities are summarized in Table 1.

Table 1. Long-lived isotopes with activities practically independent of T and $t < 600$ years.

Isotope	Half-life years	Decay	Activity C/g fiss.prod.
^{79}Se	6.5×10^{4}	β, γ	8.7×10^{-4}
^{87}Rb	6.2×10^{10}	β	6.1×10^{-10}
^{93}Zr	9.5×10^{5}	β	9.8×10^{-5}
^{99}Tc	2.1×10^{5}	β	4.3×10^{-3}
^{107}Pd	5×10^{6}	β	6.1×10^{-7}
^{115}In	6×10^{14}	β	2.6×10^{-16}
^{129}I	1.7×10^{7}	β, γ	6.6×10^{-7}
^{135}Cs	2.1×10^{6}	β	4.1×10^{-5}
^{142}Ce	5.1×10^{15}	α	1.8×10^{-12}
^{144}Nd	2.2×10^{15}	α	3.8×10^{-13}
^{147}Sm	1.3×10^{11}	α	2.8×10^{-10}

In the figures, the solid lines refer to a neutron flux of 10^{13} n cm^{-2} sec^{-1}. If other curves are obtained at lower or higher fluxes, this is indicated by dotted lines for 10^{12} and dashed lines for 10^{14} n cm^{-2} sec^{-1}. The effect of burn up of ^{235}U at high $\Phi \cdot T$ is discussed in a separate paragraph below.

In some cases in Figs. 1—7, only one isotope contributes to the activity of an element (*e.g.* ^{99}Mo, ^{140}Ba, ^{147}Nd), but in most cases the element contains several radioactive isotopes. However, generally only one isotope predominates at a time but, close to inflexion points, two isotopes may contribute about equally. By consulting the data in Part I[1], it is often possible to reveal the isotopic composition of the β-activity of the elements in Figs. 1—7.

Only nine elements are considerably influenced by variation in the neutron flux. They are Tc, Pd, Ag, Xe, Cs, Pm, Sm, Eu and Tb. The various fluxes influence the disintegration curves in two ways: the activity either increases (Pd, Ag, Cs, (Pm), Eu, Tb) or decreases (Eu, Tc, Xe, (Pm), Sm) with increasing neutron flux. In the former case a shorter lived isotope is formed through neutron capture in a longer-lived or stable isotope of the element. In the latter case a shorter-lived isotope captures a neutron forming a longer-lived or stable isotope of the element (*cf.* Part I[1]).

In Fig. 8, the total β-activity of the combined fission elements is given as a function of T and t (1 d $\leq t \leq$ 1 400 years). The curve for $T = 0$ was taken from Björnerstedt *et al.*[3]. If the curve for $T = 0$ is compared with the statistically calculated curve of Way and Wigner [13], one finds that their curve in some parts lies about a factor of 1.5 above the curve given by Björnerstedt

et al. On the other hand, the curve for $T = 0$ in Fig. 8 agrees almost completely with the curve given by Thornton and Houghton [14], who made calculations from known values of individual fission products.

Because the methods of calculation are, in principle, the same, almost complete agreement is obtained with the curves given for various irradiation and cooling times by Koontz and Jarrett [15], and from the summation of the curves for the individual isotopes given in Ref [6].

ELEMENTAL DISTRIBUTION OF FISSION PRODUCTS

The mole fractions of the fission product elements are given in Figs. 9—15 as a function of cooling time for the same irradiation times as given in the previous figures. While the relative activities of the fission elements vary considerably with cooling time, it is immediately seen from Figs. 9—15 that the mole fractions of the elements change very little with cooling times. For some elements, there is practically no change at all; these elements are given at the right margin of the figures. The considerable variation of the curves for Nb, Sb, and Pm is due to the fact that these fission elements consist mainly of relatively short-lived isotopes.

From comparisons of Figs. 1—7 and 9—15 it is seen that many of the elements, which make up a considerable fraction of the mass of the fission products contribute relatively little to the total radioactivity. Thus, about 50 % of the weight of the fission products comes from elements Xe, Nd and Cs, while these elements only make up about 1 % of the total β-activity (*cf.* Figs. 3 and 11 for $T = 1$ month and $t = 2$ months).

ESTIMATED ERRORS

The primary data used in these calculations are impaired by certain errors. One of these errors, *i.e.* unknown modes of branching decay, must here be neglected.

The half-lives are generally fairly accurate, but for long half-lives the standard deviations are assumed to be $\lesssim$ 10 %.

The fission yields and thermal neutron cross sections have often been determined with neutrons obtained from "thermal" uranium reactors, in which also fast neutrons are present. However, since the capture cross sections are higher in the thermal than in the fast neutron energy region, and the cross sections have relatively little influence on A and X except in a few cases, the errors in the cross sections can here be neglected.

The fission yields may be subject to much larger errors because of the presence of fast neutrons. In uranium reactors about 3 % of the fissions may be caused by fast neutrons [16]. This is especially important for the mass numbers in the valley of the fission yield curve, because the fission yields there increase considerably with neutron energy. The standard deviations in the fission yields are assumed to lie between 2 and 10 %.

With the complicated equations used for calculating A and X, it is very difficult to calculate exactly the errors due to uncertainties in the primary

Figs. 1—7. Elemental composition of fission product β-activity (in curies per g fiss. prod.) as a function of cooling time for three different neutron fluxes: ····· 10^{12}, ——— 10^{13} and ------- 10^{14} n cm^{-2} sec^{-1}. T = irradiation time.

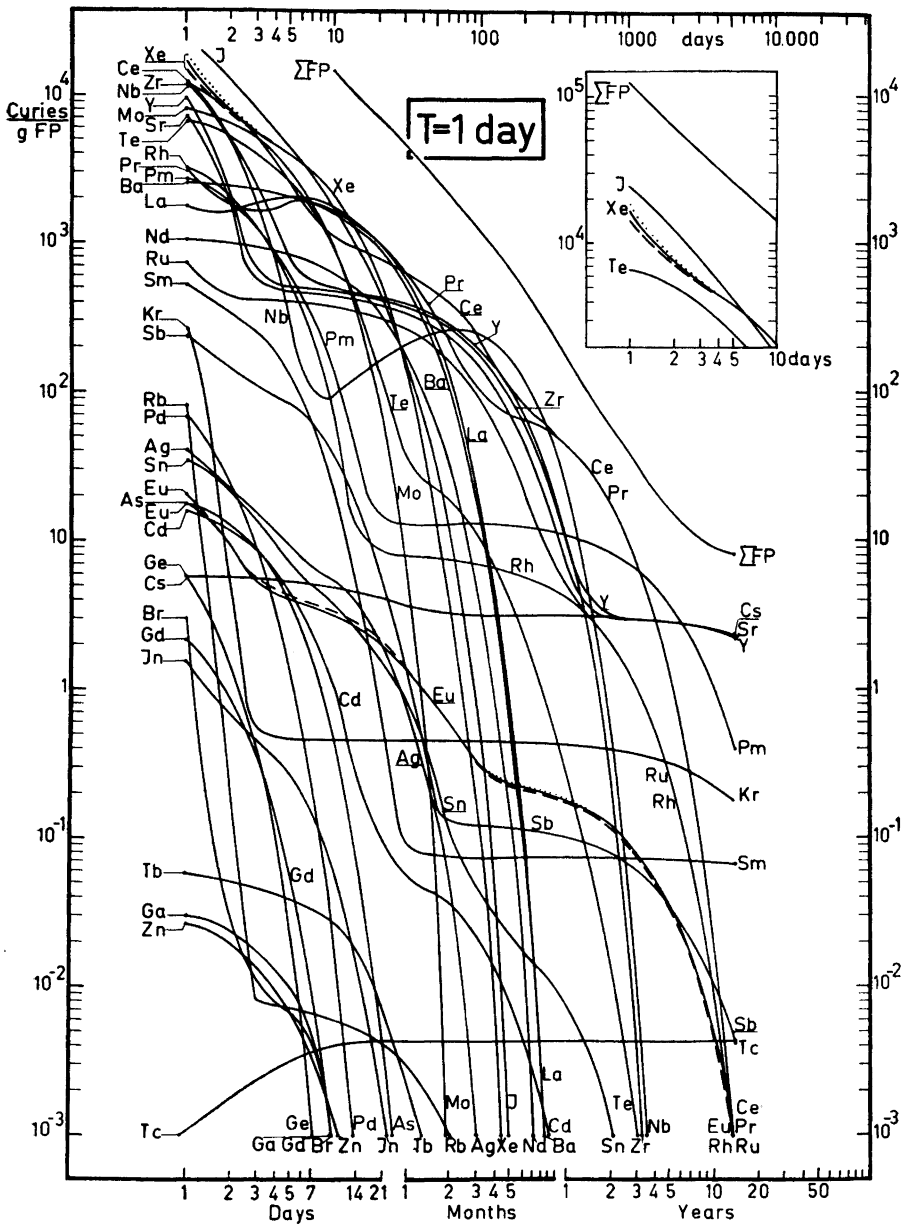

Fig. 1.

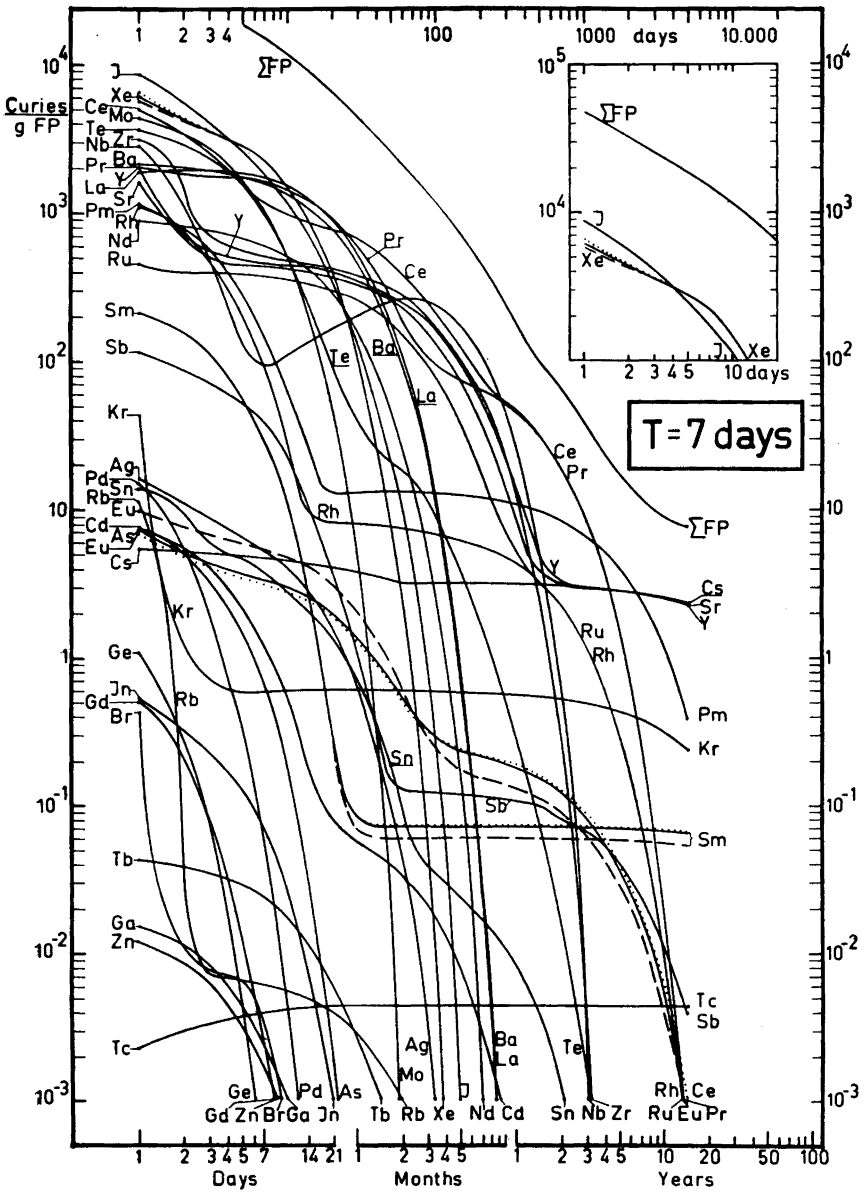
Curies
g FP
1 2 3 4
100
1000 days
10.000
ΣFP
J
Xe
Ce
Mo
Te
Nb
Zr
Pr
Ba
Y
La
Sr
Pm
Rh
Nd
Ru
Sm
Sb
Kr
Ag
Pd
Sn
Rb
Eu
Cd
As
Eu
Cs
Ge
In
Gd
Br
Tb
Ga
Zn
Tc
Pr
Ce
Te
Ba
La
Rh
Y
Ce
Pr
ΣFP
Cs
Sr
Y
Ru
Rh
Pm
Kr
Sn
Sb
Sm
Tc
Sb
Ag
Mo
Ba
La
Te
Rh
Ce
Ge
Gd Zn Br Ga In
Pd
As
Tb Rb Xe Nd Cd
Sn Nb Zr
Ru Eu Pr
Days
Months
Years
T=7 days
10 days

Fig. 2.

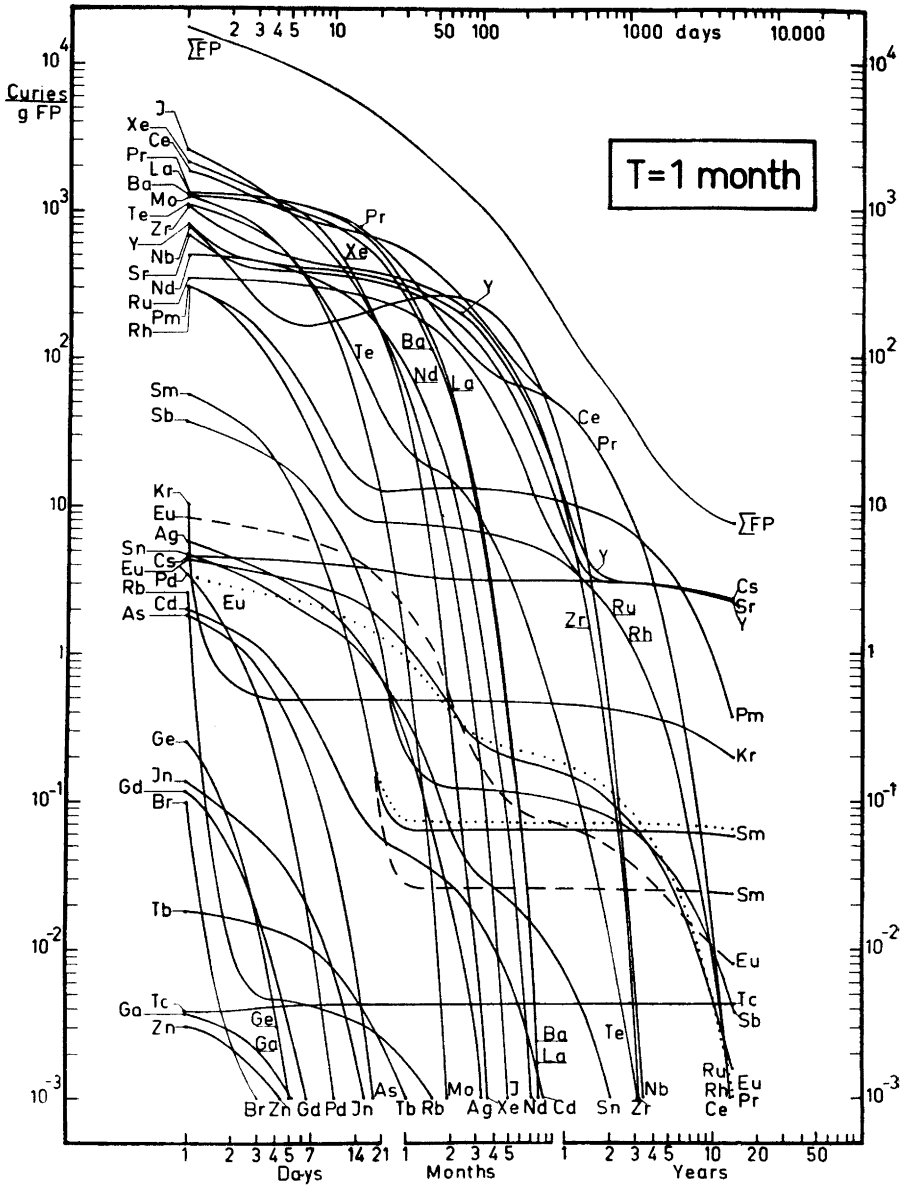
T=1 month
Curies
g FP
2 3 4 5 10 50 100 1000 days 10.000
ΣFP
Days
Months
Years
Br Zn Gd Pd In As Tb Rb Mo Ag Xe Nd Cd Sn Zr Nb Ru Rh Ce Pr Eu

Fig. 3.

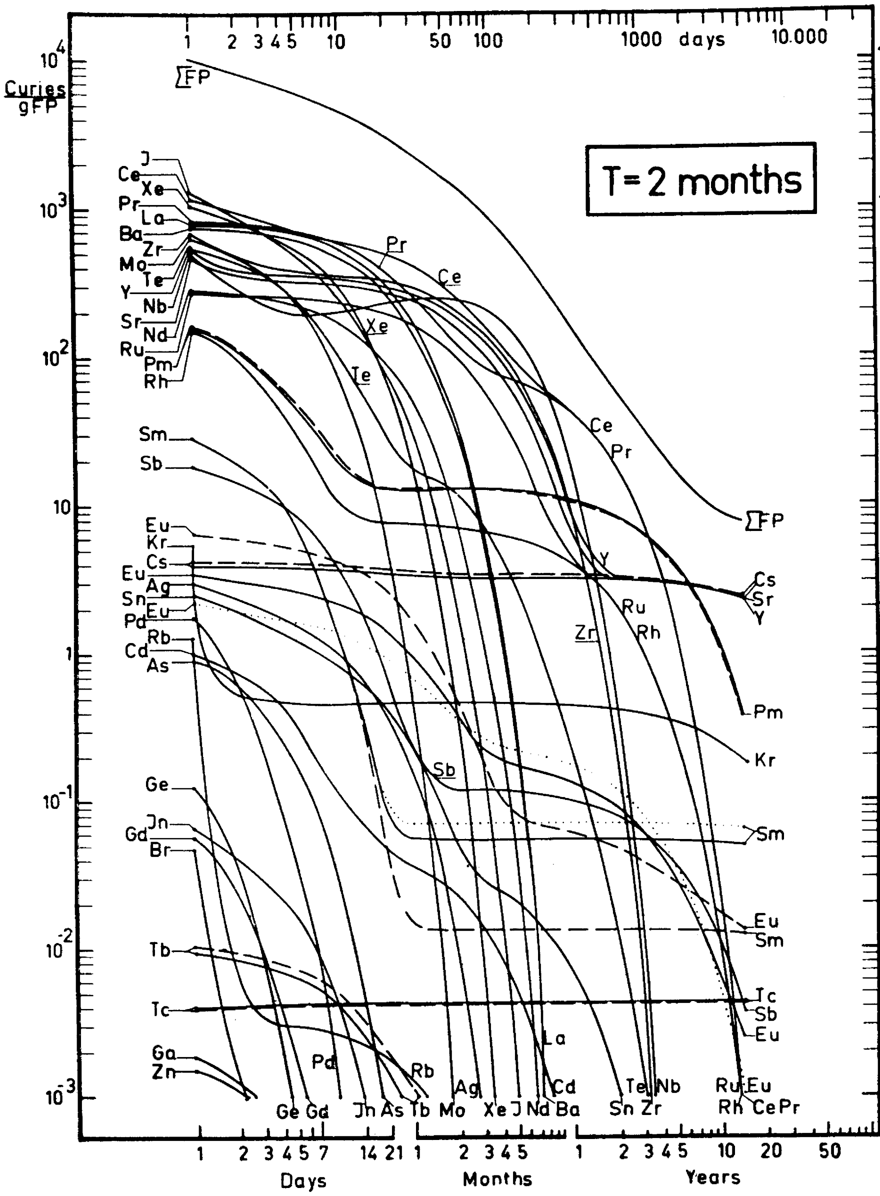
T=2 months
Curies
gFP
1 2 3 4 5 10 50 100 1000 days 10.000
1 2 3 4 5 7 14 21 1 2 3 4 5 1 2 3 4 5 10 20 50
Days
Months
Years

Fig. 4.

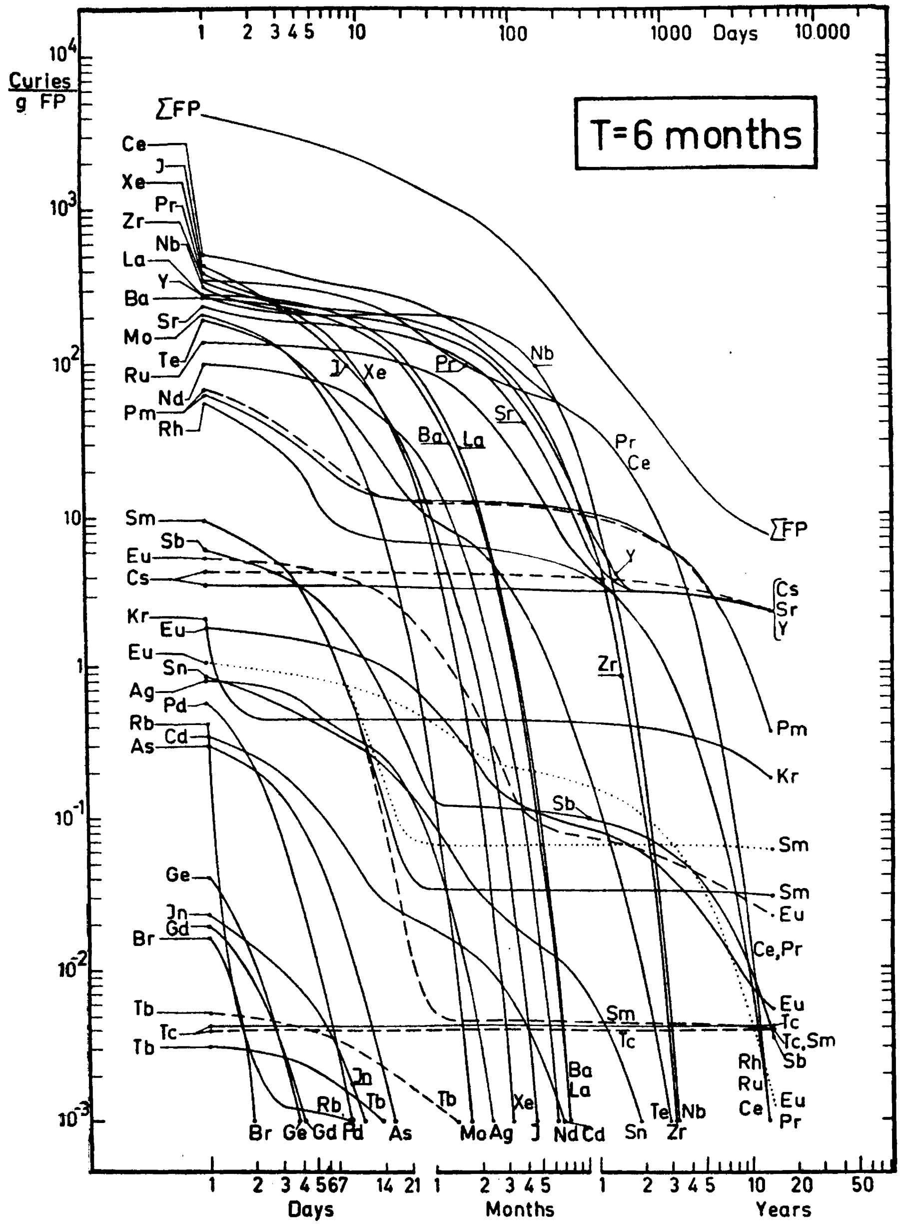

Fig. 5.

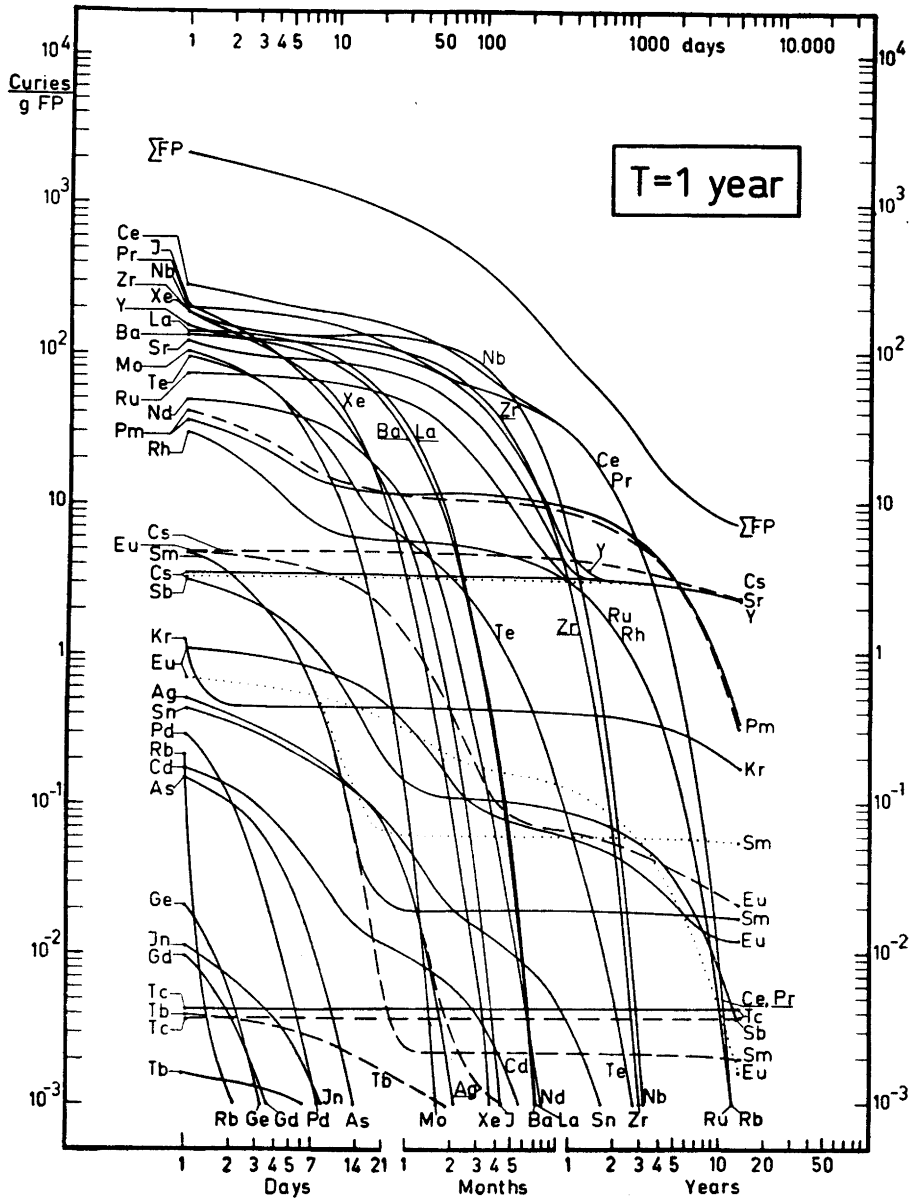
Curies
g FP
1 2 3 4 5 10 50 100 1000 days 10.000
T=1 year
∑FP
Ce
Pr
J
Nb
Zr
Xe
Y
La
Ba
Sr
Mo
Te
Ru
Nd
Pm
Rh
Cs
Eu
Sm
Sb
Kr
Ag
Sn
Pd
Rb
Cd
As
Ge
In
Gd
Tc
Tb
Days
Months
Years

Fig. 6.

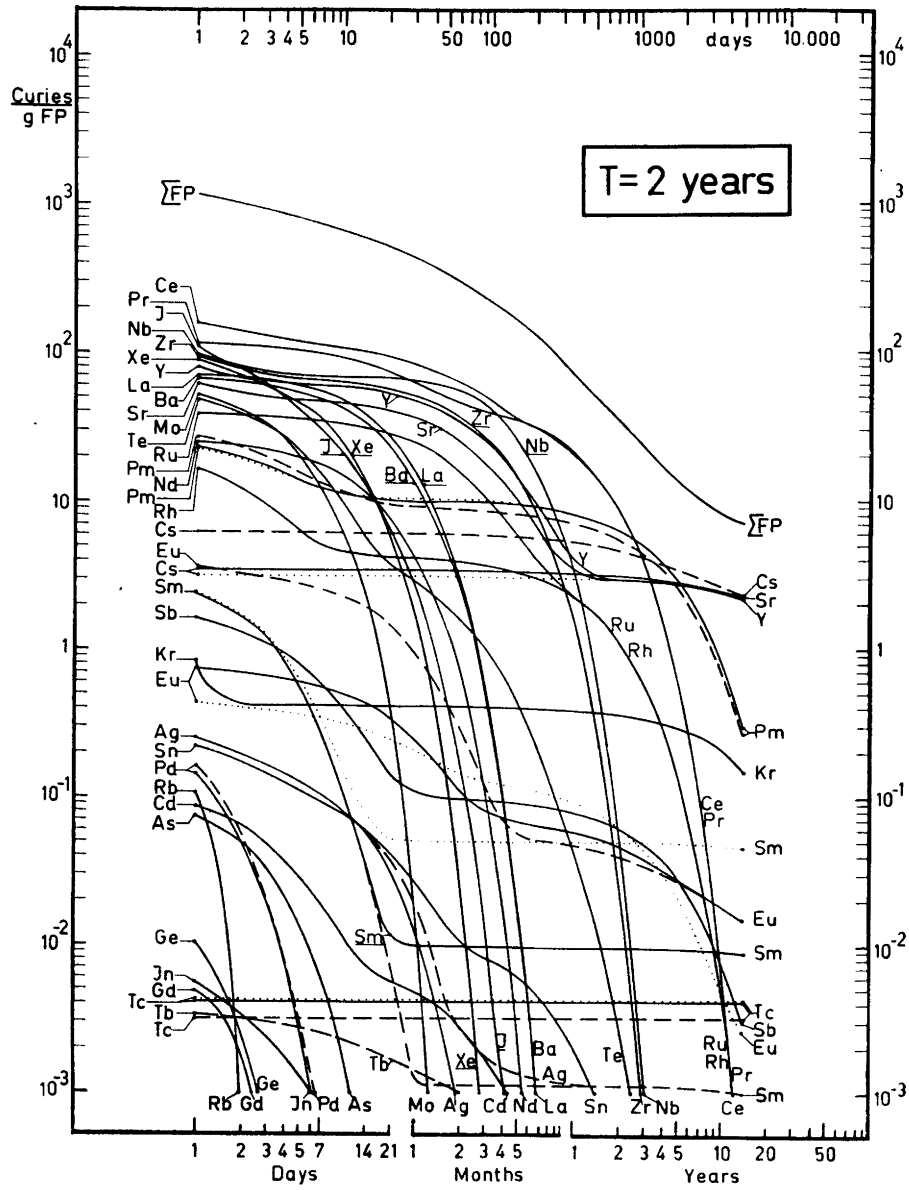
T= 2 years
Curies
g FP
1 2 3 4 5 10 50 100 1000 days 10.000
Days
Months
Years
ΣFP
Ce
Pr
Nb
Zr
Xe
Y
La
Ba
Sr
Mo
Te
Ru
Pm
Nd
Rh
Cs
Eu
Sm
Sb
Kr
Ag
Sn
Pd
Rb
Cd
As
Ge
In
Gd
Tc
Tb
I
Pm
Ce
Pr
Rb Gd In Pd As
Mo Ag Cd Nd La Sn Zr Nb Ce

Fig. 7.

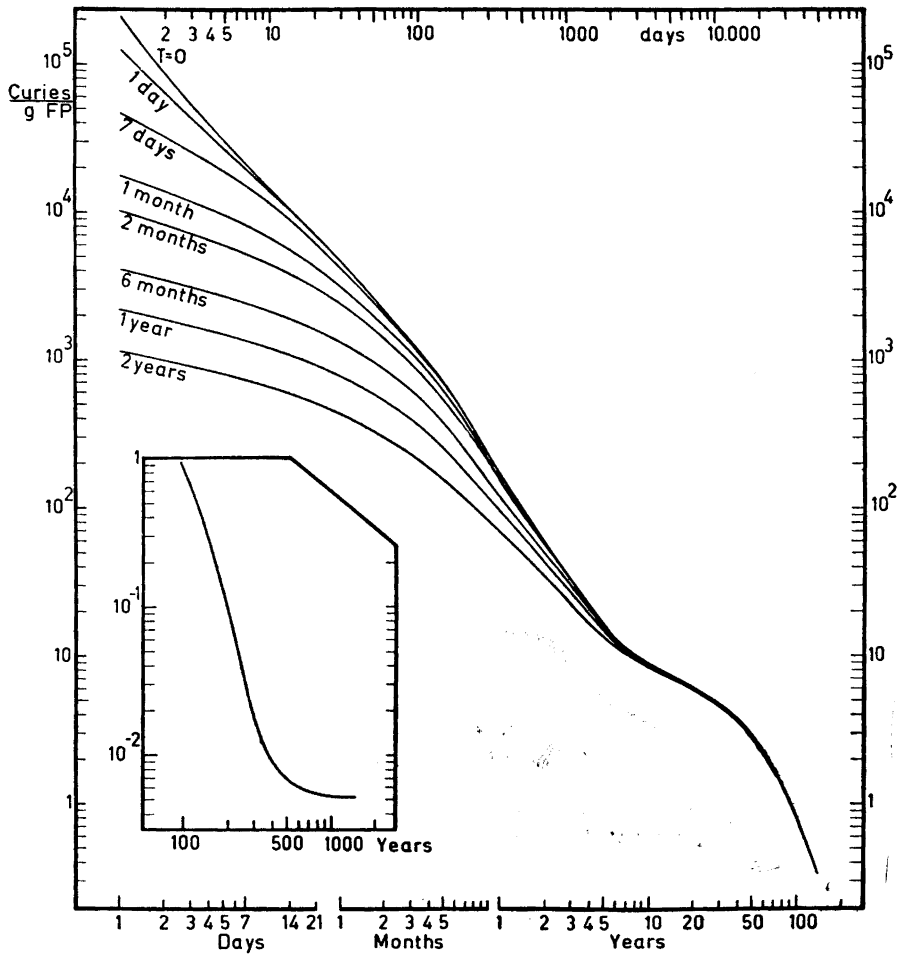

Fig. 8. Total β-activity of the fission products (in curies per g fiss. prod.) as a function of cooling time for irradiation times between 0 and 2 years. The curves are almost independent of neutron fluxes $\leq 10^{14}$ n cm^{-2} sec^{-1}.

Figs. 9—15. Mole percent of fission product elements as a function of cooling time for three different neutron fluxes: ····· 10^{12}, ———— 10^{13} and ------- 10^{14} n $cm^{-2}sec^{-1}$. T = irradiation time.

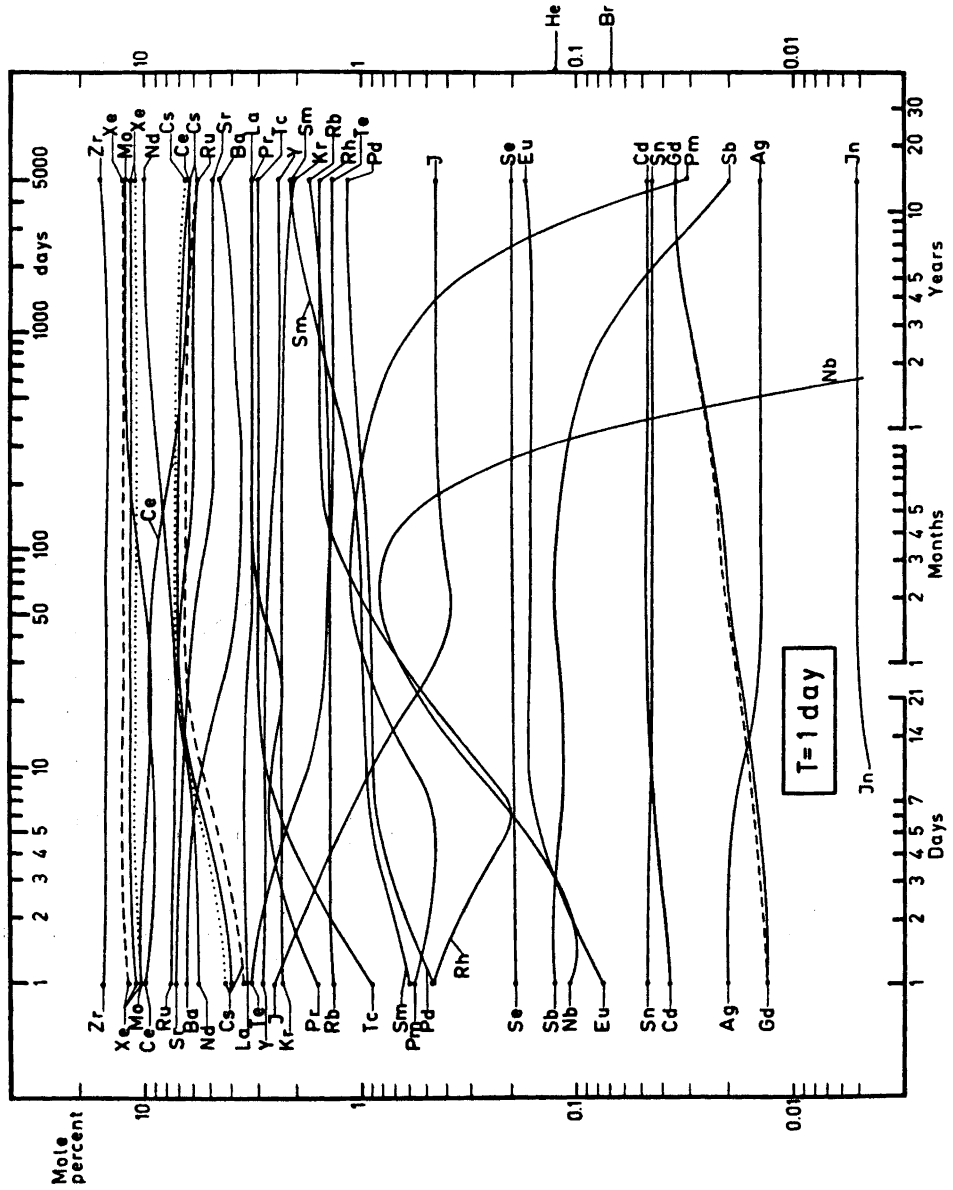

Fig. 9.

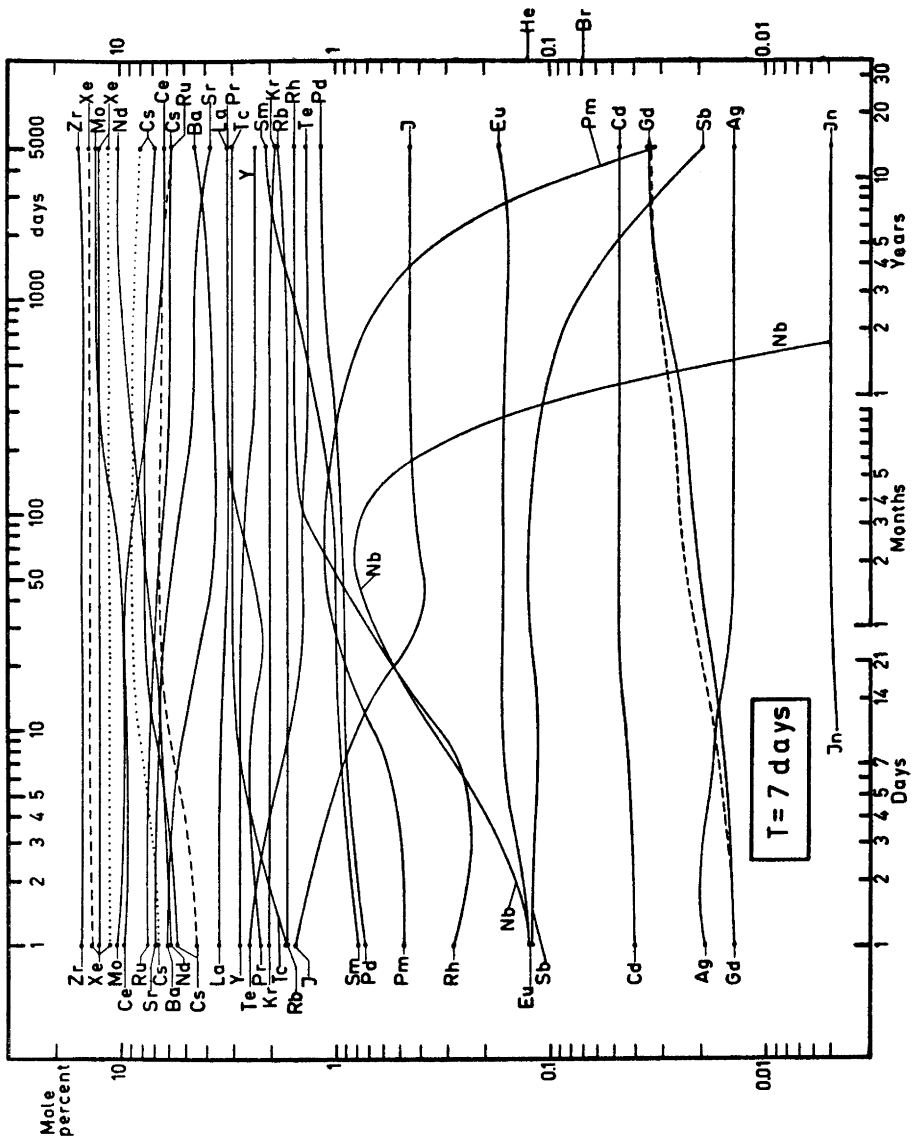
Mole percent
10
1
0.1
0.01
He
Br
0.1
0.01
days
5000
1000
100
50
10
1
Years
30
20
10
5
4
3
2
1
Months
5
4
3
2
1
Days
21
14
7
5
4
3
2
1
T = 7 days
Zr Xe Mo Nd Cs Ce Cs Ba Ru Sr La Pr Tc Y Sm Kr Rb Rh Te Pd
J
Eu
Pm Cd Gd
Sb
Ag
Jn
Nb
Nb
Nb
Zr Xe Mo Ce Ru Sr Cs Ba Nd Cs La Y Te Pr Kr Tc Rb J
Sm Pd Pm Rh Nb Eu Sb Cd Ag Gd
Jn

Fig. 10.

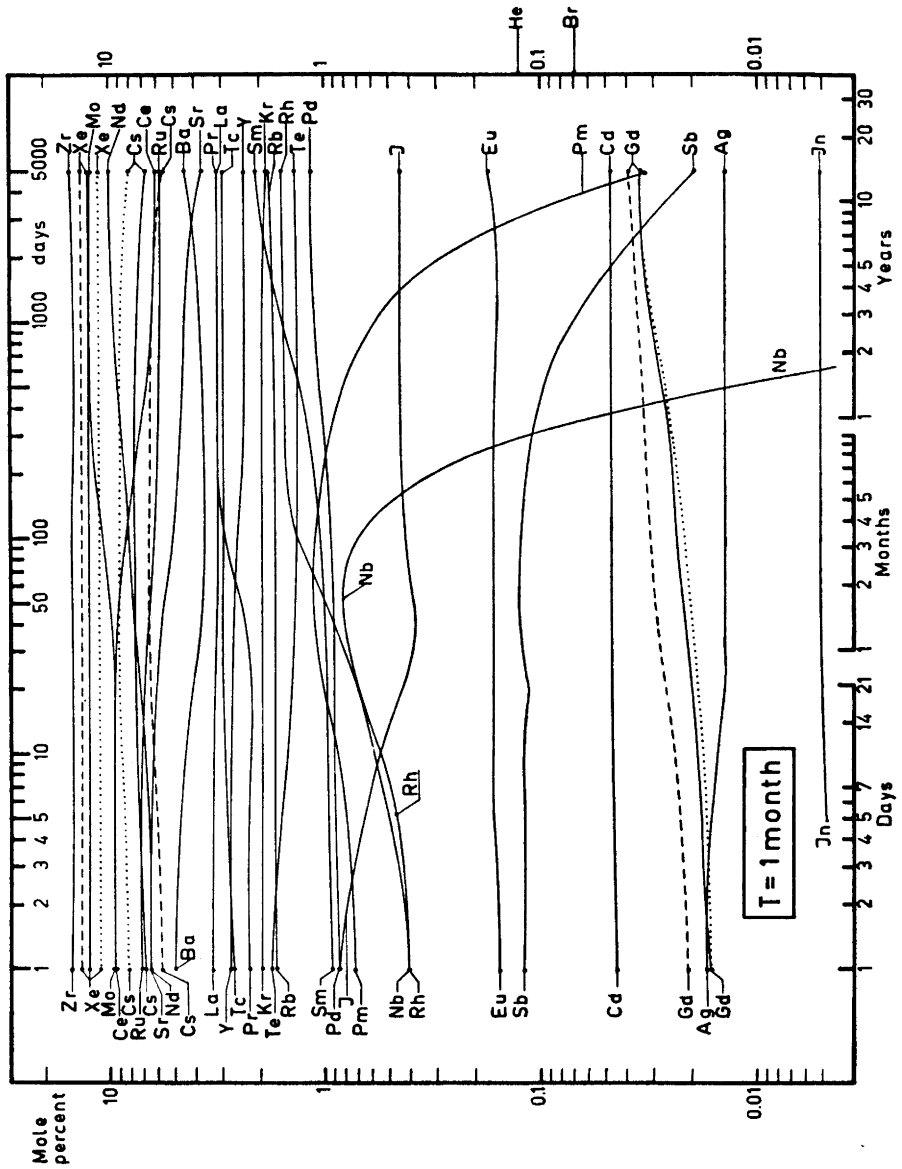
Mole percent
10
1
0.1
0.01
He
Br
days
1000
5000
100
50
10
1
Years
Months
Days
T=1month
Zr
Xe
Mo
Nd
Cs
Ce
Ru
Ba
Sr
Pr
La
Tc
Y
Sm
Kr
Rb
Rh
Te
Pd
J
Eu
Pm
Cd
Gd
Sb
Ag
In
Nb

Fig. 11.

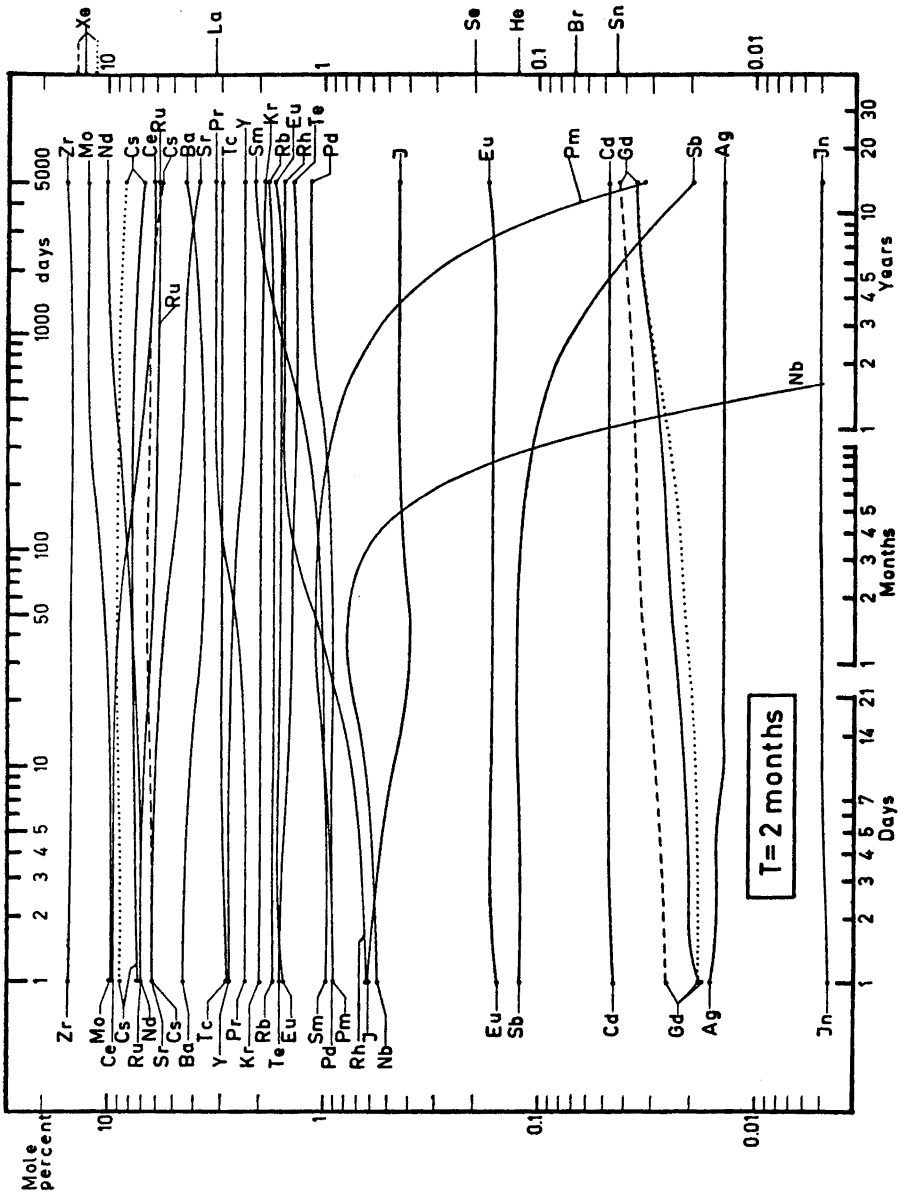
Xe
La
Se
He
Br
Sn
10
1
0.1
0.01
Zr
Mo
Nd
Cs
Ce
Ru
Cs
Ba
Sr
Pr
Tc
Y
Sm
Kr
Rb
Eu
Rh
Te
Pd
J
Eu
Pm
Cd
Gd
Sb
Ag
In
5000
1000 days
100
50
10
1
Years
Months
Days
Nb
T= 2 months
Mo
Ce
Cs
Ru
Nd
Sr
Cs
Ba
Tc
Y
Pr
Kr
Rb
Te
Eu
Sm
Pd
Pm
Rh
J
Nb
Eu
Sb
Cd
Gd
Ag
In
Mole percent

Fig. 12.

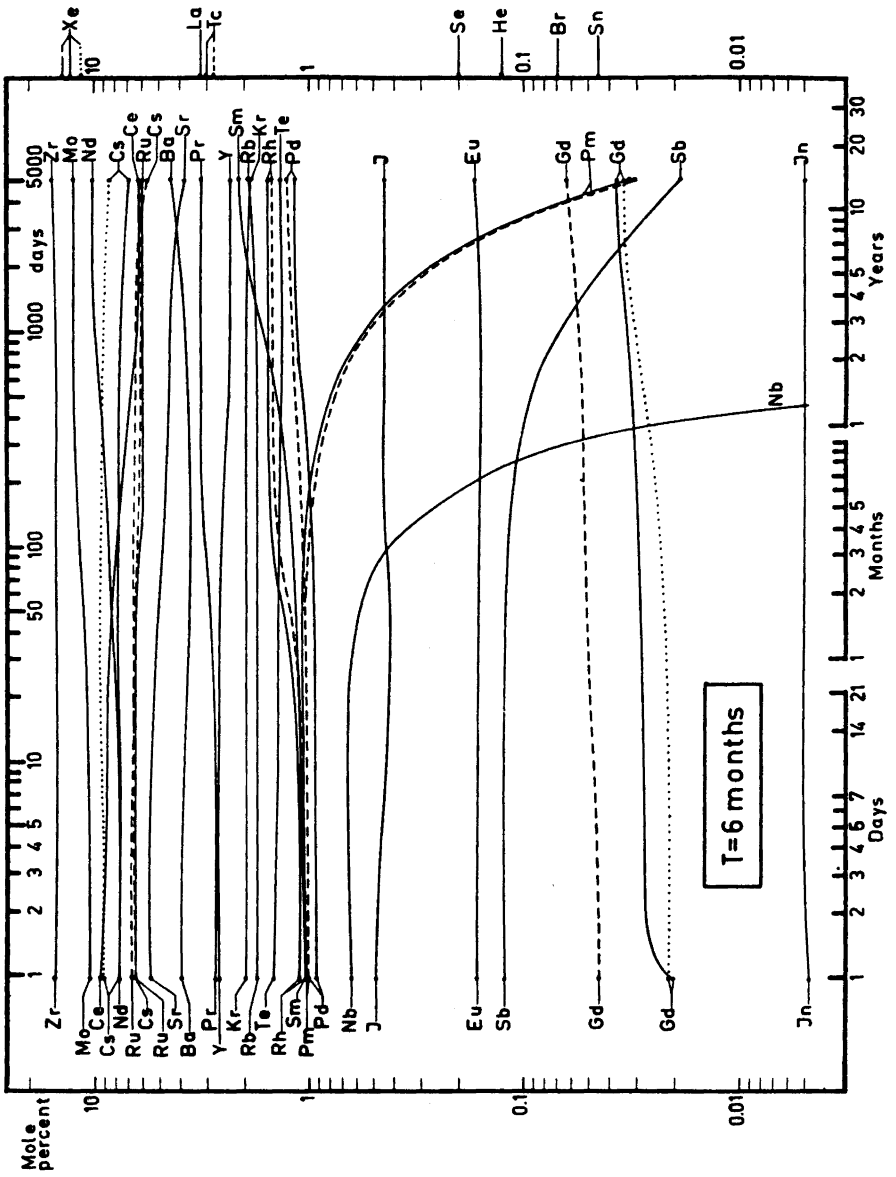
Xe
La
Tc
Se
He
Br
Sn
10
1
0.1
0.01
5000
1000
100
50
10
1
days
Years
Months
Days
Mole percent
T=6 months
Zr
Mo
Nd
Cs
Ce
Ru
Ba
Sr
Pr
Y
Sm
Rb
Kr
Rh
Te
Pd
Nb
J
Eu
Sb
Gd
Pm
In

Fig. 13.

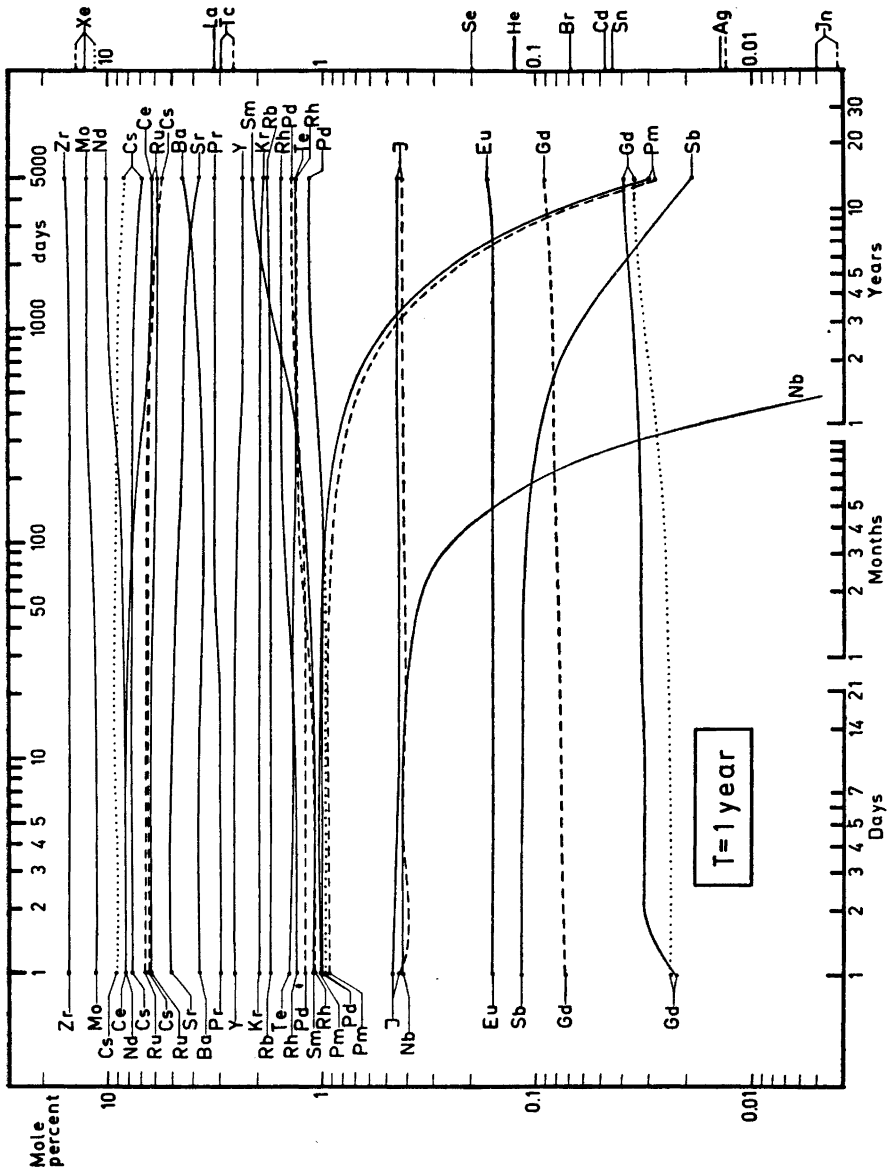
Xe
Ta
Tc
Se
He
Br
Cd
Sn
Ag
In
10
1
0.1
0.01
Zr
Mo
Nd
Cs
Ce
Ru
Ba
Sr
Pr
Y
Sm
Kr
Rb
Rh
Pd
Te
Pm
I
Nb
Eu
Sb
Gd
Gd
Pm
Sb
Nb
T=1 year
Mole percent
10
1
0.1
0.01
1000 days
5000
100
50
10
1
Years
Months
Days
1 2 3 4 5 10 20 30
1 2 3 4 5
1 2 3 4 5 7 14 21

Fig. 14.

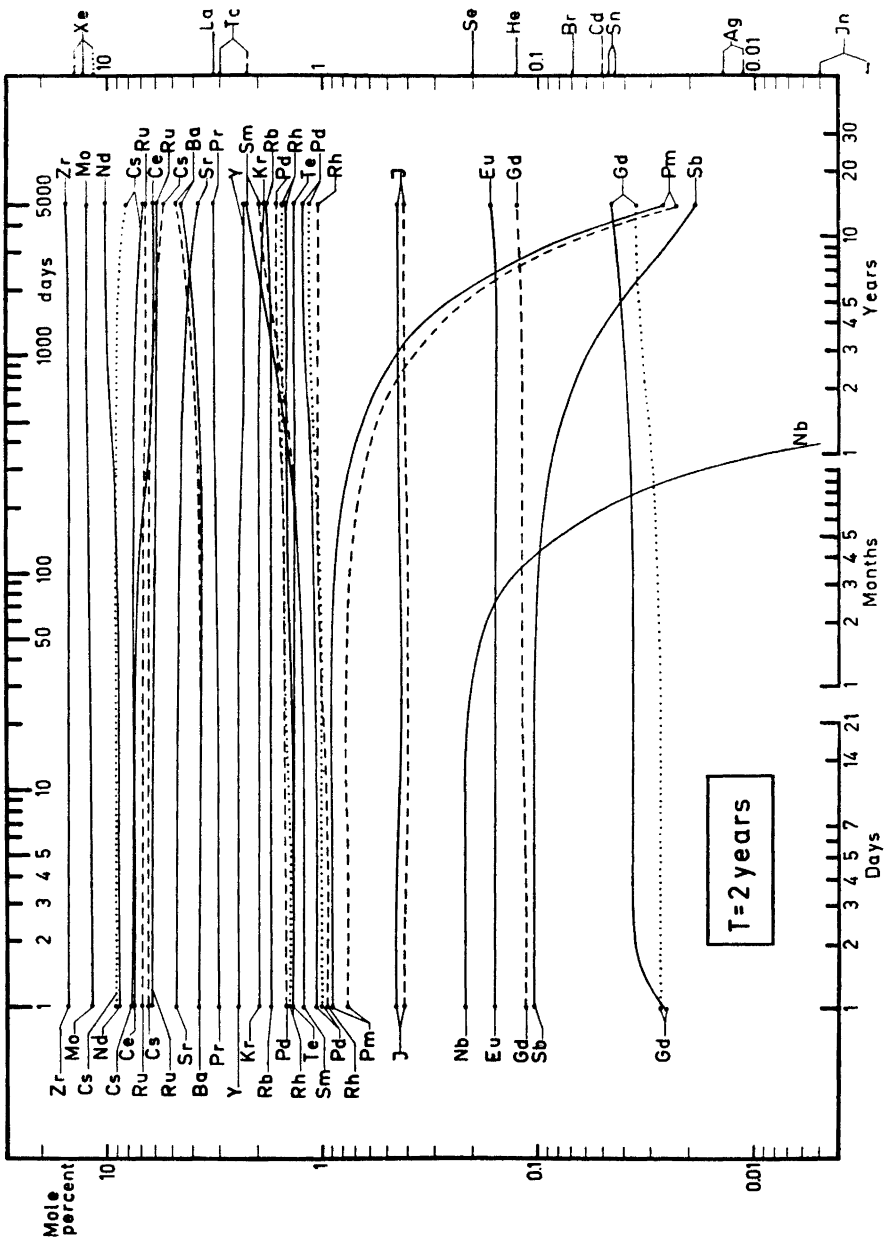
Xe
La
Tc
Se
He
Br
Cd
Sn
Ag
In
10
1
0.1
0.01
Zr
Mo
Nd
Cs
Ce
Ru
Ba
Sr
Pr
Y
Sm
Kr
Rb
Pd
Rh
Te
Tc
I
Eu
Gd
Pm
Sb
Nb
5000
1000
100
50
10
days
Years
Months
Days
T=2 years
Mole percent

Fig. 15.

data. However, an estimation has been made, which shows that for a standard deviation of 3 % in y and 1 % in λ, the deviations in A and X correspond to $\pm$ 0.2 mm in Figs. 1—15. For a standard deviation of 10 % in y and 5 % in λ, the deviations in A and X correspond to $\sim$1 mm for $t \approx \tau$, and to $\sim$2 mm for $t \approx 8\,\tau$. The errors increase for increasing ratio t/τ. However, it can be seen that these errors are of about the same size as the vertical thickness of the lines drawn in the figures.

Another limitation of the diagrams is discussed in the next paragraph.

THE INFLUENCE OF HIGH BURN UP

The calculations resulting in Figs. 1—15 are all based on the fundamental assumption that the amount of ^{235}U is constant during the irradiation time, T, (*i.e.* that the "burn up" of ^{235}U is low). Practically all published calculations of the kind presented in this paper, are based on the same assumption, even if this is not always explicitly stated.

The weight of fission products formed is proportional to $T \cdot \Phi \cdot \sigma_{fiss} \cdot N$, where N is the number of fissile atoms. Since all calculations in this paper refer to a constant weight of fission products (1 g or 1 mole) formed at a constant flux during a constant irradiation time, it follows that N also must be constant. If this is not the case owing to high burn up, either Φ must be increased in order to produce the fixed weight of fission products in the predetermined time T, or the time T must be increased to produce the fission products at a constant flux. In all cases, the conditions of the equations used in the calculations above are violated. The errors due to a burn up of about 10 % of the ^{235}U causes an error of $\lesssim$ 3 % in the activities or mole fractions given in the figures.

A burn up of 10 % is obtained at 1.5×10^{21} n cm^{-2}, *i.e.* at $\Phi = 10^{12}$ n cm^{-2} sec^{-1} after $T \approx 5$ years, at $\Phi = 10^{13}$ after $T \approx 6$ months, and at $\Phi = 10^{14}$ after $T \approx 18$ days. However, all calculations have been continued up to $T = 2$ years for fluxes $\leq 10^{14}$ n cm^{-2} sec^{-1}, even if the practical usefulness of the diagrams for $\Phi \cdot T > 1.5 \times 10^{21}$ n cm^{-2} may be limited to reactors especially designed to keep the ^{235}U amount constant without removing the fission products.

Acknowledgements. The authors wish to thank the head of FOA 1, Prof. G. Ljunggren, for his continuous interest in and support of these studies, Prof. A. Ölander, Mr. I. Österlund and Mr. P. E. Bergner for valuable discussion, and Miss M. Granstrand for drawing the diagrams. The English of this paper has been corrected by Dr. H. S. Dunsmore. Large size copies of the Figures may be obtained on request to the authors.

REFERENCES

1. Prawitz, J. and Rydberg, J. *Acta Chem. Scand.* **12** (1958) 369.
2. Hunter, H. F. and Ballou, N. E. *Nucleonics* **9** No. 5 (1951) C-1.
3. Björnerstedt, R., Löw, K. and Ulvönäs, S. *Swedish Preliminary Report to the United Nations Scientific Committee on Effects of Atomic Radiations*, July 11th 1956; Löw, K. and Björnerstedt, R. *Arkiv Fysik. In press.*
4. Lock, J. C. L. *A.E.R.E. Report C/R 1715*, Harwell 1955.

5. Moteff, J. *General Electric Co Report APEX-134*, USA 1953.
6. Appendix III, *Progress in Nuclear Chemistry, Vol. III. Process Chemistry, Vol. I*, Pergamon Press Ltd, London 1956.
7. Coryell, C. D., Brady, E. L., Ballou, N. E., Burgus, W. H., Campbell, G. W., Engelkemeir, D. W., Glendenin, L. E. and Novey, T. B. *Paper 39* of Ref.[9]
8. Rubinson, W., Metcalf, R. P., Seiler, J. A., Steinberg, E. P. and Winsberg, L. *Paper 40* of Ref.[9]
9. Coryell, C. D. and Sugerman, N. (Ed.) *Radiochemical Studies. The Fission Products*, McGraw-Hill Book Co, New York 1951.
10. Keneshea, Jr., F. J. and Saul, A. M. *Nucleonics* **11** No. 11 (1953) 26.
11. Walton, G. N. *Paper 4, A.E.R.E. Report C/R 1231*, Harwell 1953.
12. Rubinson, W. *J. Chem. Phys.* **17** (1949) 542.
13. Way, K. and Wigner, E. P. *Paper* **43** of Ref.[9]
14. Thornton, J. K. and Houghton, W. J. *J. Appl. Phys.* **24** (1953) 1374.
15. Koontz, R. L. and Jarrett, A. A. *Nucleonics* **12** No. 6 (1954) 26.
16. Tomlinson, R. H. *Private communication.*

Received November 28, 1957.