Eidg. Institut für Reaktorforschung Würenlingen
Schweiz

The possibility of continuous in-core gaseous extraction of volatile fission products in a molten fuel reactor

M. Taube

Würenlingen, Mai 1974

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Abstract

A system of continuous gas purging of a reactor core for removal of volatile fission products from the molten chloride fuel is discussed and calculations based on a computer programme are shown. The results indicate:

a) the precursors of the delayed neutrons nearly all remain

b) iodine-131 in the steady-state core is reduced approx by a factor 1000

c) caesium-137 concentration in the core decreases only by a factor 7, but the volatility is relativ low due to chloride

d) caesium isotopes 135 and 133 can be separated from Cs-137 by a factor of which make easier the Cs-137 management

e) xenon isotopes are extensively extracted

All this results in a significant improvement in the safety of this reactor type in the case of a core accident.

2

1. General Remarks

The aim of this paper is to discuss the possibility of decreasing the concentration of volatile fission products in the steady state core as a counter measure in the event of a core accident.

The design philosophy of a safe reactor is guided by the statements of the following type: (WASH 1250, 1973)

"The measures against the <u>escape</u> of radioactivity from nuclear facilities (nuclear power plants, fuel reprocessing waste disposal facilities, shipping, storage etc.) should use design features inherently favourable to safe operation e.g. by selection of fuel, coolant and core structural materials which will have inherent stability and safe characteristics."

In this paper a molten fuel fast breeder is discussed which could realise both these requirements through: - minimising the hazard of the escape of radioactive by substances by <u>continuous extraction</u> of the most volatile fission products from the molten fuel so that the steady state amounts of these are reduced by one or more orders of magnitude compared to the classical case of solid fuel periodically discharged and reprocessed.

It is well known that in the case of a reactor accident the hazards are centered around the volatile fission products (F.P.). For example according to Farmer (1973) a 1500 MW(th) reactor contains 4×10^9 curies of volatile and gaseous

fission products of which I-131 equals 50 Megacuries. The
hypothetical release having a very low probability of causing
harm to the general public (unlikely to cause one case of
fatal illness within the ensuing ten or more years) is of the
order of 5 kilocuries of I-131. In a hypothetical low pro-
bability accident they discussed a release of 5 Megacuries
of I-131 and 0.5 Megacuries of Cs-137.

2. Continuous gas extraction
────────────────────────────

The continuous in core gas extraction of volatile fission
products from the liquid fuel is based on the following
data and/or roughly estimated calculations (using the appro-
priate "GASEX" programme)

a) the reactor is fuelled with molten salt fuel - in this
    case molten chlorides $PuCl_3$ - UCl_3 - NaCl etc. (fig. 1)

b) the chemical properties of the fission products in
    this molten media are characterised by the scheme
    shown in (fig. 2)

c) the balance of the fission products - especially for
    chlorine is given in (fig. 3)

d) the thermodynamic stability of all the important
    irradiated fuel components are given in fig. 3

e) the volatility (partial pressure) of some selected cru-
    cial fission products: e.g. caesium in the form of ele-
    ment, oxide and chloride is given in fig. 4

Fig. 1   Molten chlorides fast breeder
          ~3 GW(th)

750°C   790°C

V=10m.s⁻¹

Core
~990°C   fuel

2.0 m   ~4 m

V=2m.s⁻¹

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2.36 m

5
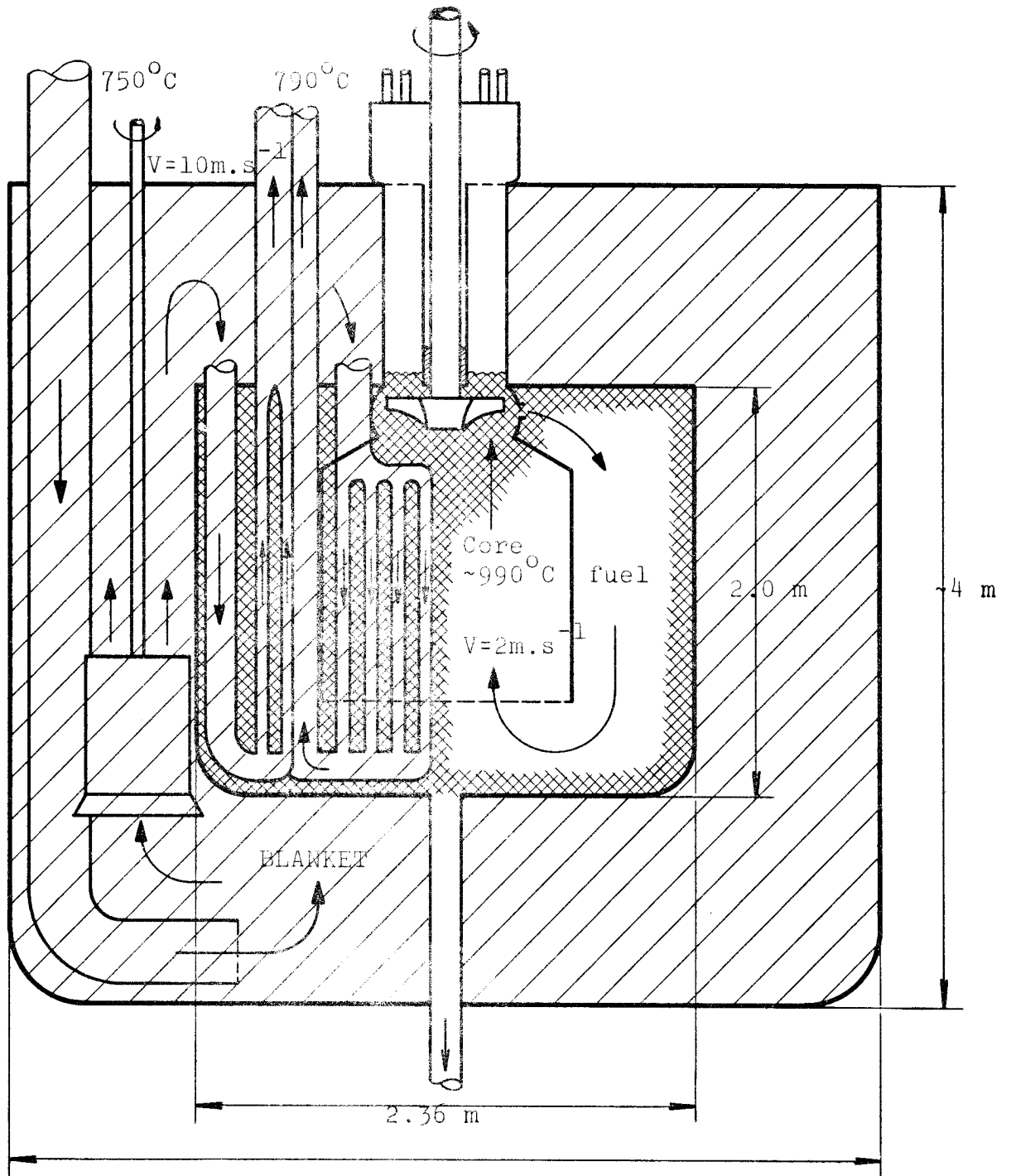
Fig. 2 Fission Products in Molten Chlorides Media

36	Kr	54	Xe
35	Br	53	I
34	Se	52	Te
33	As	51	Sb
32	Ge	50	Sn
31	Ga	49	In
30	Zn	48	Cd
		47	Ag
		46	Pd
		45	Rh
		44	Ru
		43	Tc
		42	Mo
		41	Nb
		40	Zr
		64	Gd
		63	Eu
		62	Sm
		61	Pm
		60	Nd
		59	Pr
		58	Ce
39	Y	57	La
38	Sr	56	Ba
37	Rb	55	Cs

Gas Extraction — fast, slow, very slow

Volatile chlorides

Low volatile chlorides

Non volatile metals

Low volatile chlorides

Non volatile chlorides

Low volatile chlorides

Fig. 3   Free Energy of Formation

Fission products in elementary form

Noble metals
Noble gases

Fission products in form of chloride

ΔG (KJ mol^{-1})

0

-100

-200

-300

— MoCl$_x$

— NbCl$_5$

— AgCl
— SbCl$_3$

— CdCl$_2$

— SnCl$_2$

— InCl

— UCl$_4$

— UCl$_3$ — ZrCl$_3$

— PuCl$_3$

Lanthanides

— YCl$_3$
— NdCl$_3$
— PrCl$_3$
— CeCl$_3$
— LaCl$_3$

— NaCl   — RbCl

— CsCl

— SmCl$_2$

— SrCl$_2$

— BaCl$_2$
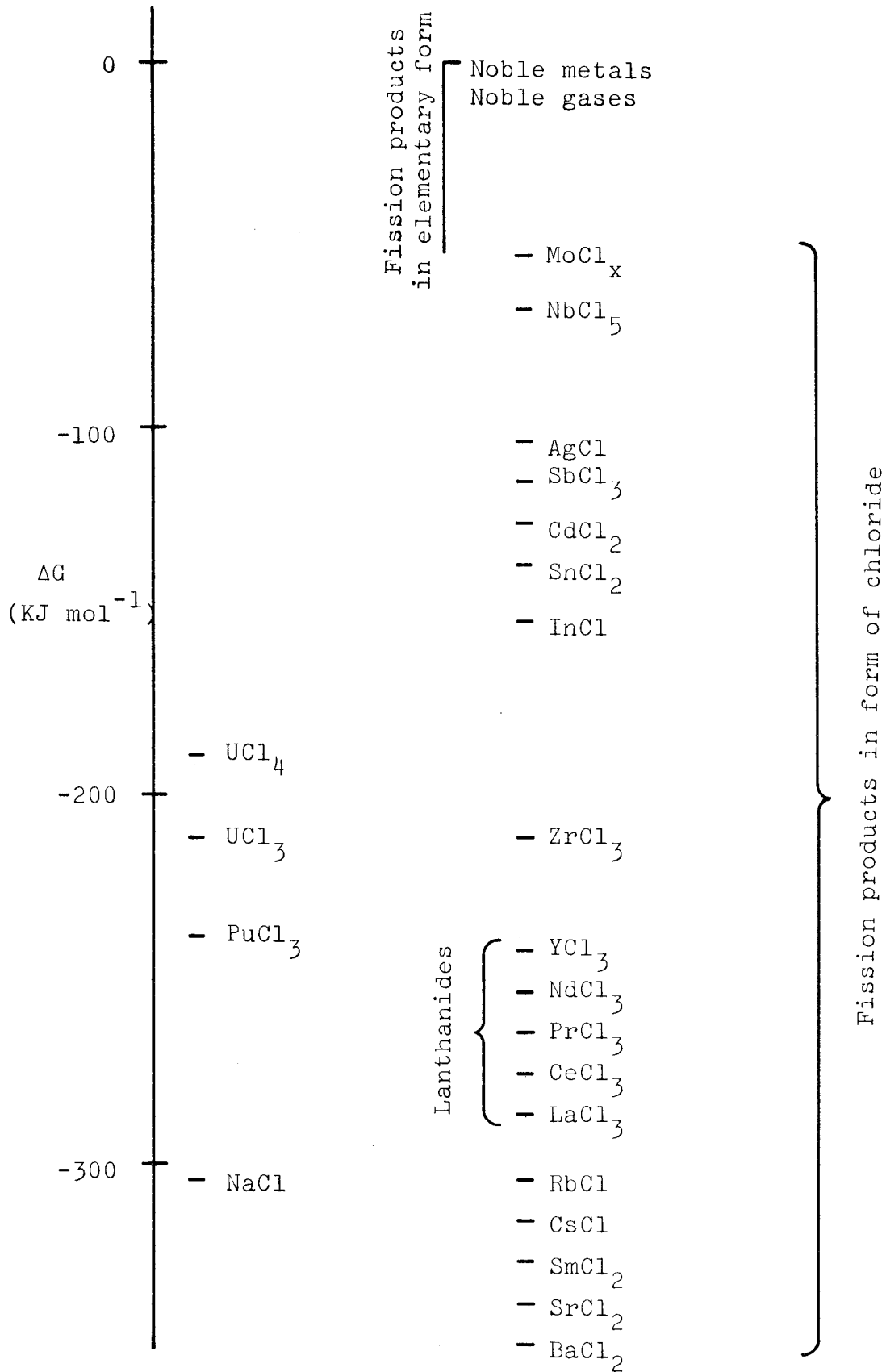
Fig. 4   Volatility Pressure of Caesium-metall (Caesium Oxide)
           and Caesium Chlorides
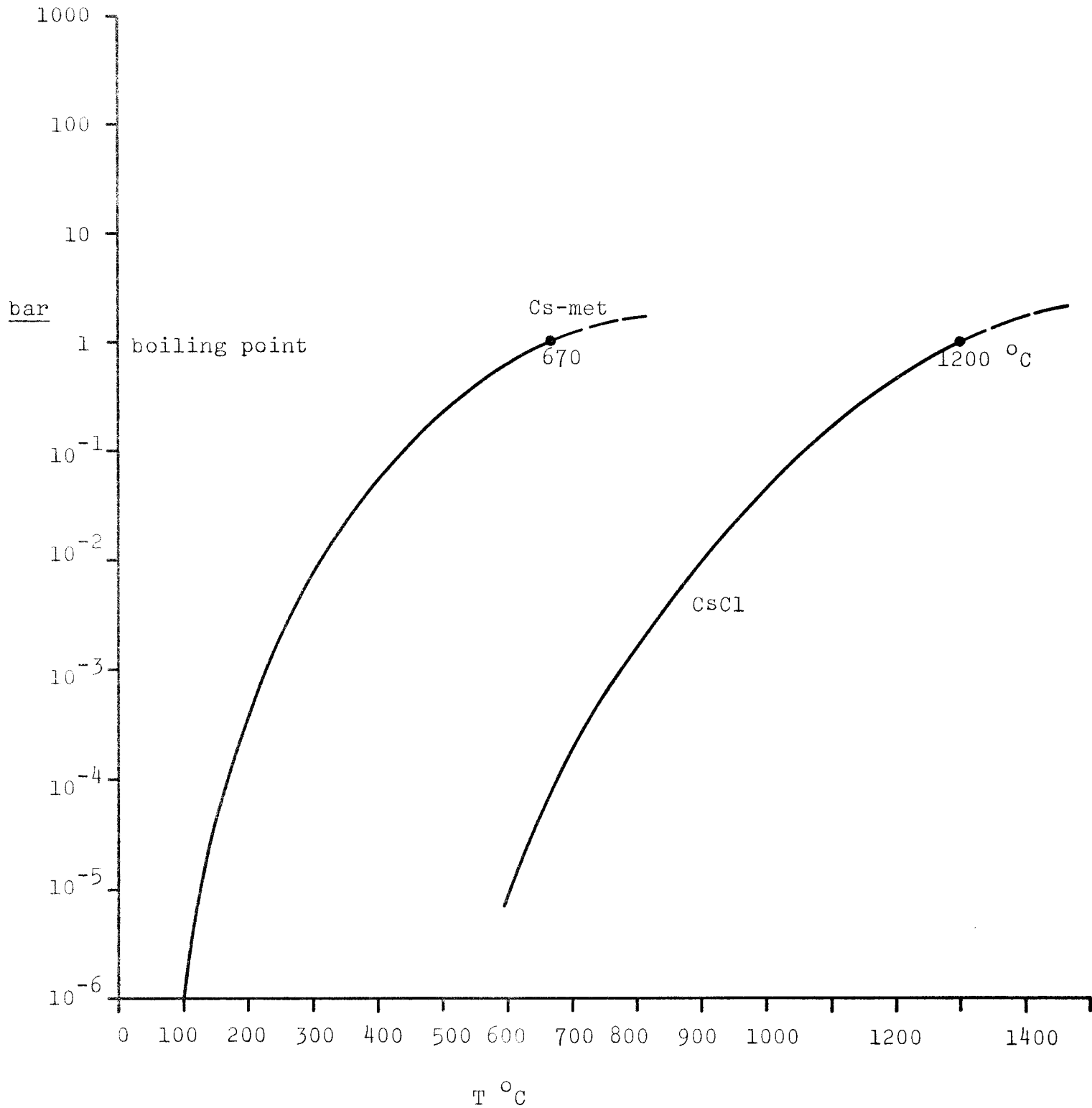
Fig. 5   Scheme of the independent Yield Calculation

A = 137

Extraction rate

51 52 53 54 55 56
Sb Te I   Xe Cs Ba

Fissioned plutonium atom

β^- β^- β^- β^- β^-

100
50
20
10

51.77 %

36.24 %

8.88 %

yield
%
according[1]
(Crouch,
1969)

2.93 %

0,1

0.16 %

0.02 %

10^7 1 year
        100 days

10^6 10 days

time
seconds

        1 day
        10 hour

10^4 1 hour

10^3 10 min.

10^2 1 min.

10

1

51 52 53 54 55 56
Sb Te I   Xe Cs Ba
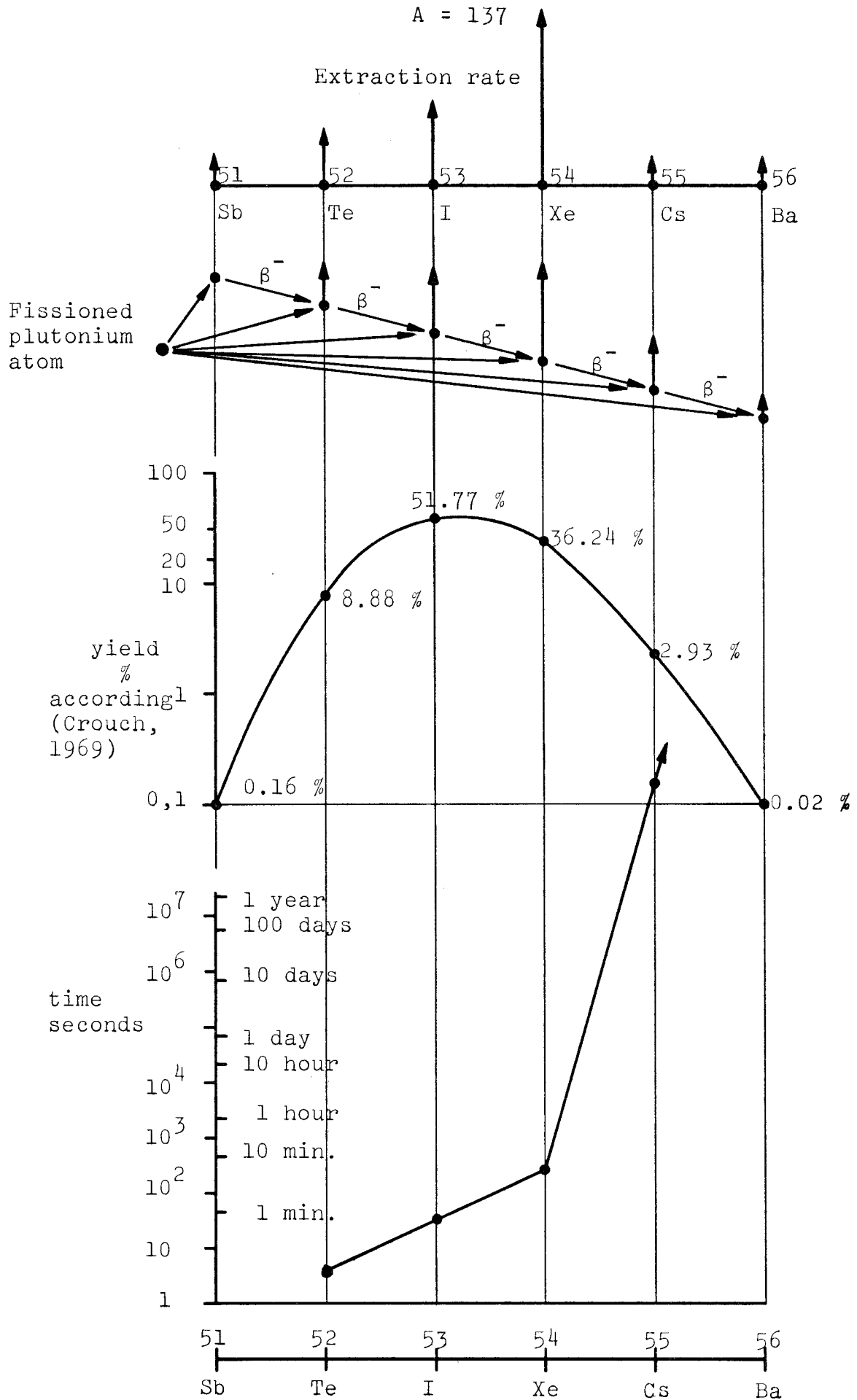
Fig. 6   Scheme of Gas-Extraction
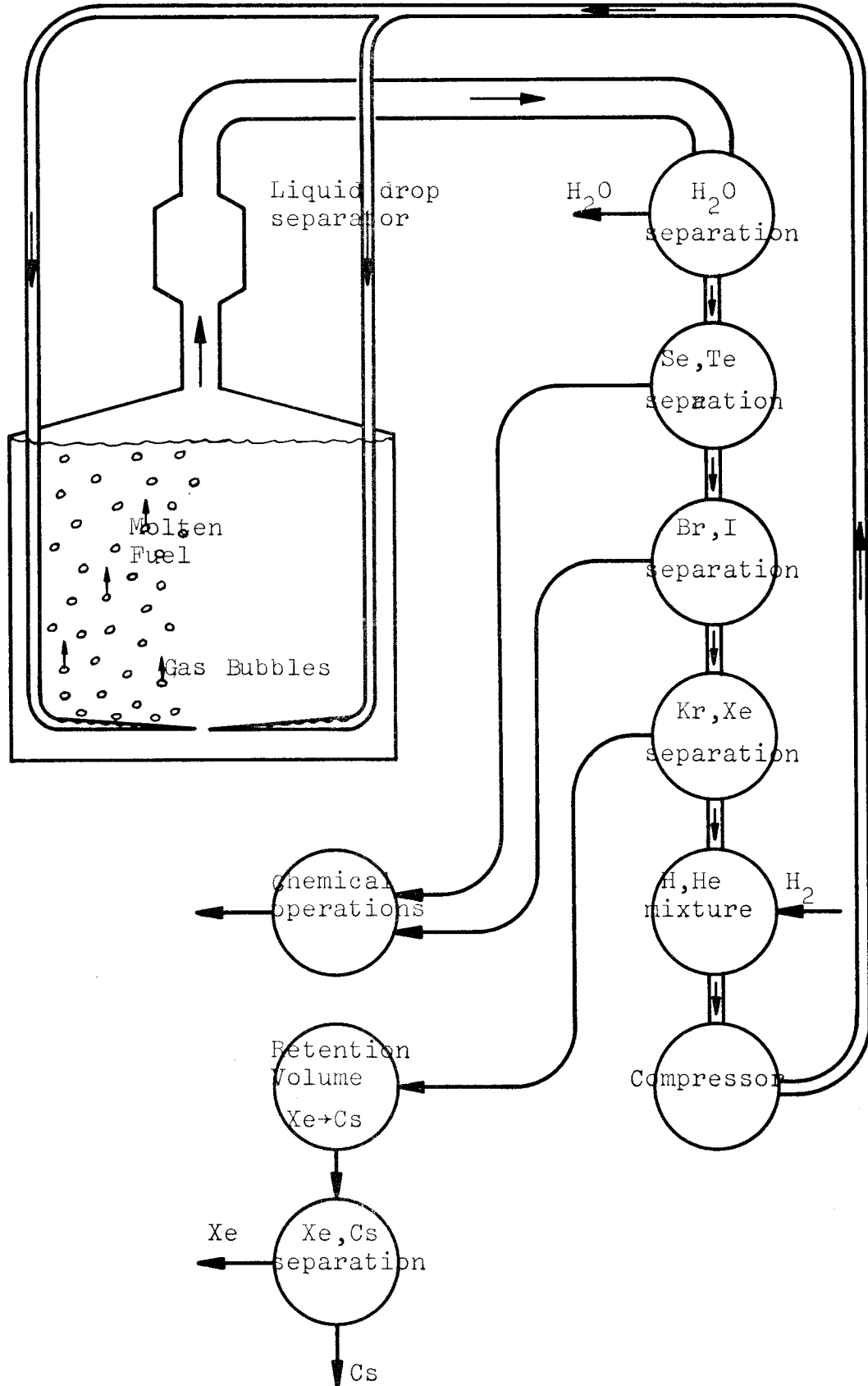
f) the history of each individual fission product is
    calculated on the basis of the independant yields:
    see fig. 5 (according to Crouch 1973)

g) the in-core molten salt medium is purged by means of
    heliumhydrogen gas bubbles which greatly influence the
    chemical behaviour of some fission products (parti-
    cularly iodine). The assumptions concerning the size
    of this stream is given in Appendix 1

3. Scheme of gas extraction and possible technology of
    gas separation

In order to give a basis for discussion on the gas extraction
systems fig. 6 gives a simplified schematic. It must be
stressed that this is a preliminary suggestion only without
any basic studies.

4. Scheme of calculation

For calculation of the gas extraction system refered to here
a computer programme "GASEX" has been prepared (using Fortran
IV for the CDC 6500/6700). The principal layout on which the
calculation is based is given in fig. 7.

Fig. 7   The Scheme of Calculation

Independent yield: η

$\dot{P} = N \cdot \sigma \cdot \emptyset \cdot \eta$

Extraction

out of core

$\dot{E} = e \cdot N$

concentration

$^A_Z N$

$^A_{(Z-1)}$

β^- decay

$\dot{D} = N \cdot \lambda$

β^- decay

$\dot{Z} = N \cdot \lambda$

$^A_{(Z+1)}$

$(N-1)$

$(Al)_Z$

Neutron Irradiation

$\dot{I} = N \cdot \sigma \cdot \emptyset$

Neutron Irradiation

$\dot{Y} = N \cdot \sigma \cdot \emptyset$

$(A+1)$

$^{(A+1)}_Z N$

from fission

Production

(n, γ)

Neutron irradiation

(A)

$^A_{(Z-1)}$ $(N-1)$

β^-

Beta decay

$^A_Z N$

β^-

Beta decay

$^A_{(Z+1)}$ $(N-1)$

Extraction

out of core

(n, γ)

Neutron irradiation

$(A-1)$

$^{(A-1)}_Z (N-1)$

$(Z-1)$ (Z) $(Z+1)$
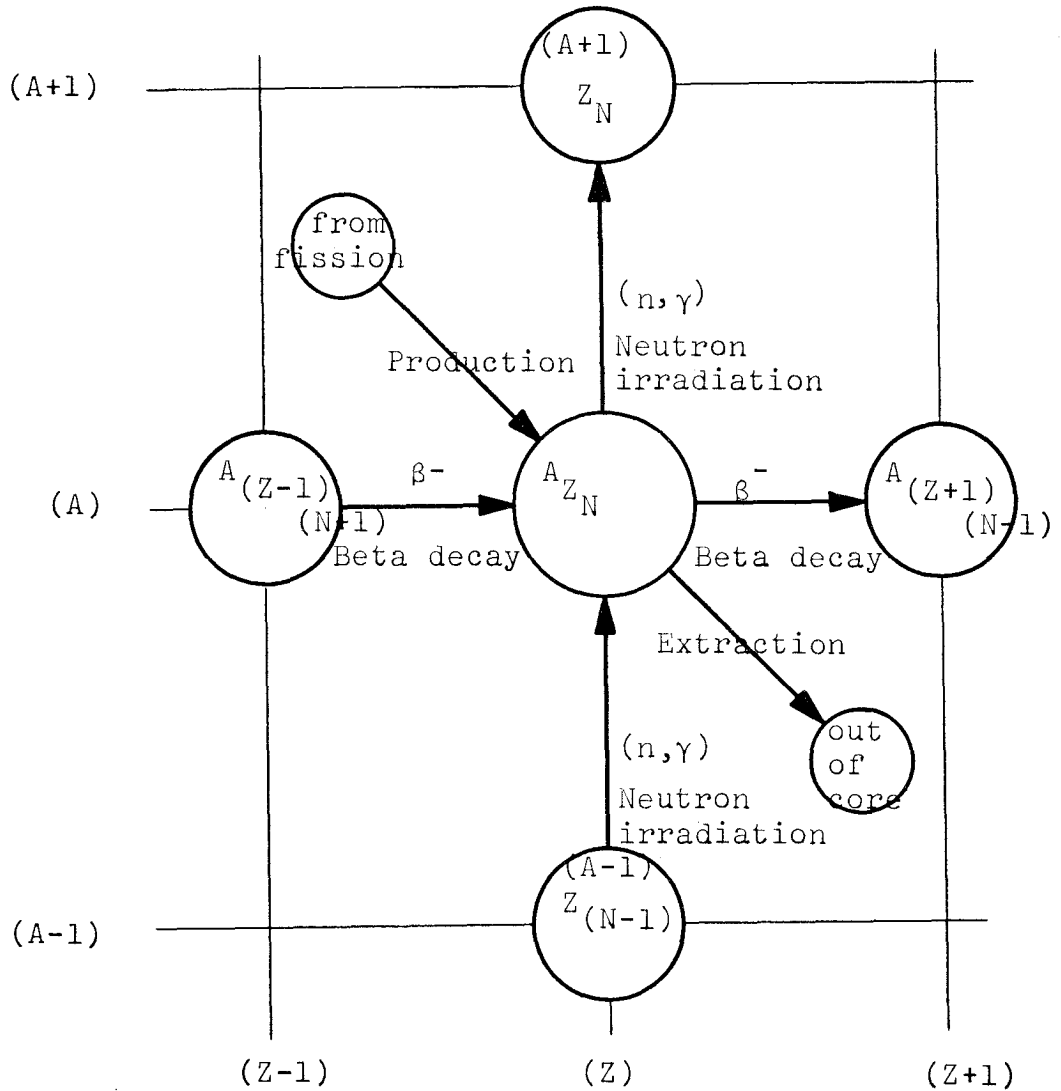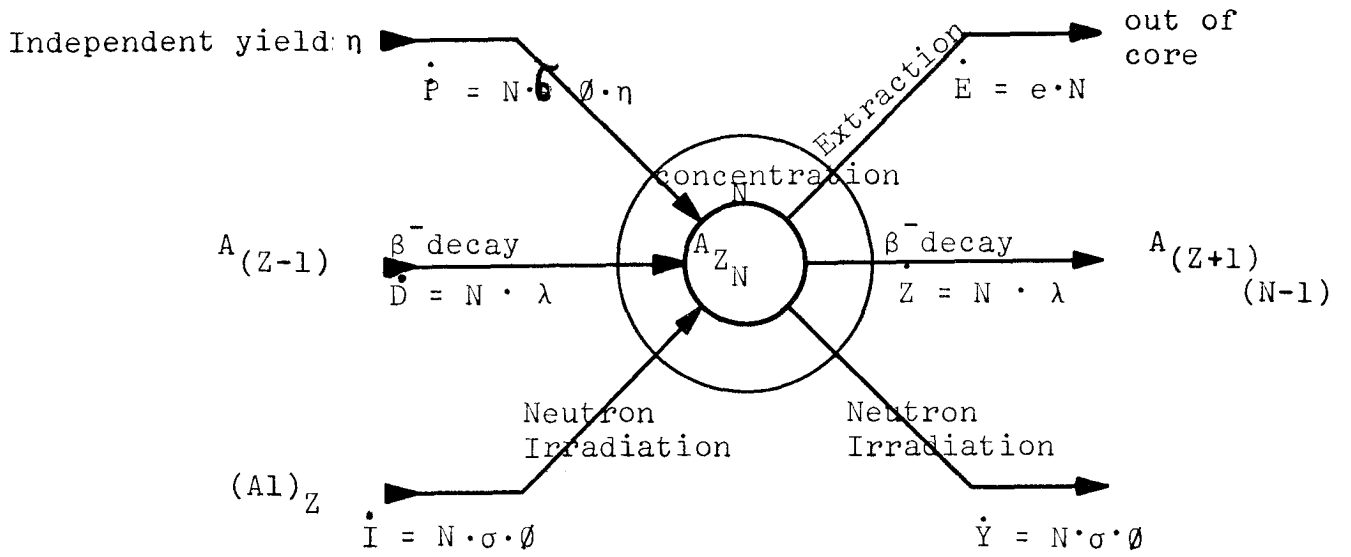
The only assumption arbitrarily made was the gas extraction rate of the volatile fission products.

The calculation was made for the following four assumptions, given in table 1.

Table 1   Core dwell time - (seconds)

	Variant			
	1	2	3	4
Se and Te	10^6	10^9	10^4	10^3
Br and I	10^6	10^4	10^3	10^2
Kr and Xe	10^6	10^3	10^2	10^1
All other elements *)	10^6	10^6	10^6	10^6

*) 10^6 seconds equals 11.57 days which is postulated as the
   reprocessing period of the total liquid fuel

5. Results of the calculations

The most important results are:

1. Stable bromine isotopes A-79 and A-87 (fig. 8) are extrac-
    ted with high efficiency.
    The short lived isotopes (t 1/2 ~ hours or minutes) (fig. 8)
    are extracted in rather small quantities (logarithmic ver-
    tical scale).

2. Short lived bromine isotopes A = 88, 89, 90, being also
   the probable precursors of delayed neutrons are in
   practice not extractable (no loss of delayed neutrons)
   (fig. 9 - linear vertical scale)

3. The very high retention of the heavy bromine isotopes
   in the molten fuel is almost independant of the rate of
   gas extraction. For the highest gas 'extraction' only
   a small extraction of bromine occurs (fig. 9).

4. The main problem for krypton isotopes is the Kr-85
   (fig. 10). The gas extraction is successful for the
   lowest case (2) and results in a reduction of the amount
   of Kr-85 by approx 1000 times.

By increasing the gas extraction rate the amounts of this
isotopes decreases by a factor 10^{-5}. The short lived krypton
isotopes are very efficiently extracted (fig. 11).

5. The gas extraction has only a small effect on the two
   radioactive strontium isotopes (fig. 12). They are de-
   creased only by a factor of 7.

6. The case of the iodine isotopes (fig. 13 and 14) is cri-
   tical for two reasons

   I) the iodine-131 is an important factor in the core acci-
      dent hazard.

The figures 8-17 are made directly by the CDC 6500/6700 prin-
ter as output from GASEX; If * appears then at least two
curves are superimposed.

Fig. 8

ALL ISOTOPES OF ATOMICNUMBER 35

```
GRAPH   1 -  ELEMENT BR  AT.NO. 35   ISOTOPE 79   STABLE 1.00E-02
1.00E-08   1.00E-06
GRAPH   2 -  ELEMENT BR  AT.NO. 35   ISOTOPE 81   STABLE
1.00E-08   1.00E-04
GRAPH   3 -  ELEMENT BR  AT.NO. 35   ISOTOPE 82   HALF-LIFE 3.540E+01 HRS
1.00E-08   1.00E-04
GRAPH   4 -  ELEMENT BR  AT.NO. 35   ISOTOPE 83   HALF-LIFE 2.400E+00 HRS
1.00E-08   1.00E-04
GRAPH   5 -  ELEMENT BR  AT.NO. 35   ISOTOPE 84   HALF-LIFE 3.200E+01 MIN
1.00E-08   1.00E-06
GRAPH   6 -  ELEMENT BR  AT.NO. 35   ISOTOPE 85   HALF-LIFE 3.000E+00 MIN
1.00E-08   1.00E-06
GRAPH   7 -  ELEMENT BR  AT.NO. 35   ISOTOPE 86   HALF-LIFE 5.400E+01 SEC
1.00E-08   1.00E-04
GRAPH   8 -  ELEMENT BR  AT.NO. 35   ISOTOPE 87   HALF-LIFE 5.540E+01 SEC
1.00E-08   1.00E-06
```

ALL ISOTOPES OF ATOMICNUMBER 35

```
     GRAPH   1 -   ELEMENT BR   AT.NO. 35   ISOTOPE   88   HALF-LIFE 1.600E+01 SEC
0.               2.00E-01                4.00E-01              6.00E-01              8.00E-01              1.00E+00
     GRAPH   2 -   ELEMENT BR   AT.NO. 35   ISOTOPE   89   HALF-LIFE 4.500E+00 SEC
0.               2.00E-01                4.00E-01              6.00E-01              8.00E-01              1.00E+00
     GRAPH   3 -   ELEMENT BR   AT.NO. 35   ISOTOPE   90   HALF-LIFE 1.600E+00 SEC
0.               2.00E-01                4.00E-01              6.00E-01              8.00E-01              1.00E+00
```

Fig. 9

ALL ISOTOPES OF ATOMICNUMBER 36

```
   GRAPH   1 -   ELEMENT KR   AT.NO. 36   ISOTOPE   82       STABLE
1.00E-08              1.00F-06                1.00E-04                 1.00E-02              1.00E+00              1.00E+02
   GRAPH   2 -   ELEMENT KR   AT.NO. 36   ISOTOPE   83       STABLE
1.00E-08              1.00F-06                1.00E-04                 1.00E-02              1.00E+00              1.00E+02
   GRAPH   3 -   ELEMENT KR   AT.NO. 36   ISOTOPE   84       STABLE
1.00E-08              1.00E-06                1.00E-04                 1.00E-02              1.00E+00              1.00E+02
   GRAPH   4 -   ELEMENT KR   AT.NO. 36   ISOTOPE   85       HALF-LIFE 1.076E+01 YRS
1.00E-08              1.00E-06                1.00E-04                 1.00E-02              1.00E+00              1.00E+02
   GRAPH   5 -   ELEMENT KR   AT.NO. 36   ISOTOPE   86       STABLE
1.00E-08              1.00F-06                1.00E-04                 1.00E-02              1.00E+00              1.00E+02
   GRAPH   6 -   ELEMENT KR   AT.NO. 36   ISOTOPE   87       HALF-LIFE 7.600E+01 MIN
1.00E-08              1.00F-06                1.00E-04                 1.00E-02              1.00E+00              1.00E+02
   GRAPH   7 -   ELEMENT KR   AT.NO. 36   ISOTOPE   88       HALF-LIFE 2.800E+00 HRS
1.00E-08              1.00E-06                1.00E-04                 1.00E-02              1.00E+00              1.00E+02
   GRAPH   8 -   ELEMENT KR   AT.NO. 36   ISOTOPE   89       HALF-LIFE 3.100E+00 MIN
1.00E-08              1.00E-06                1.00E-04                 1.00E-02              1.00E+00              1.00E+02
```


Fig. 10

16

ALL ISOTOPES OF ATOMICNUMBER 36

```
    GRAPH   1 -   ELEMENT KR   AT.NO. 36   ISOTOPE  90   HALF-LIFE 3.200E+01 SEC
0.                2.00E-01              4.00E-01              6.00E-01            8.00E-01            1.00E+00
    GRAPH   2 -   ELEMENT KR   AT.NO. 36   ISOTOPE  91   HALF-LIFE 8.400E+00 SEC
0.                2.00F-01              4.00E-01              6.00E-01            8.00E-01            1.00E+00
    GRAPH   3 -   ELEMENT KR   AT.NO. 36   ISOTOPE  92   HALF-LIFE 1.900E+00 SEC
0.                2.00E-01              4.00E-01              6.00E-01            8.00E-01            1.00E+00
    GRAPH   4 -   ELEMENT KR   AT.NO. 36   ISOTOPE  93   HALF-LIFE 1.200E+00 SEC
0.                2.00E-01              4.00E-01              6.00E-01            8.00E-01            1.00E+00
    GRAPH   5 -   ELEMENT KR   AT.NO. 36   ISOTOPE  94   HALF-LIFE 1.000E+00 SEC
0.                2.00E-01              4.00E-01              6.00E-01            8.00E-01            1.00E+00
    GRAPH   6 -   ELEMENT KR   AT.NO. 36   ISOTCPE  95   HALF-LIFE 8.000E-01 SEC
0.                2.00E-01              4.00E-01              6.00E-01            8.00E-01            1.00E+00
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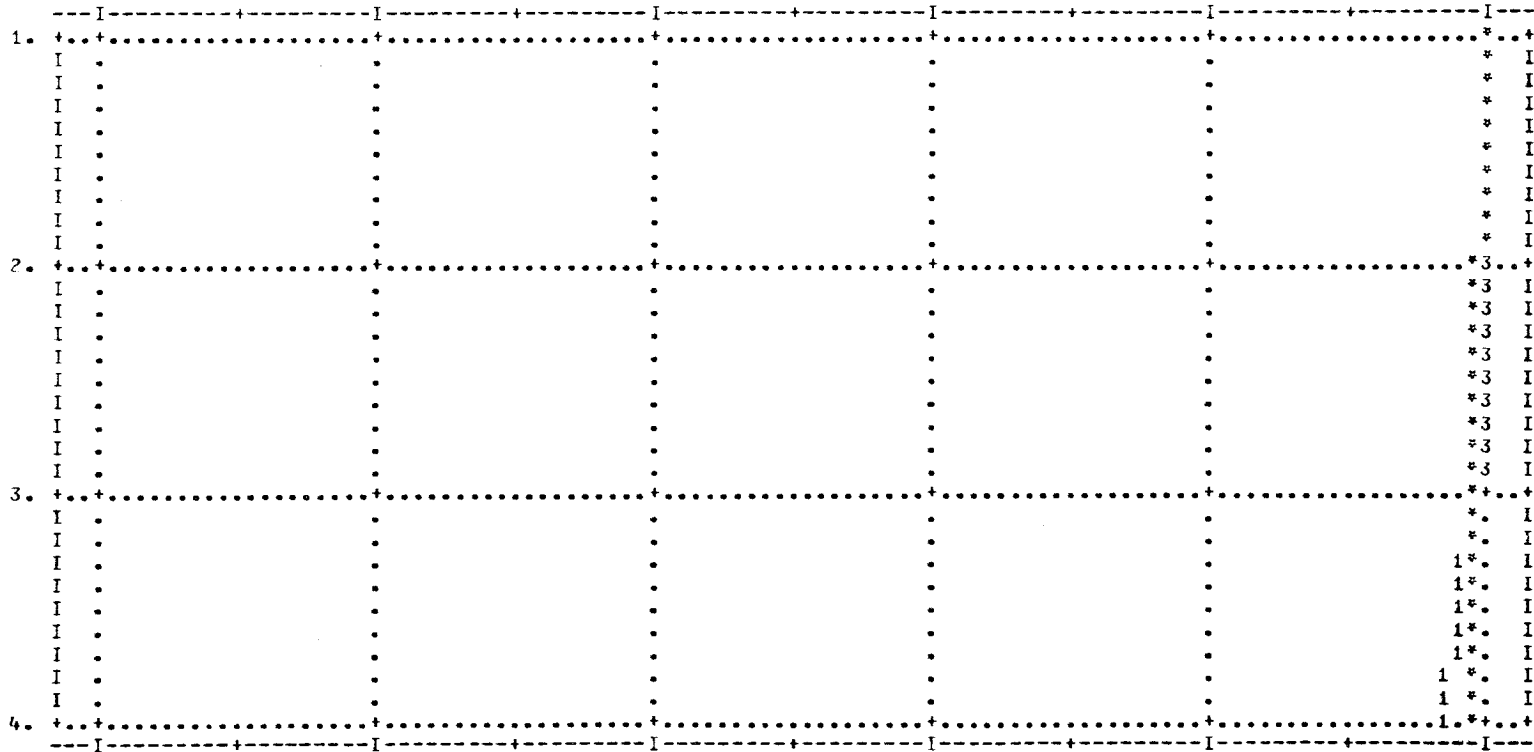
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   I  .                 .                 .                 .                 .                             1 *  I
   I  .                 .                 .                 .                 .                             1 *  I
   I  .                 .                 .                 .                 .                            1  *  I
   I  .                 .                 .                 .                 .                            1  *  I
   I  .                 .                 .                 .                 .                            1   *  I
   I  .                 .                 .                 .                 .                            1   *  I
   I  .                 .                 .                 .                 .                            1   *  I
   I  .                 .                 .                 .                 .                           1    *  I
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   I  .                 .                 .                 .                 .          111       2     *.  I
   I  .                 .                 .                 .                 .       111       2      3*.  I
   I  .                 .                 .                 .                 . 11         2      3*.  I
   I  .                 .                 .                 .              111           2      **.  I
   I  .                 .                 .                 .          111 .             2      **.  I
   I  .                 .                 .                 .       111           .       2       34*.  I
   I  .                 .                 .                 .      11             .       2       34*.  I
   I  .                 .                 .                 .    111          .        2       34*.  I
3. +..+.........................................+..111111..........+...222222.....3***.+..+
   I  .                 .                 .       11111        2222           334*6   . I
   I  .                 .                 .     11111 .       22222.        33 4*6   . I
   I  .                 .                 .   11111          22222   .       33  456    . I
   I  .                 .              11111          2222          .     33  4*56   . I
   I  .                 .            11111 .        22222           .    3   4*56   . I
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   I  .              111111          22222         .        33    44556   . I
   I  .            111111 .        2222            .       33.    4 5 6    . I
4. +..+.................1..+.....................+.........2...............+.........3.+....4.5.6...........+..+
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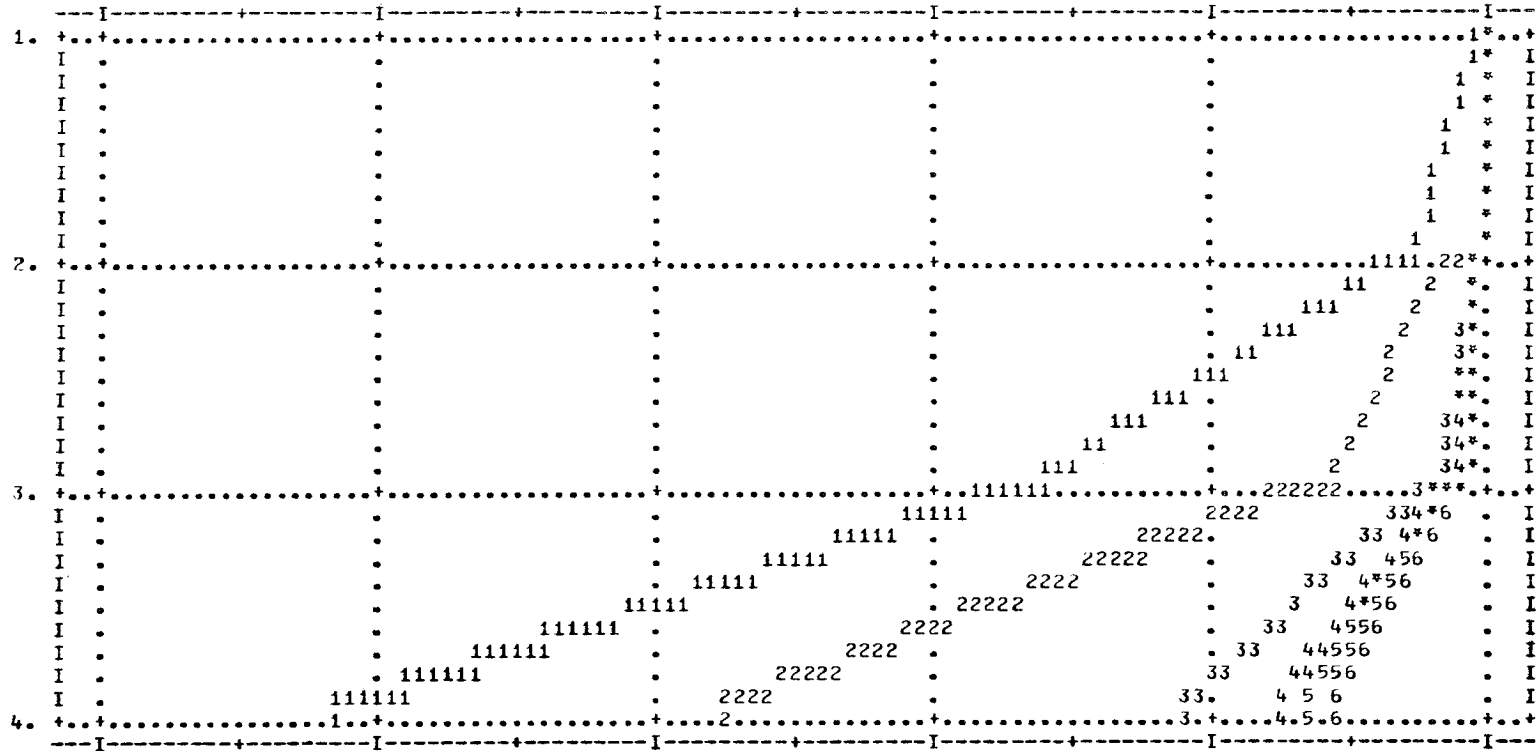ALL ISOTOPES OF ATOMICNUMBER 38

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1.00E-08          1.00E-06          1.00E-04          1.00E-02          1.00E+00          1.00E+02
   GRAPH  2 -  ELEMENT SR  AT.NO. 38  ISOTOPE  89  HALF-LIFE 5.050E+01 DAY
1.00E-08          1.00E-06          1.00E-04          1.00E-02          1.00E+00          1.00E+02
   GRAPH  3 -  ELEMENT SR  AT.NO. 38  ISOTOPE  90  HALF-LIFE 2.810E+01 YRS
1.00E-08          1.00E-06          1.00E-04          1.00E-02          1.00E+00          1.00E+02

      ---I---------+---------I---------+---------I---------+---------I---------+---------I---------+---------I---
  1. +..+...........................+....................+....................+....................1*+....................+..+
     I  .                        .                   .                   .              11* .              . I
     I  .                        .                   .                   .              11  ⋇ .            . I
     I  .                        .                   .                   .           11  32 .              . I
     I  .                        .                   .                   .          11   32 .              . I
     I  .                        .                   .                   .        11     ⋇ .               . I
     I  .                        .                   .                   .       1       32 .               . I
     I  .                        .                   .                   .     11       32 .               . I
     I  .                        .                   .                   .   11       32 .                . I
     I  .                        .                   .                   .11        32 .                  . I
  2. +..+...........................+....................+....................+1...........32......+....................+..+
     I  .                        .                   .                   .1          32 .                . I
     I  .                        .                   .                   .1          32 .                . I
     I  .                        .                   .                   .1          32 .                . I
     I  .                        .                   .                   .1          32 .                . I
     I  .                        .                   .                   .1          32 .                . I
     I  .                        .                   .                   .1          32 .                . I
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     I  .                        .                   .                   .1          32 .                . I
     I  .                        .                   .                   .1          32 .                . I
  3. +..+...........................+....................+....................+1.........32......+....................+..+
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     I  .                        .                   .                   .1          32 .                . I
     I  .                        .                   .                   .1          32 .                . I
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     I  .                        .                   .                   .1          32 .                . I
     I  .                        .                   .                   .1          32 .                . I
     I  .                        .                   .                   .1          32 .                . I
  4. +..+...........................+....................+....................+1.........32......+....................+..+
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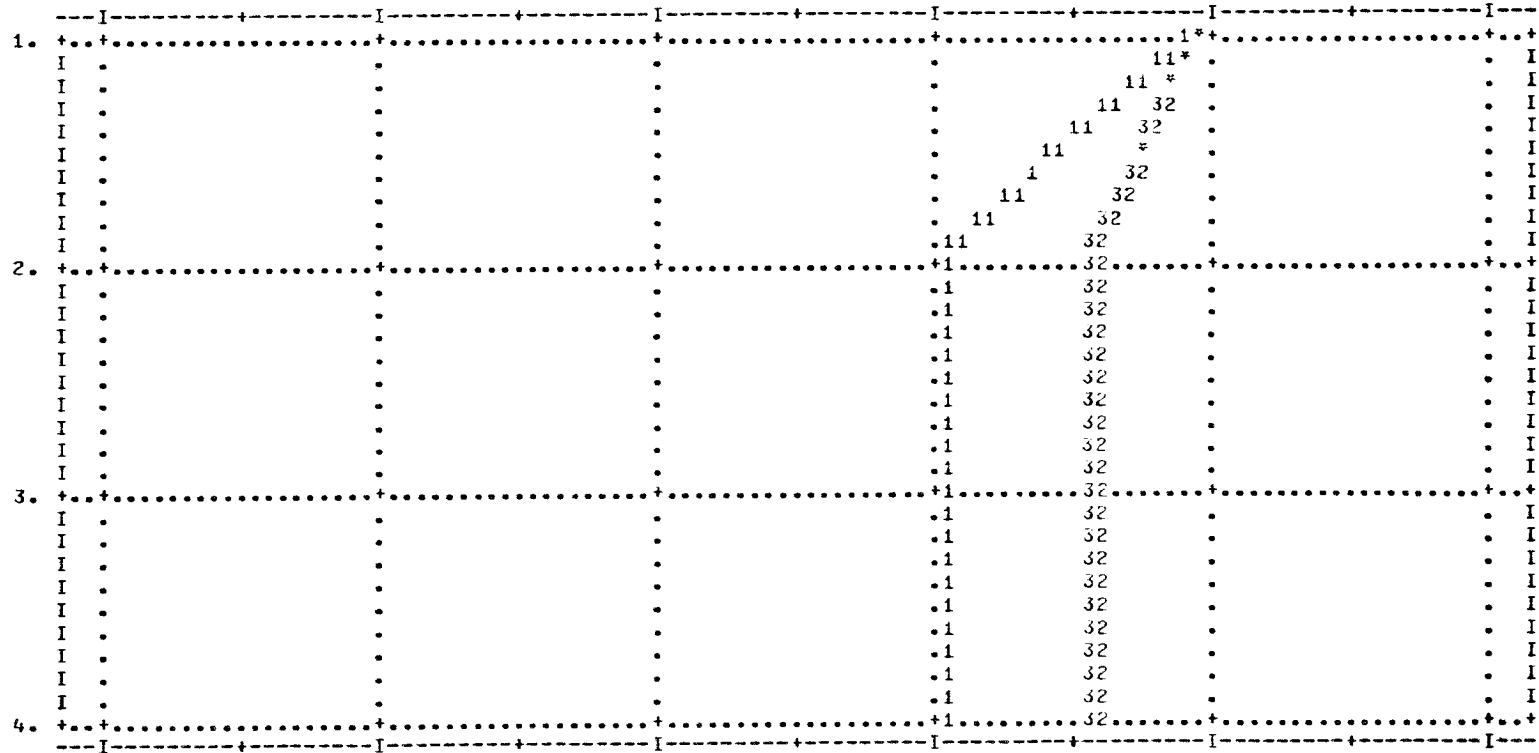
Fig. 12

18

ALL ISOTOPES OF ATOMICNUMBER 53

```
   GRAPH   1 -   ELEMENT   J   AT.NO. 53   ISOTOPE 127     STABLE
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH   2 -   ELEMENT   J   AT.NO. 53   ISOTOPE 129   HALF-LIFE 1.700E+07 YRS
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH   3 -   ELEMENT   J   AT.NO. 53   ISOTOPE 130   HALF-LIFE 1.239E+01 HRS
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH   4 -   ELEMENT   J   AT.NO. 53   ISOTOPE 131   HALF-LIFE 8.050E+00 DAY
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH   5 -   ELEMENT   J   AT.NO. 53   ISOTOPE 132   HALF-LIFE 2.400E+00 HRS
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH   6 -   ELEMENT   J   AT.NO. 53   ISOTOPE 133   HALF-LIFE 2.080E+01 HRS
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH   7 -   ELEMENT   J   AT.NO. 53   ISOTOPE 134   HALF-LIFE 5.200E+01 MIN
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH   8 -   ELEMENT   J   AT.NO. 53   ISOTOPE 135   HALF-LIFE 6.700E+00 HRS
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
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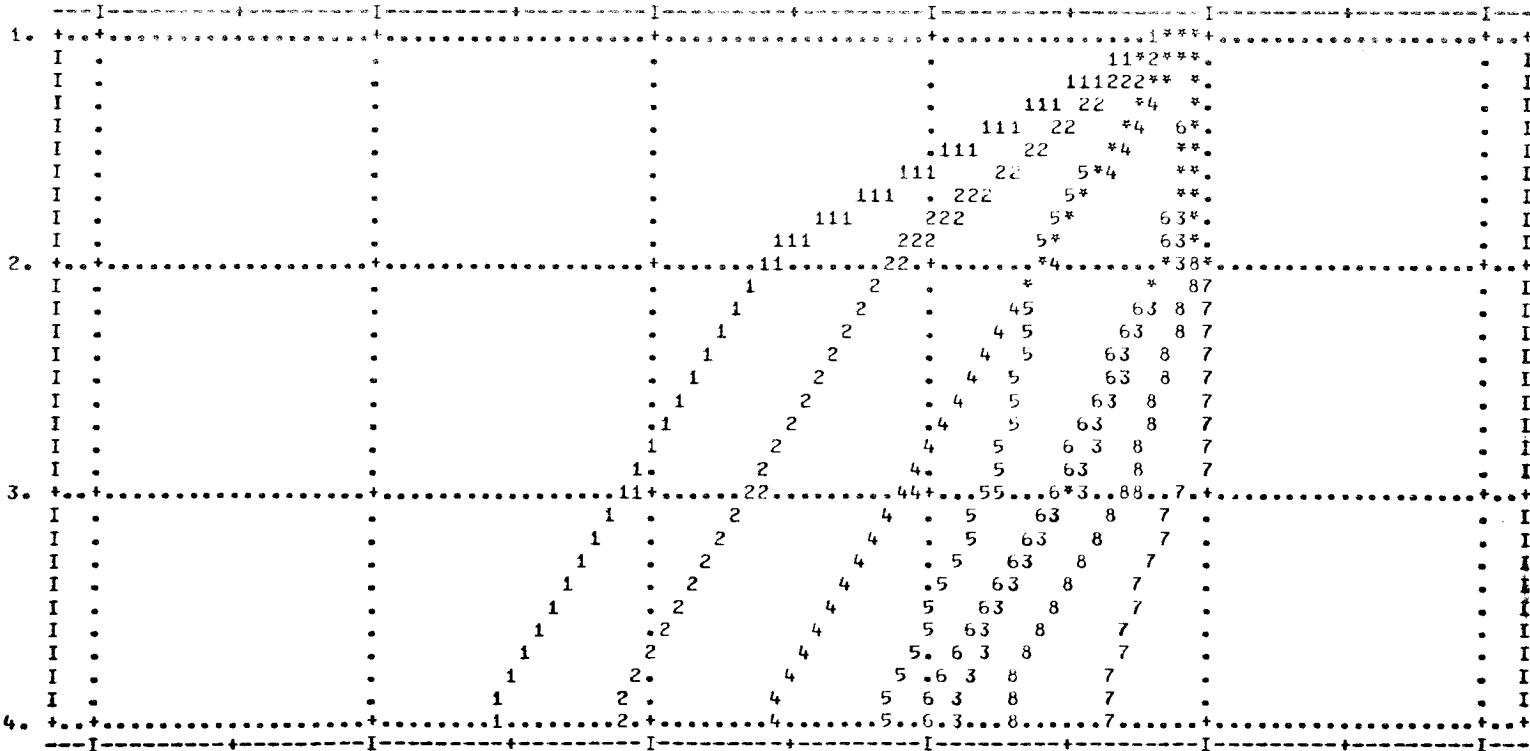

Fig. 13

ALL ISOTOPES OF ATOMICNUMBER 53

```
    GRAPH   1 -   ELEMENT   J   AT.NO.  53    ISOTOPE 136    HALF-LIFE 8.300E+01 SEC
0.               2.00E-01                 4.00E-01                6.00E-01           8.00E-01            1.00E+00
    GRAPH   2 -   ELEMENT   J   AT.NO.  53    ISOTOPE 137    HALF-LIFE 2.400E+01 SEC
0.               2.00E-01                 4.00E-01                6.00E-01           8.00E-01            1.00E+00
    GRAPH   3 -   ELEMENT   J   AT.NO.  53    ISOTOPE 138    HALF-LIFE 6.000E+00 SEC
0.               2.00F-01                 4.00E-01                6.00E-01           8.00E-01            1.00E+00
    GRAPH   4 -   ELEMENT   J   AT.NO.  53    ISOTOPE 139    HALF-LIFE 2.700E+00 SEC
0.               2.00E-01                 4.00E-01                6.00E-01           8.90E-01            1.00E+00
    GRAPH   5 -   ELEMENT   J   AT.NO.  53    ISOTOPE 140    HALF-LIFE 8.000E-01 SEC
0.               2.00E-01                 4.00E-01                6.00E-01           8.00E-01            1.00E+00
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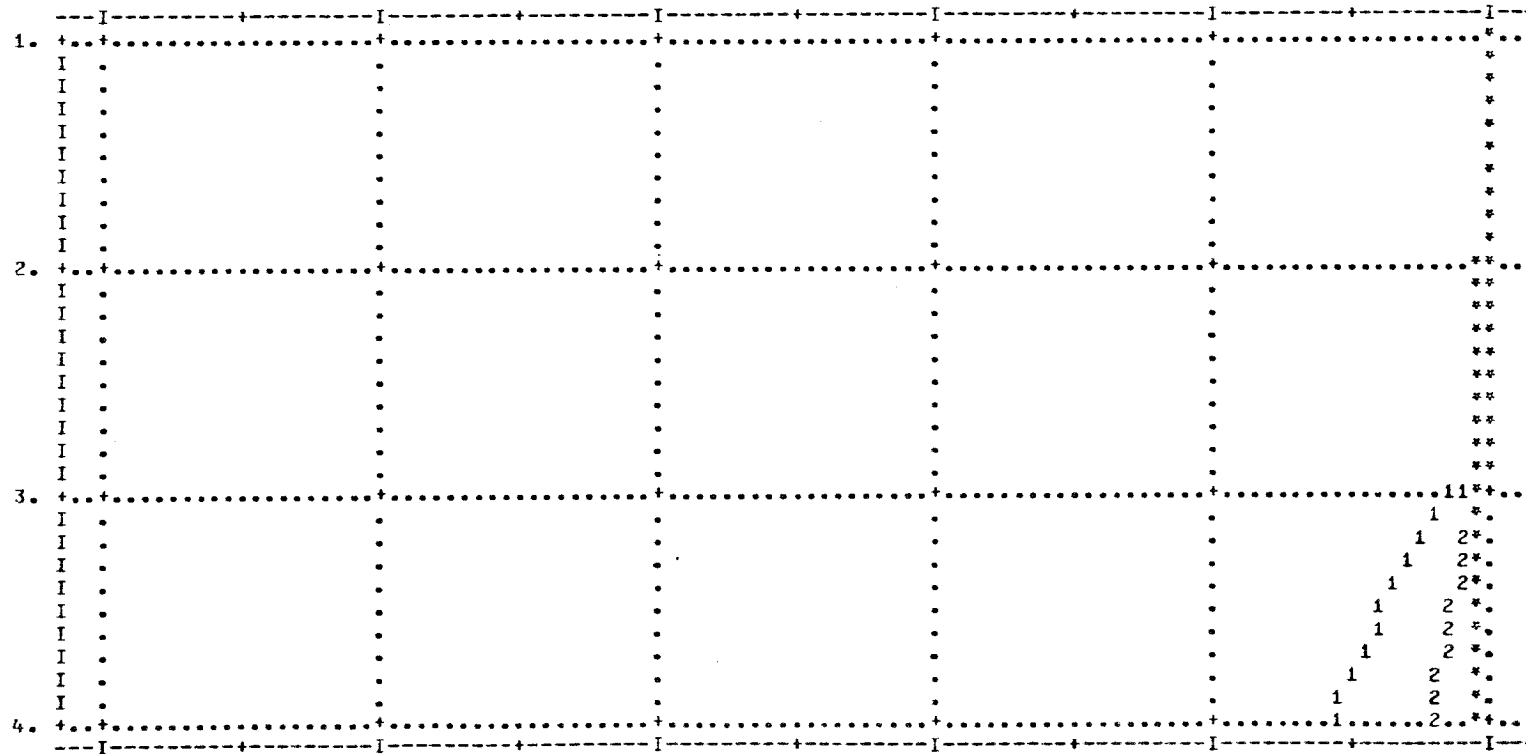
ALL ISOTOPES OF ATOMICNUMBER 54

```
   GRAPH  1 -  ELEMENT XE  AT.NO. 54   ISOTOPE 129    STABLE
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH  2 -  ELEMENT XE  AT.NO. 54   ISOTOPE 130    STABLE
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH  3 -  ELEMENT XE  AT.NO. 54   ISOTOPE 131    STABLE
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH  4 -  ELEMENT XE  AT.NO. 54   ISOTOPE 132    STABLE
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH  5 -  ELEMENT XE  AT.NO. 54   ISOTOPE 133  HALF-LIFE 5.650E+00 DAY
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH  6 -  ELEMENT XE  AT.NO. 54   ISOTOPE 134    STABLE
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH  7 -  ELEMENT XE  AT.NO. 54   ISOTOPE 135  HALF-LIFE 9.150E+00 HRS
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
   GRAPH  8 -  ELEMENT XE  AT.NO. 54   ISOTOPE 136    STABLE
1.00E-08           1.00E-06           1.00E-04           1.00E-02           1.00E+00           1.00E+02
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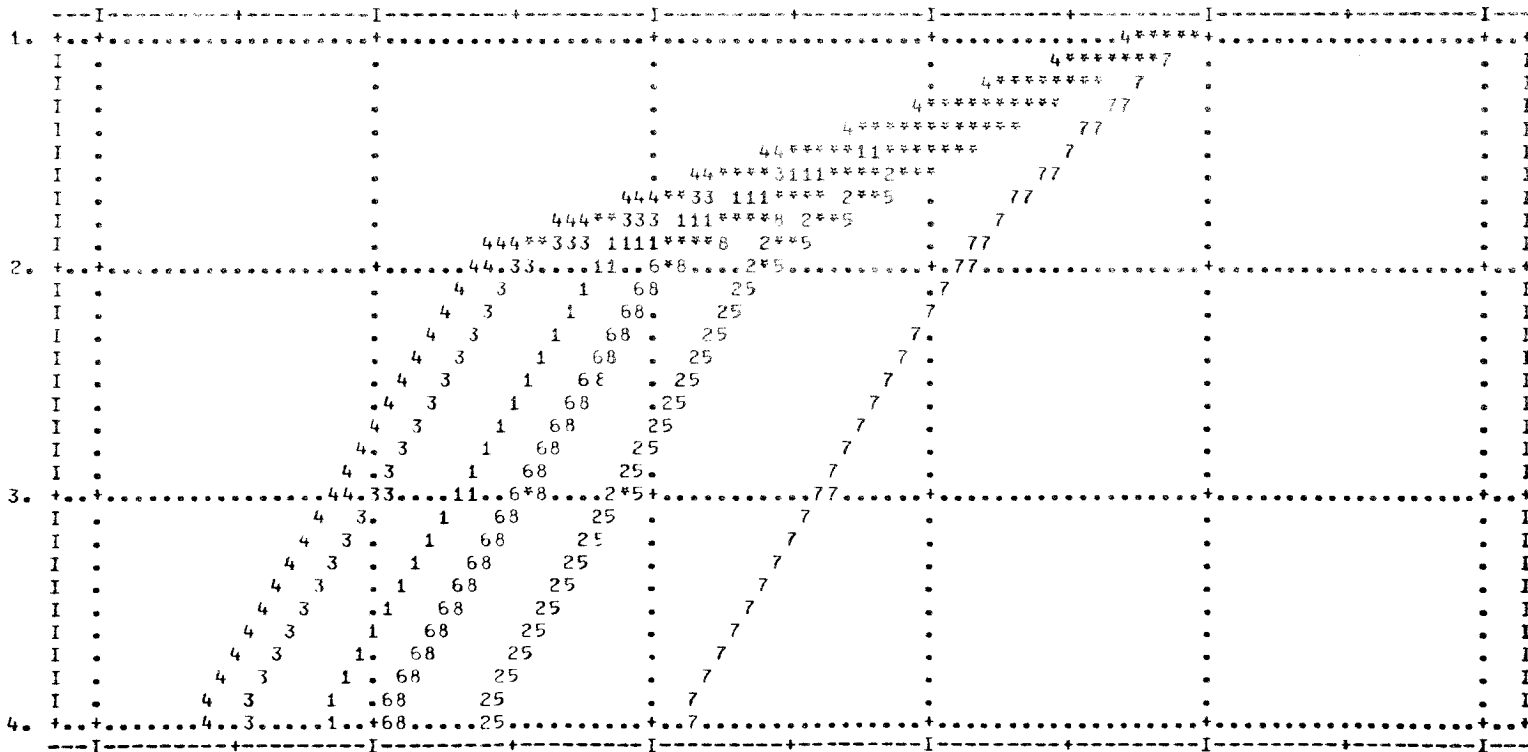


```
ELEMENT XE WITH AT.NO. 54 - ISOTOPE  144   HALF-LIFE 1.000E+00 SEC   HAS  RM = INDEF.
```

Fig. 15

21

ALL ISOTOPES OF ATOMICNUMBER 54

```
     GRAPH   1 -   ELEMENT XE   AT.NO. 54    ISOTOPE 137   HALF-LIFE 3.900E+00 MIN
1.00E-08              1.00E-06             1.00E-04            1.00E-02         1.00E+00           1.00E+02
     GRAPH   2 -   ELEMENT XE   AT.NO. 54    ISOTOPE 138   HALF-LIFE 1.410E+01 MIN
1.00E-08              1.00E-06             1.00E-04            1.00E-02         1.00E+00           1.00E+02
     GRAPH   3 -   ELEMENT XE   AT.NO. 54    ISOTOPE 139   HALF-LIFE 4.100E+01 SEC
1.00E-08              1.00E-06             1.00E-04            1.00E-02         1.00E+00           1.00E+02
     GRAPH   4 -   ELEMENT XE   AT.NO. 54    ISOTOPE 140   HALF-LIFE 1.350E+01 SEC
1.00E-08              1.00E-06             1.00E-04            1.00E-02         1.00E+00           1.00E+02
     GRAPH   5 -   ELEMENT XE   AT.NO. 54    ISOTOPE 141   HALF-LIFE 1.700E+00 SEC
1.00E-08              1.00E-06             1.00E-04            1.00E-02         1.00E+00           1.00E+02
     GRAPH   6 -   ELEMENT XE   AT.NO. 54    ISOTCPE 142   HALF-LIFE 1.200E+00 SEC
1.00E-08              1.00E-06             1.00E-04            1.00E-02         1.00E+00           1.00E+02
     GRAPH   7 -   ELEMENT XE   AT.NO. 54    ISOTOPE 143   HALF-LIFE 1.000E+00 SEC
1.00E-08              1.00E-06             1.00E-04            1.00E-02         1.00E+00           1.00E+02
```


Fig. 16

22

ALL ISOTOPES OF ATOMICNUMBER 55

Fig. 17

```
   GRAPH   1 -   ELEMENT CS   AT.NO. 55   ISOTOPE 133      STABLE
1.00E-08              1.00E-06             1.00E-04             1.00E-02            1.00E+00           1.00E+02
   GRAPH   2 -   ELEMENT CS   AT.NO. 55   ISOTOPE 135   HALF-LIFE 2.000E+06 YRS
1.00E-08              1.00E-06             1.00E-04             1.00E-02            1.00E+00           1.00E+02
   GRAPH   3 -   ELEMENT CS   AT.NO. 55   ISOTOPE 137   HALF-LIFE 3.000E+01 YRS
1.00E-08              1.00E-06             1.00E-04             1.00E-02            1.00E+00           1.00E+02
   GRAPH   4 -   ELEMENT CS   AT.NO. 55   ISOTOPE 138   HALF-LIFE 3.230E+01 MIN
1.00E-08              1.00E-06             1.00E-04             1.00E-02            1.00E+00           1.00E+02
   GRAPH   5 -   ELEMENT CS   AT.NO. 55   ISOTOPE 139   HALF-LIFE 9.000E+00 MIN
1.00E-08              1.00E-06             1.00E-04             1.00E-02            1.00E+00           1.00E+02

      ---I---------+---------I---------+---------I---------+---------I---------+---------I---------+--------I---
   1. +..+.....................+.................+...................+..................1***+..................+..+
      I .                      .                 .                  .          111*2 3*.                     . I
      I .                      .                 .                  .        1111222  3 *.                    . I
      I .                      .                 .                  .     1111  22      3  *.                 . I
      I .                      .                 .                  1111    222       3  45.                  . I
      I .                      .                 .              1111.    22         3  45.                    . I
      I .                      .                 .          1111   . 222        3    45.                      . I
      I .                      .                 .       1111      22          3     45.                      . I
      I .                      .                 .     111        222.         3    4 5.                      . I
      I .                      .                 .1111           222  .       3     4 5.                      . I
   2. +..+.....................+.................+1..............2....+..........3.....45.+..................+..+
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
   3. +..+.....................+.................+1..............2....+..........3.....45.+..................+..+
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                      .                 .1              2    .        3     45 .                     . I
      I .                                        .1              2    .        3     45 .                     .;I
   4. +..+.....................+.................+1..............2....+..........3.....45.+..................+..+
      ---I---------+---------I---------+---------I---------+---------I---------+---------I---------+--------I---
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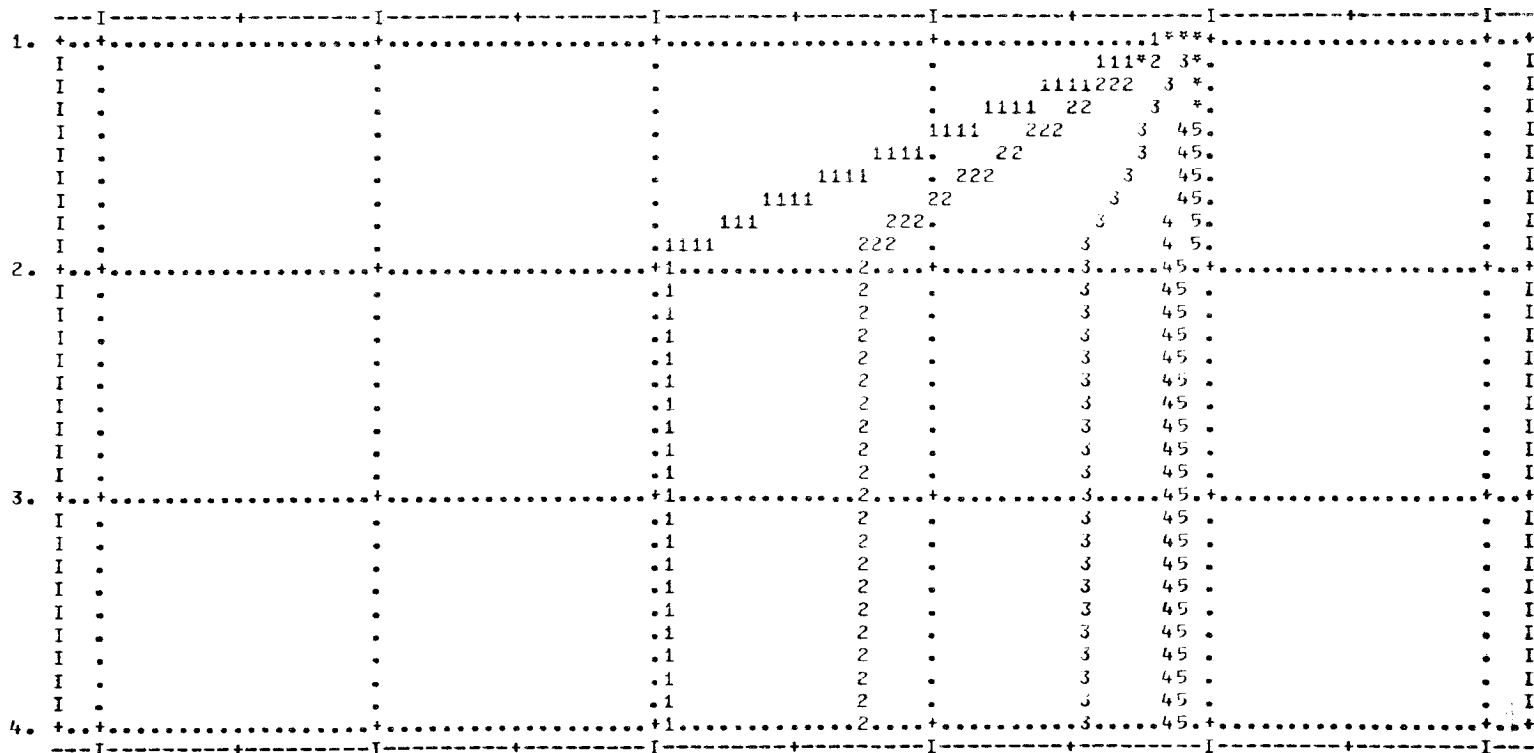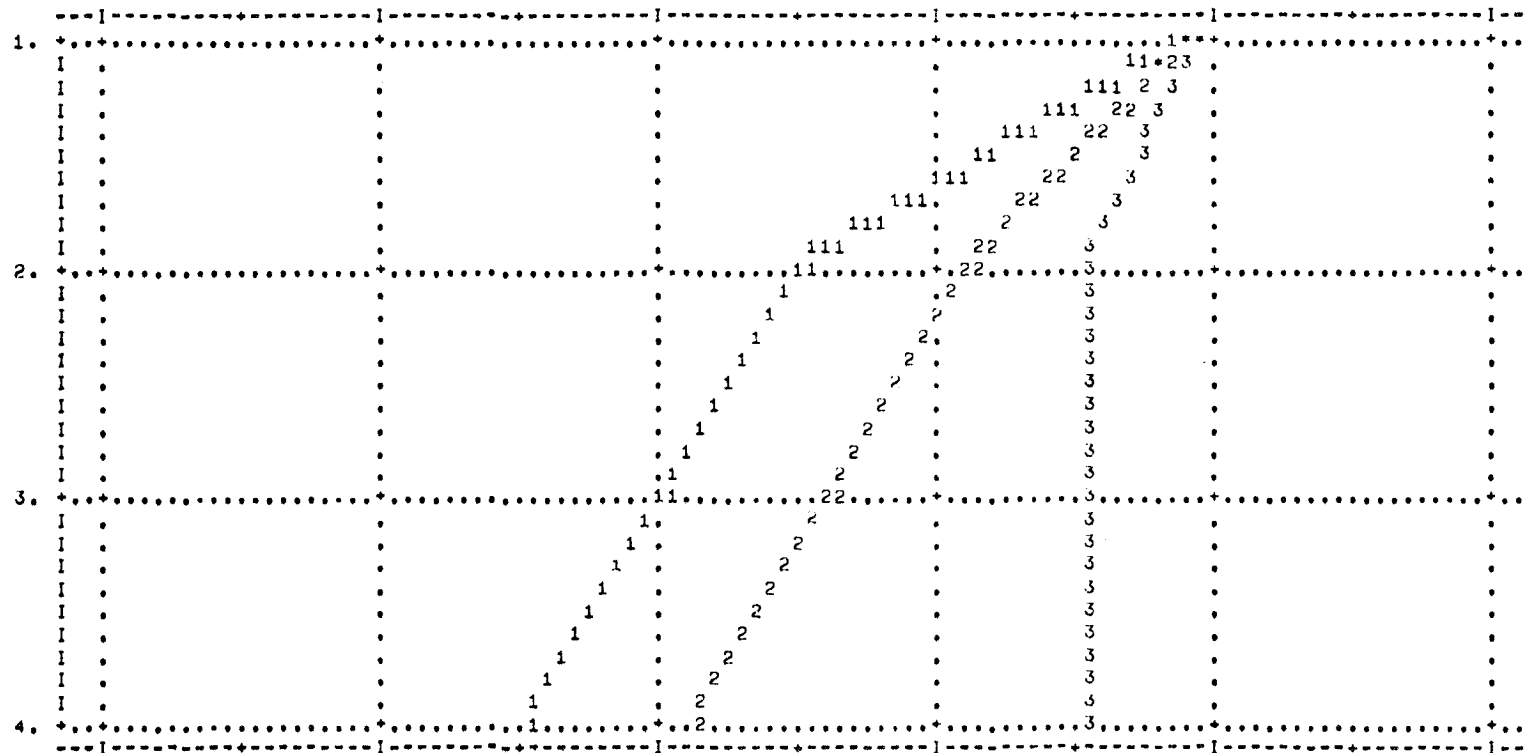
II) The isotopes I-136, I-137, I-138, I-139 and I-140
are probably the most efficient precursors of de-
layed neutrons.

Fig. 13 shows that the gas extraction of I-131 is very
efficient but rather strongly dependant on the postu-
lated gas extraction rate. For the lowest rate the I-131
decreases by a factor of approx 10, but for the middle
case - variant 3 this factor increases to more than 100
and for the highest rate (4) reaches approx 1000. For the
same gas extraction rates the loss of iodine isotope de-
layed neutron precursors is negligible (see iodine-136 -
140, fig. 14).

7. The xenon isotopes are also an important factor in the
core accident hazard. Gas extraction decreases the amount
of xenon in the core significantly including the long
lived Xe-133 and Xe-135 (fig. 15 and 16). The short life
xenon isotopes are only slightly influenced by the gas
extraction.

8. The next controlling radionuclide is Cs-137. From fig. 17
it is seen that the gas extraction does not aid in the
removal of this nuclide from the molten fuel, a factor
of 7 is all that can be obtained. But the amount is not
the only important factor, the volatility of caesium is
also of interest.

From fig. 4 it is clear that the caesium chloride has a
much lower partial pressure than that of caesium metal
or caesium oxide in the same temperature range.

A very important problem arises with the possibility of
the nuclear transformation of fission products.

ALL ISOTOPES WITH HALF-LIFE BETWEEN 0.3 AND 3 Y

```
        GRAPH   1 -  ELEMENT RU   AT.NO. 44   ISOTOPE 106   HALF-LIFE 1.000E+00 YRS
1.00E-08             1.00E-06              1.00E-04              1.00E-02           1.00E+00              1.00E+02
        GRAPH   2 -  ELEMENT SB   AT.NO. 51   ISOTOPE 125   HALF-LIFE 2.700E+00 YRS
1.00E-08             1.00E-06              1.00E-04              1.00E-02           1.00E+00              1.00E+02
        GRAPH   3 -  ELEMENT CE   AT.NO. 58   ISOTOPE 144   HALF-LIFE 2.840E+02 DAY
1.00E-08             1.00E-06              1.00E-04              1.00E-02           1.00E+00              1.00E+02
        GRAPH   4 -  ELEMENT PM   AT.NO. 61   ISOTOPE 147   HALF-LIFE 2.620E+00 YRS
1.00E-08             1.00E-06              1.00E-04              1.00E-02           1.00E+00              1.00E+02
        GRAPH   5 -  ELEMENT EU   AT.NO. 63   ISOTOPE 155   HALF-LIFE 1.810E+00 YRS
1.00E-08             1.00E-06              1.00E-04              1.00E-02           1.00E+00              1.00E+02
```


Fig. 18

25
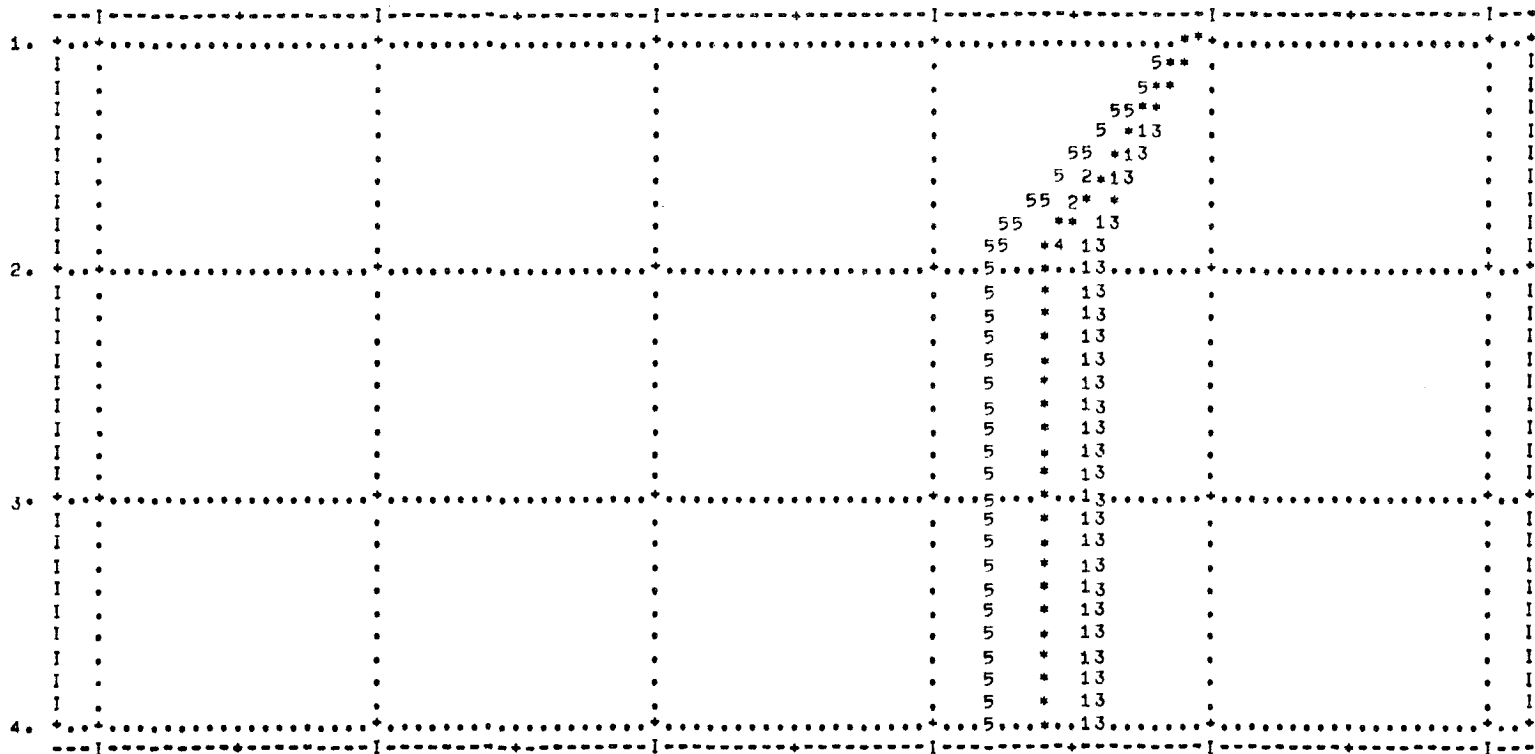
N36 WITH IS85,N54 WITH IS135,N55 WITH IS137

        GRAPH   1 -   ELEMENT KR   AT.NO. 36   ISOTOPE   85   HALF-LIFE 1.076E+01 YRS
1.00E-08   1.00E-06   1.00E-04   1.00E-02   1.00E+00   1.00E+02
        GRAPH   2 -   ELEMENT XE   AT.NO. 54   ISOTOPE 135   HALF-LIFE 9.150E+00 HRS
1.00E-08   1.00E-04   1.00E-04   1.00E-02   1.00E+00   1.00E+02
        GRAPH   3 -   ELEMENT CS   AT.NO. 55   ISOTOPE 137   HALF-LIFE 3.000E+01 YRS
1.00E-08   1.00E-06   1.00E-04   1.00E-02   1.00E+00   1.00E+02

Fig. 19

26

One of the methods especially for Cs-137 and Sr-90 is their
irradiation in a fast high flux reactor. (The use of fast
reactors is important because the absorption cross sections
for thermal neutrons are relatively small - 0.1 and 0.8 barns.)
It is important that the nuclide in question is more or less
isotopically pure for two reasons

    I) the neutron absorption in other isotopes

   II) the increased volume.

In the case of caesium there are three isotopes of importance
for neutron irradiation techniques they are: (yield for fast
fission of Pu-239)

Cs-135	stable	yield 6.91 atom %
Cs-135	t 1/2 - 2.1 x 10^6 years	yield 7.54 atom %
Cs-137	t 1/2 - 30 years	yield 6.69 atom %

If isotopic separation is not possible the amount of caesium
for neutron irradiation must be approx 3.15 times bigger than
for pure Cs-137 and the macroscopic absorption cross section
is probably more than 20 times bigger.

With the continuous in-core gas extraction system proposed
here the separation of caesium isotopes - particularly Cs-137
from Cs-135 (long lived) and Cs-133 (stable) is possible, as
is described elsewhere (Taube 1974).

8.   Other nuclides with 'mean life times' greater than 0.3
    years and shorter than 3 years as shown in fig. 18 - that
    is Ru-106, Sb-125, Ce-144, Pm-147 and Eu-155 are all little
    affected by the gas extraction.

9. Fig. 19 shows the most hazardous nuclides together Kr-85, Sr-90, Xe-135, Cs-137.

10. Fig. 20, 21, 22 show the estimated values of radioactivity for some selected nuclides in a 2000 MW(th) fast reactor for different cases

   a) without continuous gas extraction and with 85 days irradiation period.

   b) with gas extraction at the highest rate

   c) with gas extraction at the lowest rate.

Conclusions

The model of a possible continuous in-core gas-extraction process described here for a core fuelled by molten chlorides has the following features.

1. The extraction rate of the precursors of the delayed neutrons can be reduced to an insignificant value - even if these are very volatile. e.g. Kr, Xe, Br, I etc.

2. The extraction rate of I-131, the most hazardous fission product, prominent in any severe core-accident, is so high that the steady state concentration of this nuclide is reduced by more than 1000 times which significantly improves the reactor safety.

Fig. 20   Krypton-85

Fig. 21   Iodine-131

Fig. 22   Xenon-135

decay only

Rate of Extr: 10^{-3}

Rate of Extr: 10^{-1}

Activity
Curie

Time of fuel irradiation
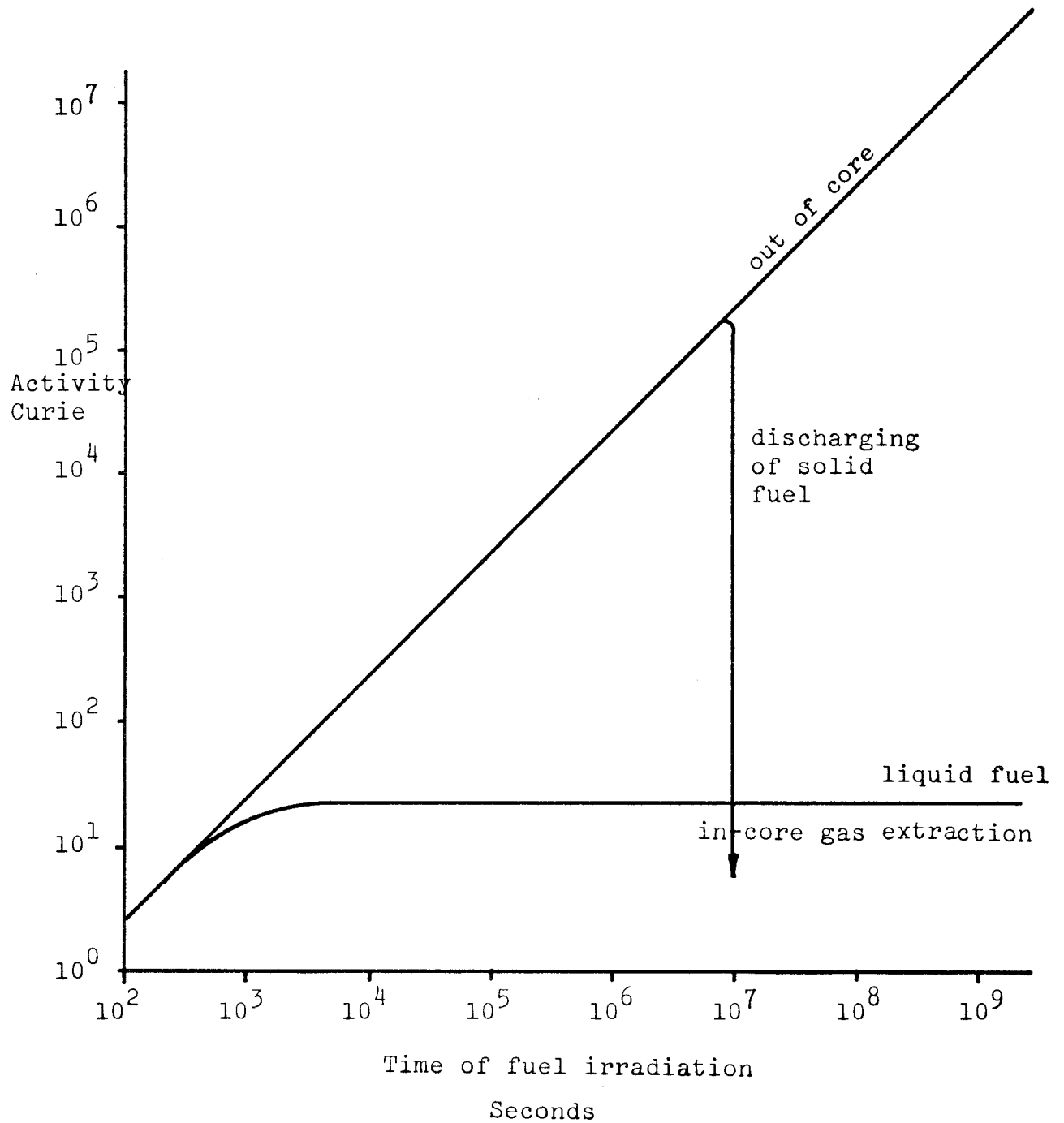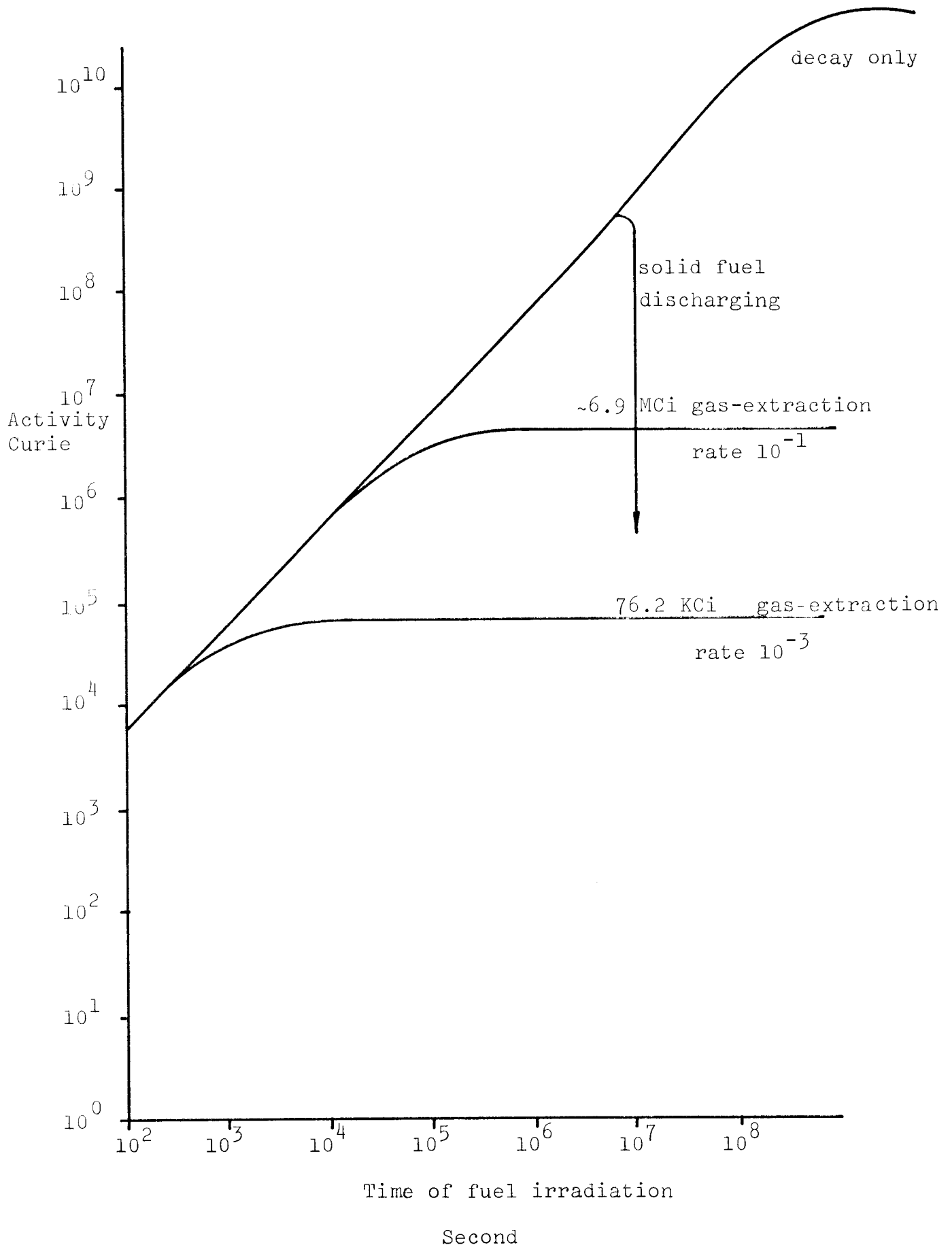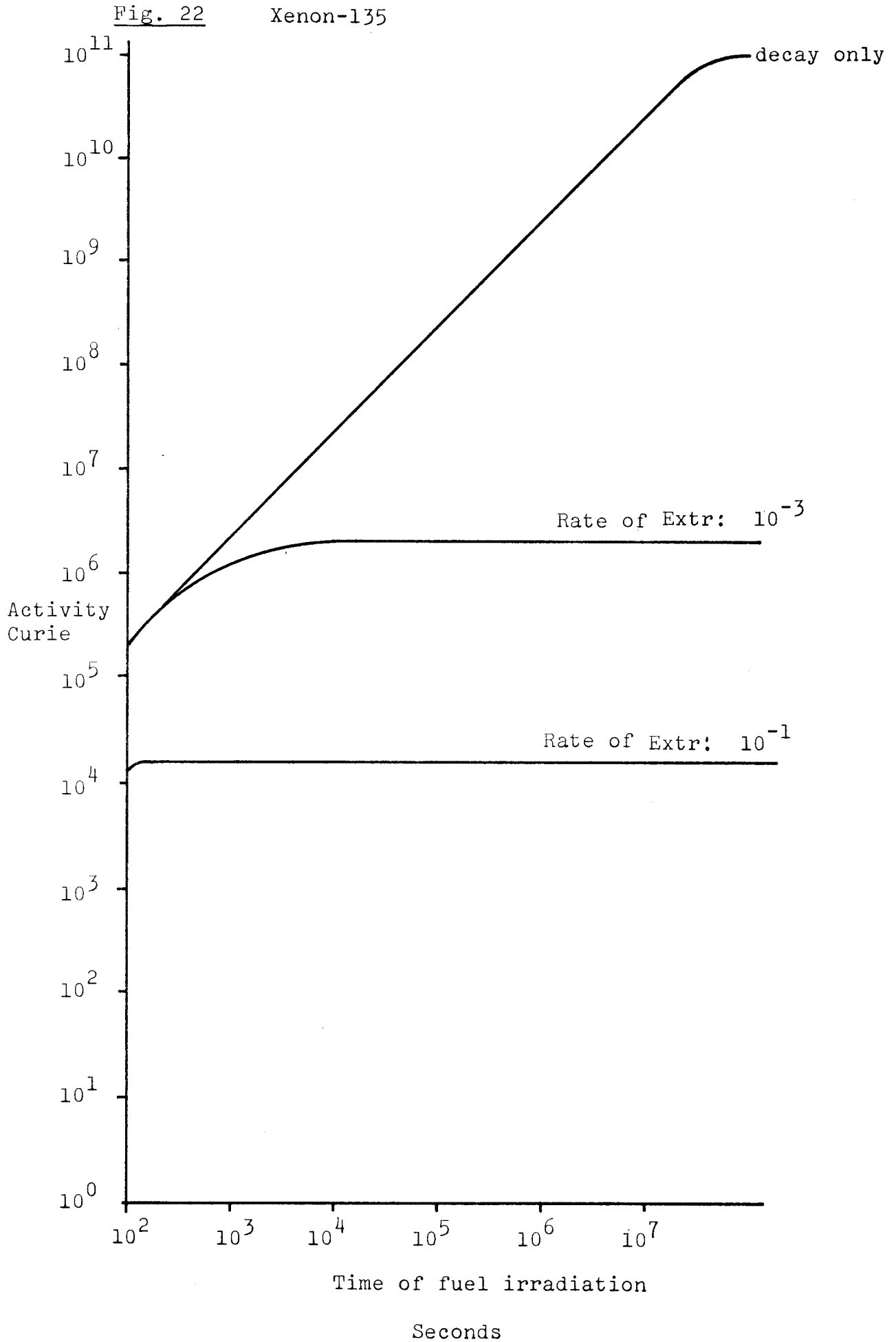
Seconds

3. The extraction rate of Cs-137 is rather small and results in the diminishing of the caesium concentration by only 7 times when compared to the normal liquid fuel reprocessing having a time constant of 10^6 seconds or $\simeq$ 10 days mean dwelling time of the liquid fuel in the reactor. In the solid fuel case the same amounts of Cs-137 are accumulated after approx 100 days that is ~3.5 months dwell time in the core. A special feature of the molten chlorides fuel is that the vapour pressure of CsCl is much lower than CsO_2/Cs metal in solid oxide or carbide fuels, decreasing the amount of vapourised caesium in the case of a core accident. Another important point is that it appears possible to separate the caesium isotopes 135 and 137.

4. Further hazardous fission products such as Xe-133 and Xe-135 can also be effectively extracted and the steady state level is >100 times lower than in the unextracted core.

5. The gas extraction rate influences also the oxygen extraction from the molten fuel which considerably improves the corrosion behaviour of metallic molybdenum and also the stability of the uranium and plutonium chlorides (Taube 1974).

Acknowledgements

Appendix I
<u></u>

Calculation of the gas stream rates.

The 2000 MW(th) reactor has a fission rate of about

$$2 \times 10^9 \text{ W} \times 3.1 \times 10^{10} \frac{\text{fiss/s}}{\text{W}} = \frac{1}{6.02 \times 10^{23}} \simeq 100 \text{ }\mu\text{M Pu/}_\text{s}$$

$$(\mu\text{M} = \text{micro Mol} = 6.02 \times 10^{23} \times 10^{-6} = 6.02 \times 10^{17} \text{ atoms})$$

The relative amounts of volatile nuclides is estimated very roughly here as 0.2, that is 2×20 $\mu\text{M/}_\text{s}$ of gaseous volatile elements. The volume of these elements at ~950 $^\text{o}$C and ~10 bars pressure is

$$40 \times 10^{-6} \text{ mol/}_\text{s} \times \frac{22.2 \times 10^3 \text{ cm}^3}{\text{mol}} \times \frac{1253 \text{ K}}{273 \text{ K}} \times \frac{1}{10} \simeq 0.4 \text{ cm}^3\text{/}_\text{s}$$

This amount of 0.4 $\text{cm}^3\text{/}_\text{s}$ must be removed from the core by means of continuous gas extraction. If we postulate a helium stream (with probably 1 mol % of hydrogen) of 4 $\text{cm}^3\text{/}_\text{s}$ then it seems the volatile elements can be removed. The assumed dwelling time of the gas bubbles in the core is 10 to 1000 s. The volume of the gaseous phase in the core is from 40 to 4000 cm^3.

The total volume of fuel in the core equals

$$8.75 \frac{\text{m}^3}{\text{core}} \times 0.386 \text{ fuel fraction} = 3.377 \text{ m}^3$$

The volume of gas bubbles given above equals from 10^{-5} to 10^{-3} of the fuel volume. This should not give rise to problems of criticality particularly in the steady state.

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