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Fast Reactors Using Molten Chloride Salts as Fuel

Final Report (1972-1977)

M. Taube

Würenlingen, Januar 1978

FAST REACTORS USING
MOLTEN CHLORIDE SALTS AS FUEL

FINAL REPORT (1972 - 1977)

prepared by M. Taube

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Summary

This report deals with a rather exotic "paper reactor" in which the fuel is in the form of molten chlorides.

(a) Fast breeder reactor with a mixed fuel cycle of thorium/uranium-233 and uranium 238/plutonium in which all of the plutonium can be burned *in situ* and in which a denatured mixture of uranium-233 and uranium-238 is used to supply further reactors. The breeding ratio is relatively high, 1.58 and the specific power is 0.75 GW(th)/m^3 of core.

(b) Fast breeder reactor with two and three zones (internal fertile zone, intermediate fuel zone, external fertile zone) with an extremely high breeding ratio of 1.75 and a specific power of 1.1 GW(th)/m^3 of core.

(c) Extremely high flux reactor for the transmutation of the fission products: strontium-90 and caesium-137. The efficiency of transmutation is approximately 15 times greater than the spontaneous beta decay. This high flux burner reactor is intended as part of a complex breeder/burner system.

(d) Internally cooled fast breeder in which the cooling agent is the molten fertile material, the same as in the blanket zone. This reactor has a moderate breeding ratio of 1.38, a specific power of 0.22 GW(th)/m^3 of core and very good inherent safety properties.

All of these reactors have the fuel in the form of molten chlorides: $PuCl_3$ as fissile, UCl_3 as fertile (if needed) and NaCl as dilutent. The fertile material can be $^{238}UCl_3$ as fertile and NaCl as dilutent. In mixed fuel cycles the $^{233}UCl_3$ is also a fissile component with $^{232}ThCl_4$ as the fertile constituent.

In some special cases a hypothetical molten fluoride has been checked using PuF_3 as the fissile, UF_3 as the fertile and NaF and ZrF_4 as the dilutents components. In this case one obtains a lower but still respectable breeding ratio of 1.5.

In all cases a directly coupled continuously operating reprocessing plant is proposed. Some of the technological problems of reprocessing are discussed. Furthermore the report touches on some of the difficulties associated with corrosion arising from the use of these molten media coupled with the irradiation effects such as structural damage from fast neutrons.

The thermohydraulic studies show that even under the extreme operating conditions of very high neutron fluxes and high specific power, cooling is possible, in most cases by out-of core cooling but also in one or two cases cooling internally in the core.

Some molten salt reactor specific safety problems are discussed.

The influence of fast neutrons on the chlorine, forming sulphur by the (n,p) reaction has been experimentally investigated and the results are reported briefly.

With this report the work of several years at the Swiss Federal Institute for Reactor Research is brought to a conclusion.

CONTENTS

FORWARD

The history of the development of fission reactor concepts using molten salt as fuel media is as old and as complex as the history of the development of nuclear power itself. The ups and downs have followed those of the parent technology but the swings have been if anything more violent. In 1976 for example molten salt technology all but died out but then in 1977 a new attempt at revival was begun this time associated with the new interest in proliferation-proof systems.

The author of the present paper has a profound belief that the concept of molten slat reactors coupled with continuous reprocessing and the associated waste management will become an important feature of nuclear strategy perhaps in 10 or 20 years time.

In this report the efforts in this field over the last six years are summarised.

1. MOLTEN SALT REACTORS. GENERAL DESCRIPTION

1.1 Methods of classification

There are many ways of classifying a reactor type. One such possibility is shown here.

a) Method of cooling

b) Flux intensity related also to specific power density

c) Number of zones in the reactor

d) Kind of fissile nuclides and fuel cycles

e) Neutron energy

f) Purpose of the reactor

g) Dilutent for the molten salt

It is clear that such an arbitrary classification is not necessarily internally compatible and not all reactor types fall easily into the scheme chosen.

1.2 Method of cooling. External: Internal

Molten fuel reactors differ from the point of view of the cooling system. The following are three types of molten fuel reactors:

Externally cooled, where the molten fuel is pumped out of the core to the external heat exchanger. In this type of reactor, only fuel and fertile material are present in the core (no coolant). The large amount of molten fuel ouside the core does not of course contribute to the critical mass.

This type of reactor has been discussed for example by Nelson, (Argonne 1967) and Lane (USA 1970) especially as a high flux materials testing fast reactor.

In externally cooled fast reactors the loss of a portion of the delayed neutrons could adversely affect reactor control. Also the biological shielding outside the core is very expensive. In this paper most of the reactors discussed are externally cooled.

Internally, direct cooled reactors: here the cooling agent is pumped directly into the core where, after mixing, the fuel in the lower part of the core is separated and pumped out of the core to the heat exchanger. The direct contact of molten fuel with molten coolant has several particular advantages! very good heat transfer, no coolant tubes (or cladding), possibility of transporting fission products.

The disadvantages are unfortunately, also numerous: problems of mixing and separating the fuel and coolant, corrosion, etc. This type of reactor has also been studies, e.g. cooled by molten lead (Long, Harwell and Killingback, Winfrith 1967), cooled by boiling mercury (Taube, Warsaw 1966) and cooled by boiling aluminium chloride (Taube, Warsaw 1966). This type of reactor must be considered as an "extremely exotic type", and only some references are given here.

Internally indirectly cooled reactor: here the cooling agent flows through tubes in the core. Heat is transferred from fuel to coolant across the tubes. No direct contact between molten fuel and liquid or gaseous coolant is permitted. These types have also been studied, in most cases using sodium as a coolant, (Nelson, Argonne 1967) or molten chlorides of uranium (Taube, 1970). See Fig. 11.

1.3 Intensity of neutron Flux

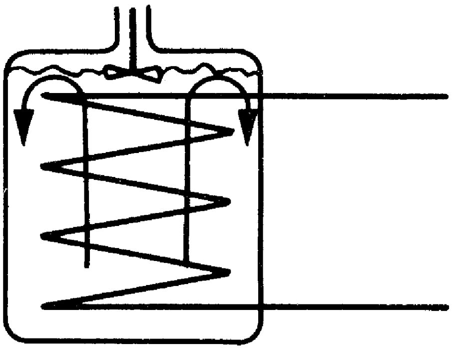
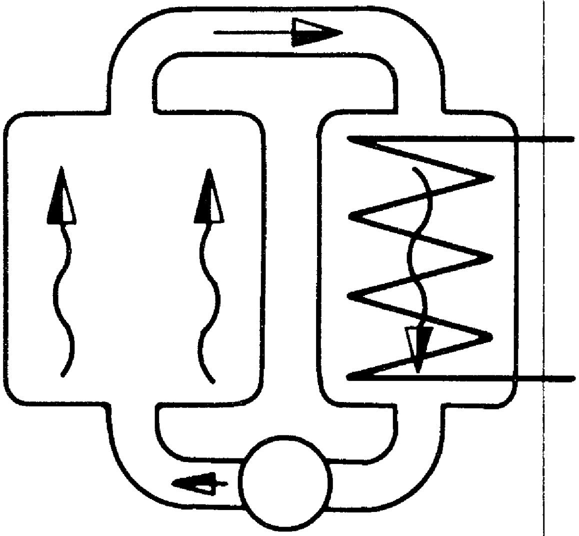
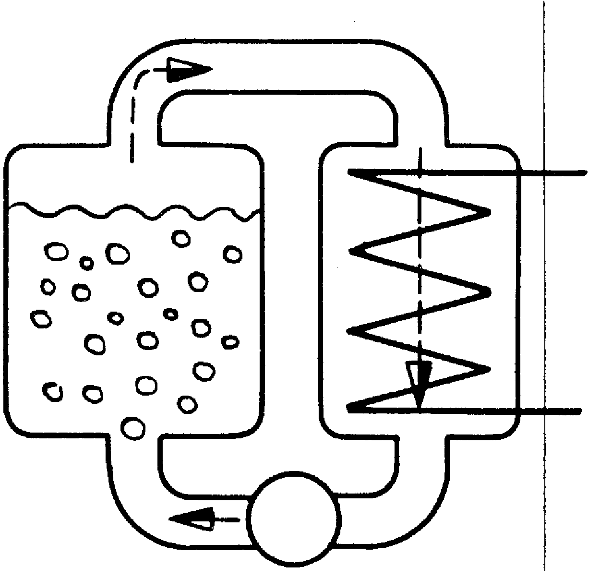
The molten salt reactors discussed here can be used for two more or less quite different purposes.

- power production and fissile breeding, which is self evident
- neutron production for nuclear transmutation of the long radionuclides produced in power reactors.

In this report both types have been considered

- power breeding ractors with a mean power level of approx 3 GW(th) and steam production with over critical parameters.
- burner reactors with a very high neutron flux particularly in the internal zone for neutron moderation when the thermal flux reaches 3×10^{16} n $cm^{-2}s^{-1}$.

Fig. 1.1 TYPES OF REACTOR COOLING SYSTEMS

TYPE	SCHEME	CHAPTER
INTERNAL INDIRECT COOLING		CHAPTER 5
EXTERNAL INDIRECT COOLING		CHAPTER 2, 3, 4
INTERNAL DIRECT COOLING (BOILING)		here not discussed

1.4 Number of core zones

The division of the reactor into several zones must be considered from the point of view of neutronics, thermohydraulics and safety.

The organisation of multiple zones is easier in the case of molten fuel reactors than for solid fuel reactors. In this report two types are discussed

- with two zones
- with three zones including outer and inner fertile zone, (see Fig. 1.2).

1.5 Type of Fissile Nuclide. Plutonium: Uranium

The fast reactors show excellent neutron properties, not only for the fuel cycle:

Uranium -238/Plutonium but also
Thorium -232/Uranium-233

Also a mixed fuel cycle of both types has some spectial advantages. Fig. 1.3 shows the nuclear properties of the fissile nuclides.

1.6 Neutron energy: Thermal and Fast

The reactors discussed here are all fast reactors. Thermal reactors however have also been extensively and intensively investigated during the 1960's and 1970's in Oak Ridge National Laboratory USA. (Rosenthal at all, 1972)

Fig. 1.2 TYPES OF REACTORS

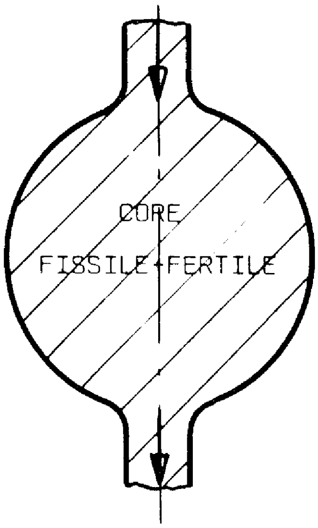
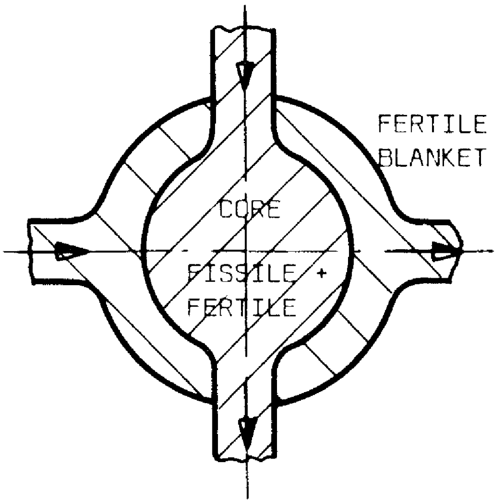
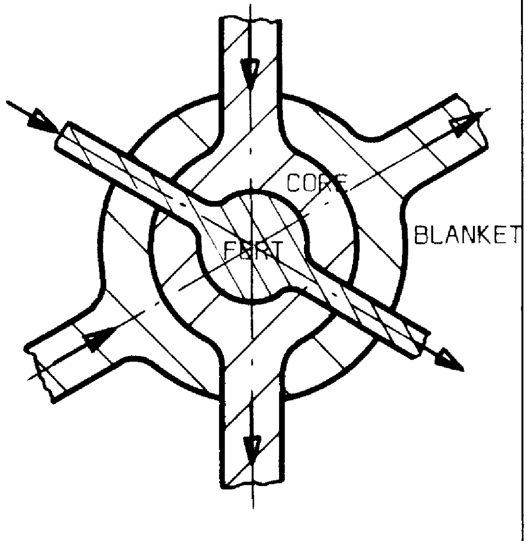
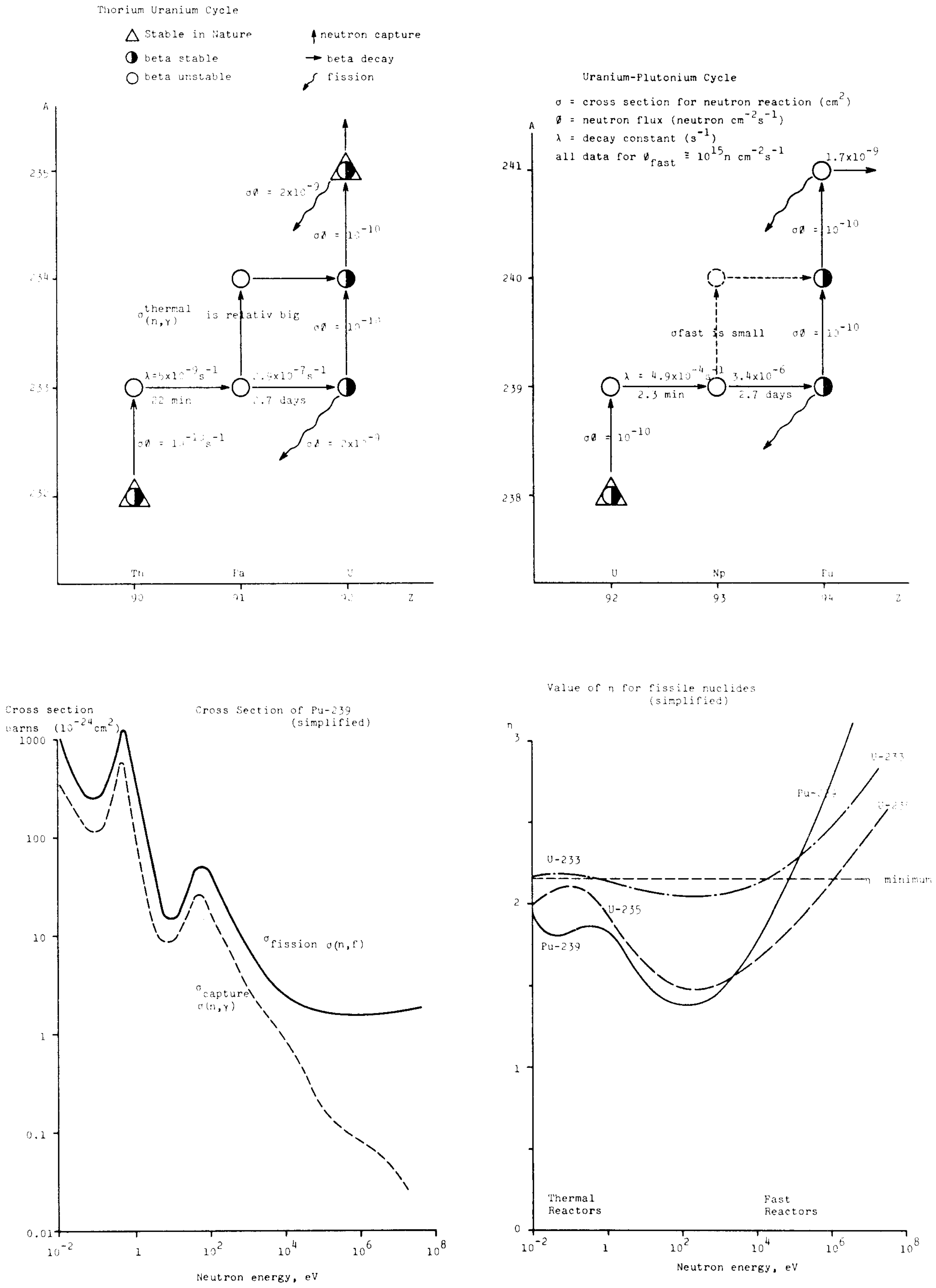
NUMBER ZONE	GEOMETRY	PROPERTIES	
		POSITIVE	NEGATIVE
ONE		SIMLICITY OF TECHNOLOGY	RELATIV LOW BREEDING RATIO, BECAUSE OF TOO SOFT NEUTRON.
TWO		THE OPTIMAL DESIGN	
THREE		GOOD USE OF NEUTRONS	THE GEOMETRY IS VERY COMPLEX THE NEUTRON FLUX DISTRIBUTION IS DISTURB

Fig. 1.3


Thorium Uranium Cycle
Stable in Nature
beta stable
beta unstable
neutron capture
beta decay
fission
A
235
234
233
232
σØ = 2x10^-9
σØ = 10^-10
σ thermal (n,γ) is relativ big
σØ = 10^-10
λ=5x10^-4 s^-1
22 min
2.9x10^-7 s^-1
2.7 days
σØ = 10^-10 s^-1
σØ = 2x10^-9
Th
Pa
U
90
91
92
Z
Uranium-Plutonium Cycle
σ = cross section for neutron reaction (cm^2)
Ø = neutron flux (neutron cm^-2 s^-1)
λ = decay constant (s^-1)
all data for Ø fast ≡ 10^15 n cm^-2 s^-1
A
241
240
239
238
1.7x10^-9
σØ = 10^-10
σfast is small
σØ = 10^-10
λ = 4.9x10^-4 s^-1
2.3 min
3.4x10^-6
2.7 days
σØ = 10^-10
U
Np
Pu
92
93
94
Z
Cross section
barns (10^-24 cm^2)
Cross Section of Pu-239 (simplified)
1000
100
10
1
0.1
0.01
σ fission σ(n,f)
σ capture σ(n,γ)
10^-2
1
10^2
10^4
10^6
10^8
Neutron energy, eV
Value of η for fissile nuclides (simplified)
η
3
2
1
0
U-233
Pu-239
U-235
U-233
U-235
Pu-239
η minimum
Thermal Reactors
Fast Reactors
10^-2
1
10^2
10^4
10^6
10^8
Neutron energy, eV

1.7 Purpose of the reactors

The principle purposes of the large ractors proposed can be classified as follows. (Table 1.1)

Table 1.1

Reactor type	Primary Aim	Secondary Aim	Comments to be found in chapter
Power	Electrical energy $T > 600^{o}C$	Production of fissile nuclides $BR > 1$	ch.5
Breeder	Production of fissile nuclide B.R. ∿ optimum	Production of electrical energy	ch.3
High flux	Neutron Flux Φ $(n\ cm^{-2}s^{-1})$ $> 10^{16}$ for transmutation	Production of electrical energy	ch.4
High Temperature	$T > 850^{o}C$ for chemical reactions	Production of electrical energy	not discussed here
Non-proliferating	Maximum security. No plutonium output	Production of electrical energy	ch.2
Propulsion	Heat for steam turbine		not discussed here
Space Heating	Heat with $100^{o}C < T < 200^{o}C$		not discussed here

1.8 Fuel Components. For Molten salt, fluoride and chloride

In the thermal molten salt reactor the best fuel compound is unduobtedly the fluoride.

For fast reactors the use of chlorine as the compound seems to be preferable but the use fo fluorine (as zirconium and sodium fluoride) as dilutent is not excluded.

1.9 Short resumé of the classification

Table 1.2 brings together all these characteristics in an attempt at classification.

Table 1.2

		This work:	
		yes	no
Method of Cooling	External	x	
	Internal Direct		x
	Internal Indirect	x	
Flux Intensity	High	x	
	Low	x	
Number of zones	One		x
	Two	x	
	Three	x	
Fissile	Plutonium	x	
	Uranium	x	
	Mixed	x	
Energy of Neutrons	Thermal		x
	Intermediate		x
	Fast	x	
Aim	Power	x	
	Breeder	x	
	High Flux burner	x	
Dilutents	Fluoride	x	
	Chloride	x	

1.10 Method of Neutronic Calculation

Almost all results given here have been obtained using the following calculational method

- the reactor code: ANISN
- number of zones: 5, 6 or 7
- 40 - 100 spatial positions
- order of quadrature S_4 checked by S_8
- neutron groups: 22 or 23 groups including the thermal neutrons (see Table 1.3)
- anisotropy by first order Legendre expansions
- library ENDF/BI, BII and BIV processed by code GGC-3 and GGC-4
- the management of additional sub-routines have been realised by RSYST.

Table 1.3 Relative Fluxes in Each Group

g	Upper boundary	Mean value	Δu (Lethargy)	Centre	Core boundary
1	15 MeV	12.2 MeV	0.4	10^{-5} (.0002)	10^{-4}
2	10	8.18	0.4	.0010 (.0026)	.0006
3	6.7	5.49	0.4	.0045 (.0116)	.0029
4	4.5	3.67	0.4	.0112 (.0286)	.0072
5	5.0	2.46	0.4	.0222 (.0552)	.0146
6	2.0	1.65	0.4	.0407 (.0901)	.0285
7	1.35	1.23	0.2	.0264 (.0536)	.0193
8	1.11	1.00	0.2	.0281 (.0536)	.0215
9	0.91	0.78	0.3	.0475 (.0837)	.0379
10	0.67	0.55	0.4	.0924 (.1373)	.0787
11	0.45	0.37	0.4	.1072 (.1254)	.0972
12	0.30	0.25	0.4	.1162 (.1042)	.1136
13	0.20	0.165	0.4	.1044 (.0749)	.1064
14	0.135	0.108	0.45	.1046 (.0662)	.1108
15	86.5 keV	59.5 keV	0.75	.1061 (.0549)	.1208
16	40.8	25.0	1.00	.1043 (.0394)	.1262
17	15.0	8.0	1.25	.0581 (.0145)	.0758
18	4.31	2.94	0.75	.0047 (.0011)	.0066
19	2.03	1.39	0.75	.0148 (.0025)	.0208
20	0.96	0.67	0.75	.0048 (.0004)	.0079
21	0.45	0.24	1.25	.0008 (10^{-4})	.0017
22	0.13 keV	0.4 eV	5.75	$< 10^{-4}$ $<$ (10^{-4})	$< 10^{-4}$
Total	0.4 eV -	15 MeV	$\Sigma = 17.40$	1.0000	1.0000

2.1 BREEDER REACTOR WITH PLUTONIUM BURNING *IN SITU*

2.1.1 Aim of this Concept

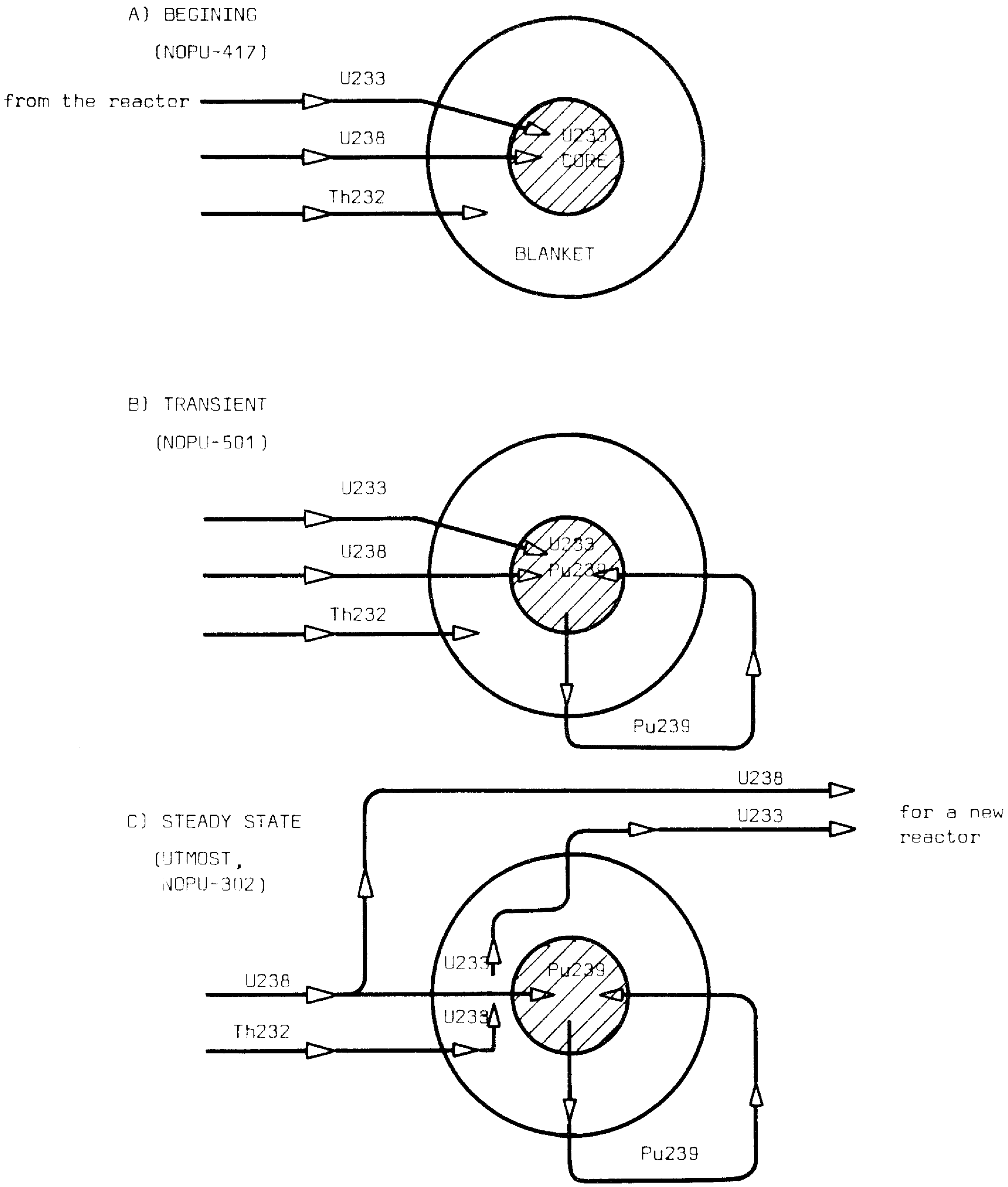
The aim here is to demonstrate the possibility of using a molten chlorides fast breeder reactor with external cooling as a device for consuming all plutonium produced, *in situ*. At first the reactor is fuelled with denatured uranium -233/uranium -238 and this is changed stepwise to a feed of thorium and depleted or natural uranium only.

Such a reactor will have the following phases in its fuel cycle (Table 2.1) See Fig. 2.1

Table 2.1

Phase	Fuel input		Fissile burned *in situ*	Fuel output	
	Fertile	Fissile		Fissile	Fertile
Start. Build-up phase. (Fig. 2.1 A)	U-238: 70% Th	U-233: 30%	U-233	none	none
Transient Phase (Fig. 2.1 B)	U-238: 70% Th	U-233: 30%	Pu-239 U-233	none	none
Steady State	U-238 Th	none	Pu-239 + other Pu-isotopes	mixed U-233: 30%	U-238

Fig. 2.1 Two-zones Reactors with uranium-233/plutonium-239



2.1.2 Reactor in the Build-up phase

At the start of the cycle the reactor core is fuelled by uranium-233 denatured with uranium 238 (see Fig. 2.1). (Table 2.2)

Table 2.3 gives information concerning

- the method of calculation
- densities of elements in each of the 5 zones (core, wall, blanket, wall, reflector)

  Table 2.4 shows the neutron balance in the core and blanket

Table 2.5 gives the breeding ratio calculated by a microscopic method of the form:

$$BR_{micr} = \left(\frac{\nu-1+\alpha}{1+\alpha}\right) + \left(\frac{(\nu-1)\delta}{1+\alpha}\right) - \frac{L_{tot}+\alpha}{1+\alpha}$$

- and the macroscopic method by

$$BR_{macr} = \frac{\text{production rate of fissile nuclide}}{\text{rate of destruction of fissile nuclide}}$$

- and the maximum neutron flux which gives information on the flux spectrum in the core.

Table 2.6 shows the geometry of this reactor e.g. see Fig. 2.2

- radius: 0.955 m
- volume: 3.65 m^3

and

- specific power: 0.75 GW(therm)/m^3 of core
- total power: 2.8 GW(therm)

and inventories of fissile and fertile materials.

Table 2.7 gives some information concerning

- the material flux in this type of reactor. (for more see section 2.3).

TABLE 2.2

OBJECT : Thorium-uranium Breeder with Plutonium burning in situ

REACTOR TYPE : Power, Breeder

GEOMETRY : INTERNAL ZONE : Fuel

WALL :

INTERMEDIATE ZONE : See Fig. 2.2

WALL :

EXTERNAL ZONE : Fertile zone

WALL, REFLECTOR :

POWER (GW thermal) : 2.8

POWER DENSITY (GW therm/m^3 core) : 0.75

NEUTRON FLUX, MEAN (n/cm^2s) : 1.6×10^{16}

FISSILE NUCLIDE : Pu-239/Pu-241 in core, fuel
: U233 in fertile

FERTILE NUCLIDE : U238 in core, Th232 in fertile

DILUTENT : Chloride

COOLING SYSTEM : Outer

BREEDING RATIO : 1.58

PARAMETER STUDIED : Make up reactor with U233/U238
: transient reactor with U233 + Pu239
: steady state reactor with Pu239
:

METHOD OF NEUTRONIC CALCULATION : See table 1.10

Fig. 2.2 Three zones reactor, 2.8 GW th (meter)

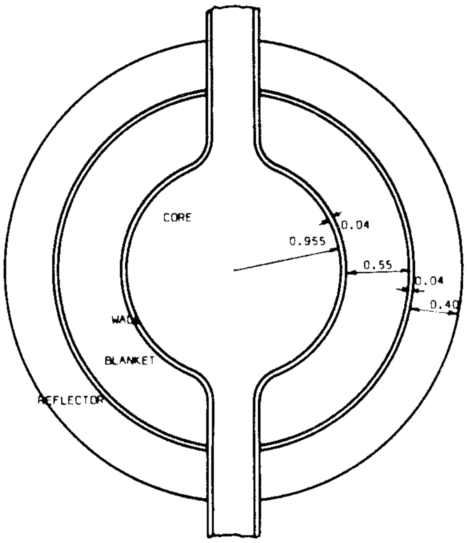


Table 2.3

```
                                    UTMOST-417,NOPU.
                                    ********************

TITLE : SPHERIC GEOM.
DATE  :  08/09/77


                          INPUT DATA (RSYST FORMALISM)
                          *****************************

        ORDER OF SCATTERING              1
        QUADRATURE ORDER                 4
        NO. OF ZONES                     5
        NO. OF INTERVALS                40
        NO. OF GROUPS                   22
        EIGENVALUE MODIFIER        -2.000E-01
        PRECISION DESIRED           1.000E-03
        NORM. FACTOR                2.291E+20
        NEW PARM. MOD. SRCH.        1.000E+00


                          DENSITIES(ATOMS*1E-24/CM3)
                          **************************

                          CORE
                          ¬¬¬¬

    NA    = 7.64E-03     CL    = 1.30E-02     U238  = 4.75E-03
    U233  = 5.66E-04     PU239 = 7.31E-07     PU240 = 7.31E-07
    PU241 = 7.31E-07     FP239 = 1.61E-07

    RATIO U3/(PUTOT + U3) =    8.37

                          FIRST WALL
                          ¬¬¬¬¬¬¬¬¬¬

    C     = 6.62E-02     FE    = 2.88E-02

                          BLANKET
                          ¬¬¬¬¬¬¬

    NA    = 5.41E-03     CL    = 1.08E-02     TH232 = 5.41E-03
    PA    = 1.30E-05     U233  = 1.08E-05

                          2-ND WALL
                          ¬¬¬¬¬¬¬¬¬

    C     = 1.37E-02     FE    = 7.31E-02

                          REFLECTOR
                          ¬¬¬¬¬¬¬¬¬

    C     =   .0         FE    = 8.45E-02
```

Table 2.4

```
                 UTMOST-417,NOPU.
                 ********************

   TITLE : SPHERIC GEOM.
   DATE  :  08/09/77

*****************************************************************
*                                                               *
*                    NEUTRONICS MACR                            *
*                    ***************                            *
*                                                               *
*  ELEMENT         ABSORPTION             PRODUCTION            *
*  ¬¬¬¬¬¬¬         ¬¬¬¬¬¬¬¬¬¬             ¬¬¬¬¬¬¬¬¬¬            *
*    CORE                                                       *
*    ¬¬¬¬                                                       *
*  U233         69.992E+18      31.77 16.499E+19      71.95     *
*  U238         65.894E+18      29.91 59.530E+18      25.96     *
*  PU239        73.674E+15        .03 18.217E+16        .08     *
*  PU240        32.018E+15        .01 57.914E+15        .03     *
*  PU241        87.895E+15        .04 22.947E+16        .10     *
*  CL           11.638E+18       5.28   .0              .00     *
*  FE           21.973E+17       1.00   .0              .00     *
*  NA           10.552E+17        .48   .0              .00     *
*  C            13.334E+16        .06   .0              .00     *
*  FP           12.735E+14        .00   .0              .00     *
*                                                               *
*  TOTAL CORE   15.110E+19      68.60 22.499E+19      98.11     *
*                                                               *
*    BLANKET                                                    *
*    ¬¬¬¬¬¬¬                                                    *
*  TH232        50.643E+18      22.99 17.989E+17        .78     *
*  U233         11.181E+17        .51 25.407E+17       1.11     *
*  PA           69.619E+15        .32   .0              .00     *
*  CL           62.067E+17       2.82   .0              .00     *
*  FE           10.236E+18       4.65   .0              .00     *
*  NA           27.541E+16        .13   .0              .00     *
*  C            72.251E+13        .00   .0              .00     *
*                                                               *
*  TOT. BLAN.   69.177E+18      31.40 43.296E+17       1.89     *
*                                                               *
*  ENTWO *                            42.168E+17       1.81     *
*  LEAKAGE      13.285E+18       5.69                           *
*                                                               *
*  T O T A L    22.028E+19     100.00 22.921E+19     100.00     *
*                                                               *
*****************************************************************
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THE REACTION (n,2n)

Table 2.5

```
                        UTMOST-417,NOPU.
                        ********************

 TITLE : SPHERIC GEOM.
 DATE  :  08/09/77


***************************************************************
*                                                             *
*                    NEUTRONICS MICR                          *
*                    ****************                         *
*          ENTITY              SYMBOL             VALUE       *
*          ------              ------             -----       *
*     NI0                                         3.183       *
*     NI1                                         3.023       *
*     NI2                                         2.455       *
*     NI3                                         2.556       *
*     NI8                                         2.945       *
*     NI9                                         2.996       *
*     ALFM                                         .085       *
*     FASB                                         .319       *
*     ABPAR                                        .173       *
*     LEAG                                         .058       *
*     PRODUCTION                                  1.357       *
*     FAST BONUS                                   .572       *
*     ABSOR.PARS                                   .159       *
*     LEAKAGE                                      .137       *
*     BRED.RATIO MICR                             1.633       *
*     PU39 PRODUC                                 4.568       *
*     PU39 DESTRO                                  .007       *
*     U33 PRODUC                                  4.991       *
*     U33 DESTRO                                  7.111       *
*     BALANCE U233             BALA33            -2.126       *
*     BALANCE PU239            BALA39             4.561       *
*     TOTAL BREEDING MACR      BRTOT              1.342       *
*                                                             *
*   MAX. FLUX IS : 2.42E+15 FOR GROUP 11 IN INTERVAL   1      *
*                                                             *
***************************************************************
*COMMENT                                                      *
*FASB, N*SIGMA FISS.FERT / N*SIGMA FISS.FISSIONABLE.          *
*ABPAR, N*SIGMA PARAS / N*CAPT.(FERT + FISS).                 *
*LEAG, LEAKAGE / TOTAL ABSOR.(FROM TABLE NEUTRONICS).         *
*PRODUC, (ZNIM - 1 - ALFM) / (1 + ALF).                       *
*FAST BONUS, (FASB * (NI8 - 1)) / (1 + ALF).                  *
*ABSOR.PARS, ABPAR / (1 + ALF).                               *
*LEAKAGE, LEAG*ZNIM / (1 + ALF).                              *
*BRTOT, N*SIGMA TRANSMUTATION (TH + U8 + PU0) / N*SIGMA.      *
*CAPT. (U3 + PU9 + PU1).                                      *
***************************************************************
```

Table [illegible].8

```
                         UTMOST-417,NOPU.
                         ****************

 TITLE [illegible]
 DATE [illegible]

**********************************************************************
*                                                                    *
*    [illegible]            SYMBOL      UNIT            VALUE        *
*    -----------            ------      ----            -----        *
*      GEOMETRY                                                      *
*                                                                    *
*    CORE RADIUS            R(1)        M                 .955       *
*    WALL 1 THICKNESS       THICK(2)    M                 .040       *
*    BLANKET THICKNESS      THICK(3)    M                 .550       *
*    WALL 2 THICKNESS       THICK(4)    M                 .040       *
*    HEIGHT OF CYLINDER     HEIGHT      M                 .000       *
*    [illegible] RATIO      HTRAT       -                 .000       *
*    VOLUME OF CORE         VOLCOR      M**3             3.653       *
*    VOLUME OF BLANKET      VOLBL       M**3            11.328       *
*                                                                    *
*      POWER                                                         *
*                                                                    *
*    [illegible]            [illegible] GW               2.739       *
*    [illegible]            [illegible] GW                .057       *
*    [illegible]            POWTO       GW               2.796       *
*    [illegible]            [illegible] GW/M**3           .750       *
*    [illegible] BLANKET    [illegible] GW/M**3           .005       *
*    [illegible]            [illegible] MW/KG          857.336       *
*                                                                    *
*      INVENTORY                                                     *
*                                                                    *
*    TOTAL [illegible] INVENT. [illegible] KG            1.060       *
*    TOTAL PU INVENT.       PUTOT       KG               3.195       *
*    U233 IN CORE           U3C         KG             800.212       *
*    U233 IN BLANKET        U3B         KG              47.430       *
*    TOTAL U233 INVENT.     U3TOT       KG            6864.340       *
*    TOTAL TH INVENT.       THTOT       KG           23613.092       *
*    RATIO [illegible]      RATUB       -             2148.611       *
*                                                                    *
*      [illegible]                                                   *
*                                                                    *
*    [illegible] BURNUP     BURNR       -                 .002       *
*    [illegible]            TIME        YEARS             .010       *
*    [illegible]            [illegible] NEUT/S*CM2 1.260E+16       *
*                                                                    *
**********************************************************************
```

Table 2.7

```
*************************************************************
*                         TABLE                             *
*                        *******                            *
*         MATERIAL BALANCE OF THE BREEDER SYSTEM            *
*                  INCLUDING NEW REACTOR                    *
*************************************************************
*   *   THOR    *    PA    *   U233   *   U238   *   PU39   *
*************************************************************
*ORS   2.33E+06   6.06E+02   5.97E+02  -1.87E+02   5.69E+02*
*SBL   2.36E+04   5.69E+01   4.74E+01                      *
*FBL   8.84E+00   8.49E+00   7.99E+02                      *
*FTR   6.06E+02   5.97E+02              5.69E+02           *
*SCO                         8.00E+02   6.86E+03   1.06E+03*
*FFP                                    2.52E+02   9.21E-01*
*FCB   6.15E+02                         8.21E+02           *
*FLR   2.36E+00   5.69E-03   4.74E-03   6.86E+00   1.06E-03*
*SRE   2.36E+00   5.69E-03   8.00E-01   6.86E+00   1.06E-03*
*FWA   6.15E-01             -2.02E-01   6.86E-04   1.06E-04*
*FNW   2.33E+06             -2.02E+02  -1.01E+03           *
*SNW   2.36E+04             -2.04E+00  -1.02E+01           *
*************************************************************

WHERE

ORS = FLOW FROM THE ORE OR SYNTH. IN KG / YEAR
SBL = STEADY STATE IN THE BLANKET IN KG.
FBL = FLOW FISS. + PARAS.IN BLANKET IN KG / YEAR
FTR = FLOW FOR TRANSMUTATION IN KG / YEAR
SCO = STEADY STATE IN THE CORE IN KG
FFP = FLOW FISS.+ PARASITIC IN CORE IN KG / YEAR
FCB = FLOW TO THE CORE OR BLANKET IN KG / YEAR
FLR = FLOWTO REPROCESS AND RETURN IN KG / YEAR
SRE = STEADY STATE IN THE RE-PROCESSING IN KG
FWA = FLOW TO THE WASTE IN KG / YEAR
FNW = FLOW TO THE NEW IN KG / YEAR
SNW = IN THE NEW AFTER DTIME IN KG
```

The data given indicates that in the given geometry the nominal power and all related values: temperature, temperature gradient, velocity of circulating fuel, heat exchange etc. are as for the steady state reactor with only plutonium fuel in the core (see section 2.1.4).

2.1.3 The Transient Reactor

Very shortly after the start up of the reactor, a significant amount of plutonium has been produced in the core.

The total amount of plutonium chloride, after having been separated from the fission products but mixed with the uranium chloride and sodium chloride is circulated back into the core. The amount of fresh U-233 required is correspondingly smaller.

The reactor now burns two fissile nuclides, the uranium-233 and the reprocessed plutonium. The data given below refers to the case where approximately half of the fissile uranium-233 is replaced by plutonium-239 and plutonium-241.

Based on rather simplified assumptions concerning the isotopic composition of the plutonium it can be shown that the same core design is capable of burning the mixture of fissile materials maintaining approximately the same power level.

Tables 2.8, 2.9, 2.10, 2.11 and 2.12 give the equivalent values previously shown for the build-up phase.

These confirm the suitablility of the core design for both the build-up phase and the transient phase.

2.1.4 The steady state reactor

This reactor phase is in fact the main object of the work covered in this report, the steady state fast breeder reactor having the following features.

Table 2.8

```
                         UTMOST-501, PU+U.
                         *********************

TITLE : SPHERIC GEOM.
DATE  :  16/09///

                     INPUT DATA (RSYST FORMALISM)
                     ******************************

          ORDER OF SCATTERING             1
          QUADRATURE ORDER                4
          NO. OF ZONES                    5
          NO. OF INTERVALS               40
          NO. OF GROUPS                  22
          EIGENVALUE MODIFIER       -2.000E-01
          PRECISION DESIRED          1.000E-03
          NORM. FACTOR               2.435E+20
          NEW PARM. MOD. SRCH.       1.000E+00

                      DENSITIES(ATOMS*1E-24/CM3)
                      ****************************

                      CORE

    NA     = 7.59E-03    CL     = 1.30E-02    U238  = 4.72E-03
    U233   = 2.83E-04    PU239  = 2.54E-04    PU240 = 7.26E-05
    PU241  = 3.63E-05    FP239  = 5.59E-05

    RATIO U8/(PUTOT + U3) =   7.70

                      FIRST_WALL

    C      = 6.62E-02    FE     = 2.88E-02

                      BLANKET

    NA     = 5.41E-03    CL     = 1.08E-02    TH232 = 5.41E-03
    PA     = 1.30E-05    U233   = 1.08E-05

                      2-ND_WALL

    C      = 1.37E-02    FE     = 7.31E-02

                      REFLECTOR

    C      =  .0         FE     = 8.46E-02
```

Table 2.9

```
                    UTMOST-501, PU+U.
                    *********************

TITLE : SPHERIC GEOM.
DATE  :  16/09/77


*********************************************************************
*                                                                   *
*                     NEUTRONICS MACR                               *
*                     ****************                              *
*                                                                   *
*  ELEMENT         ABSORPTION                  PRODUCTION           *
*                                                                   *
*    CORE                                                           *
*                                                                   *
*  U233         38.077E+18       16.27  89.679E+18          36.80   *
*  U238         71.188E+18       30.42  62.698E+18          25.73   *
*  PU239        27.072E+18       11.82  68.323E+18          28.04   *
*  PU240        33.993E+17        1.45  61.259E+17           2.51   *
*  PU241        47.359E+17        2.02  12.348E+18           5.07   *
*  CL           12.350E+18        5.28    .0                  .00   *
*  FE           23.588E+17        1.01    .0                  .00   *
*  NA           11.275E+17         .48    .0                  .00   *
*  C            13.080E+16         .06    .0                  .00   *
*  FP           48.166E+16         .21    .0                  .00   *
*                                                                   *
*  TOTAL CORE   16.153E+19       69.01  23.917E+19          98.15   *
*                                                                   *
*    BLANKET                                                        *
*                                                                   *
*  TH232        53.378E+18       22.81  18.379E+17            .75   *
*  U233         11.761E+17         .50  26.719E+17           1.10   *
*  PA           73.403E+16         .31    .0                  .00   *
*  CL           65.000E+17        2.78    .0                  .00   *
*  FE           10.450E+18        4.46    .0                  .00   *
*  NA           28.913E+16         .12    .0                  .00   *
*  C            74.329E+13         .00    .0                  .00   *
*                                                                   *
*  TOT. BLAN.   72.528E+18       30.99  45.098E+17           1.85   *
*                                                                   *
*  EN TWO                               44.660E+17           1.80   *
*  LEAKAGE      14.126E+18        5.69                              *
*                                                                   *
*  T O T A L    23.405E+19      100.00  24.364E+19         100.00   *
*                                                                   *
*********************************************************************
```

Table 2.10

```
                         UTMOST-501, PU+U.
                         ********************

  TITLE : SPHERIC GEOM.
  DATE  :  16/09///


*******************************************************************
*                                                                 *
*                  NEUTRONICS MICR                                *
*                  ****************                               *
*       ENTITY                SYMBOL                VALUE         *
*                                                                 *
*    NIC                                            3.182         *
*    NI1                                            3.022         *
*    NI2                                            2.456         *
*    NI3                                            2.554         *
*    NI8                                            2.946         *
*    NI9                                            2.994         *
*    ALFM                                            .136         *
*    FASB                                            .380         *
*    ABPAR                                           .172         *
*    LEAG                                            .058         *
*    PRODUCTION                                     1.426         *
*    FAST BONUS                                      .651         *
*    ABSOR.PARS                                      .152         *
*    LEAKAGE                                         .141         *
*    BRED.RATIO MICR                                1.784         *
*    PU39 PRODUC                                    4.991         *
*    PU39 DESTRO                                    2.767         *
*    U33 PRODUC                                     5.263         *
*    U33 DESTRO                                     3.925         *
*    BALANCE U233             BALA33                1.338         *
*    BALANCE PU239            BALA39                2.223         *
*    TOTAL BREEDING MACR      BRTOT                 1.451         *
*                                                                 *
*  MAX. FLUX IS : 2.58E+15 FOR GROUP 11 IN INTERVAL   1           *
*                                                                 *
*******************************************************************
*COMMENT                                                          *
*FASB, N*SIGMA FISS.FERT / N*SIGMA FISS.FISSIONABLE.              *
*ABPAR, N*SIGMA PARAS / N*CAPT.(FERT + FISS).                     *
*LEAG, LEAKAGE / TOTAL ABSOR.(FROM TABLE NEUTRONICS).             *
*PRODUC, (ZNIM - 1 - ALFM) / (1 + ALF).                           *
*FAST BONUS, (FASB * (NIC - 1)) / (1 + ALF).                      *
*ABSOR.PARS, ABPAR / (1 + ALF).                                   *
*LEAKAGE, LEAG*ZNIM / (1 + ALF).                                  *
*BRTOT, N*SIGMA TRANSMUTATION (TH + U8 + PU0) / N*SIGMA.          *
*CAPT. (U3 + PU9 + PU1).                                          *
*******************************************************************
```

Table 2.11

```
                     UTMOST-501, PU+U.
                     ********************

 TITLE : SPHERIC GEOM.
 DATE  :  16/09/77


*****************************************************
*                                                   *
*      ENTITY           SYMBOL    UNIT       VALUE  *
*                                                   *
*     GEOMETRY                                      *
*                                                   *
*   CORE RADIUS         R(1)      M           .963  *
*   WALL 1 THICKNESS    THICK(2)  M           .040  *
*   BLANKET THICKNESS   THICK(3)  M           .550  *
*   WALL 2 THICKNESS    THICK(4)  M           .040  *
*   HEIGHT OF CYLINDER  HEIGHT    M           .000  *
*   HEIGHT/DIAM. RATIO  HDRAT     -           .000  *
*   VOLUME OF CORE      VOLCOR    M**3       3.740  *
*   VOLUME OF BLANKET   VOLBL     M**3      11.461  *
*                                                   *
*     POWER                                         *
*                                                   *
*   POWER IN CORE       POINC     GW         2.749  *
*   POWER IN BLANKET    POINB     GW          .059  *
*   TOTAL POWER         POWTO     GW         2.808  *
*   POWER DENS. CORE    PODEC     GW/M**3     .735  *
*   POWER DENS. BLANKET PODEB     GW/M**3     .005  *
*   POWER RATING OF PU  POWRAT    MW/KG      5.091  *
*                                                   *
*     INVENTORY                                     *
*                                                   *
*   TOTAL PU239 INVENT. P9TOT     KG       377.326  *
*   TOTAL PU INVENT.    PUTOT     KG       539.940  *
*   U233 IN CORE        U3C       KG       409.894  *
*   U233 IN BLANKET     U3B       KG        47.984  *
*   TOTAL U238 INVENT.  U8TOT     KG      6978.166  *
*   TOTAL TH INVENT.    THTOT     KG     23889.212  *
*   RATIO U38/(PU+U33)  RATU8     -         12.924  *
*                                                   *
*     OTHER DATA                                    *
*                                                   *
*   BURN. RATIO (PU/U)  BURNR     -           .826  *
*   DOUBLING TIME       DTIME     YEARS      1.291  *
*   MEAN FLUX CORE      FLMEAN    NEUT/S*CM2 1.334E+16 *
*                                                   *
*****************************************************
```

Table 2.12

```
***********************************************************************
*                               TABLE                                 *
*                              *******                                *
*           MATERIAL BALANCE OF THE BREEDER SYSTEM                    *
*                   INCLUDING NEW REACTOR                             *
***********************************************************************
*     *    THOR     *    PA     *   U233    *   U238    *   PU39      *
***********************************************************************
*ORS   1.92E+04    6.39E+02    6.30E+02    1.83E+03    6.22E+02*
*SBL   2.39E+04    5.76E+01    4.80E+01                        *
*FBL   9.99E+00    8.95E+00    4.41E+02                        *
*FTR   6.39E+02    6.30E+02                6.22E+02            *
*SCO                           4.10E+02    6.98E+03    3.77E+02*
*FFP                                       2.65E+02    3.46E+02*
*FCB   6.49E+02                            8.87E+02            *
*FLR   2.39E+00    5.76E-03    4.80E-03    6.98E+00    3.77E-01*
*SRE   2.39E+00    5.76E-03    4.10E-01    6.98E+00    3.77E-01*
*FWA   6.49E-01                1.89E-01    6.98E-04    3.77E-02*
*FNW   1.85E+04                1.89E+02    9.44E+02            *
*SNW   2.39E+04                2.44E+ 2    1.22E+03            *
***********************************************************************

WHERE

ORS = FLOW FROM THE ORE OR SYNTH. IN KG / YEAR
SBL = STEADY STATE IN THE BLANKET IN KG.
FBL = FLOW FISS. + PARAS.IN BLANKET IN KG / YEAR
FTR = FLOW FOR TRANSMUTATION IN KG / YEAR
SCO = STEADY STATE IN THE CORE IN K
FFP = FLOW FISS.+ PARASITIC IN CORE IN KG / YEAR
FCB = FLOW TO THE CORE OR BLANKET IN KG / YEAR
FLR = FLOWTO REPROCESS AND RETURN IN KG / YEAR
SRE = STEADY STATE IN THE RE-PROCESSING IN KG
FWA = FLOW TO THE WASTE IN KG / YEAR
FNW = FLOW TO THE NEW IN KG / YEAR
SNW = IN THE NEW AFTER DTIME IN KG
```

- all the freshly bred plutonium can be burned in the same reactor (the history of the transplutonium elements is neglected here).
- to achieve this *in situ* burning of plutonium the reprocessing of the irradiated fuel is limited to the separation of all or just the most neutron absorbing fission products and directly coupled to the reactor
- for the next generation of reactors the fuel is produced in the form of denatured uranium, that is an isotopic mixture of

  10.5% uranium -233 (produced in this breeder)
  89.5% uranium -238 (from the mine or depleted uranium stockpiles)

- for over 90% of the lifetime of this plant, that is ∿30 years, the reactor burns its own plutonium and produces the uranium -233 for the next breeder generation. Tables 2.13 to 2.17 give the corresponding data for this reactor phase as before.

2.1.5 Comparison of the three phases

In spite of this rather over-simplification it seems that the proposed reactor design is suited for burning:

uranium -233 in th first phase
uranium -233 and plutonium in the second phase
plutonium in the steady state phase.
(see Fig. 2.3)

As a first approximation the timetable of such a reactor will be as follows

- doubling time approximately 4 years (including the out of core inventory).
- transient period (going from U-233 of plutonium) approximately 2 years.
- steady state period approx 30 years including shutdown periods for the exchange of the core vessel etc. See Table 2.18

[illegible]

```
                    UTICST-302,NOPU.
                    ********************

TITLE : SPHERIC [illegible]
DATE  :  03/[illegible]

                 (INPUT DATA (FSYST FORMALISM)
                 *****************************

      ORDER OF SCATTERING             [illegible]
      QUADRATURE ORDER                    4
      NO. OF ZONES                        5
      NO. OF INTERVALS                   41
      NO. OF GROUPS                      22
      EIGENVALUE MODIFIER         -2.000E-01
      PRECISION DESIRED            1.000E-03
      NORM. FACTOR                 2.520E+20
      NEW PARM. [illegible]        1.000E+00

                  DENSITIES(ATOMS*E+24/CM3)
                  *************************

                  CORE

    NA    = 7.48E-03     FE    = 1.09E-2     U238  = 4.71E-3
    U233  = 7.36E-05     PU239 = 7.1[illegible]E-4     PU240 = 1.43E-4
    PU241 = 8.17E-05     PU242 = 1.1[illegible]E-4

    RATIO U8/(PUTOT + U3) =   6.79

                  FIRST WALL

    C     = 6.62E-02     FE    = 2.88E- 2

                  BLANKET

    NA    = 5.41E-03     FE    = 1.08E-02    TH232 = 5.41E- 3
    PA    = 1.30E-08     U233  = 1.08E-05

                  SECOND WALL

    C     = 1.37E-02     FE    = 7.31E- 2

                  REFLECTOR

    C     =  .0          FE    = 8.46E-02
```

Table 2.14

```
                        UTHOST-3C2,NOPU.
                        *********************

 TITLE : SPHERIC GEOM.
 DATE  :  03/09/77

**********************************************************************
*                                                                    *
*                    NEUTRONICS MACR                                 *
*                    ****************                                *
*                                                                    *
*  ELEMENT          ABSORPTION               PRODUCTION              *
*                                                                    *
*    CORE                                                            *
*                                                                    *
*  U233          10.599E+16         .04 24.936E+16           .10     *
*  U238          76.015E+18       30.64 65.729E+18         25.45     *
*  PU239         58.061E+18       23.65 14.513E+19         56.19     *
*  PU240         69.111E+17        2.79 12.726E+18          4.93     *
*  PU241         11.388E+18        4.59 20.652E+18         11.48     *
*  CL            13.083E+18        5.27   .0                 .00     *
*  FE            25.411E+17        1.02   .0                 .00     *
*  NA            11.833E+17         .48   .0                 .00     *
*  C             14.230E+16         .06   .0                 .00     *
*  FP            10.387E+17         .42   .0                 .00     *
*                                                                    *
*  TOTAL CORE    17.107E+19       68.96 25.349E+19         98.14     *
*                                                                    *
*    BLANKET                                                         *
*                                                                    *
*  TH232         57.083E+18       23.01 19.305E+17           .75     *
*  U233          12.680E+17         .51 28.730E+17          1.11     *
*  PA            75.191E+16         .30   .0                 .00     *
*  CL            66.744E+17        2.69   .0                 .00     *
*  FE            10.910E+18        4.40   .0                 .00     *
*  NA            30.440E+16         .12   .0                 .00     *
*  C             77.050E+13         .00   .0                 .00     *
*                                                                    *
*  TOT. BLAN.    76.992E+18       31.04 48.036E+17          1.86     *
*                                                                    *
*  ENTWO                                45.919E+17          1.78     *
*  LEAKAGE       14.056E+18        5.65                              *
*                                                                    *
*  T O T A L     24.806E+19      100.00 25.808E+19        100.00     *
*                                                                    *
**********************************************************************
```

Table 2.15

```
                              UTMOST-302,NOPU.
                              *****************

 TITLE : SPHERIC GEOM.
 DATE  :  03/09/77


*****************************************************************
*                                                               *
*                     NEUTRONICS MICR                           *
*                     ***************                           *
*       ENTITY                SYMBOL               VALUE        *
*                                                               *
*     NI0                                          3.182        *
*     NI1                                          3.020        *
*     NI2                                          2.454        *
*     NI3                                          2.515        *
*     NI8                                          2.947        *
*     NI9                                          2.993        *
*     ALFM                                          .200        *
*     FASB                                          .455        *
*     ABPAR                                         .173        *
*     LEAG                                          .058        *
*     PRODUCTION                                   1.501        *
*     FAST BONUS                                    .738        *
*     ABSOR.PARS                                    .144        *
*     LEAKAGE                                       .144        *
*     BRED.RATIO MICR                              1.951        *
*     PU39 PRODUC                                  5.371        *
*     PU39 DESTRO                                  5.866        *
*     U33 PRODUC                                   5.630        *
*     U33 DESTRO                                    .137        *
*     BALANCE U233            BALA33               5.492        *
*     BALANCE PU239           BALA39               -.495        *
*     TOTAL BREEDING MACR     BRTOT                1.581        *
*                                                               *
*   MAX. FLUX IS : 2.81E+15 FOR GROUP 11 IN INTERVAL  1         *
*                                                               *
*****************************************************************
*COMMENT                                                        *
*FASB, N*SIGMA FISS.FERT / N*SIGMA FISS.FISSIONABLE.            *
*ABPAR, N*SIGMA PAPAS / N*CAPT.(FERT + FISS).                   *
*LEAG, LEAKAGE / TOTAL ABSOR.(FROM TABLE NEUTRONICS).           *
*PRODUC, (ZNIM - 1 - ALFM) / (1 + ALF).                         *
*FAST BONUS, (FASB * (NI8 - 1)) / (1 + ALF).                    *
*ABSOR.PARS, ABPAR / (1 + ALF).                                 *
*LEAKAGE, LEAG*ZNIM / (1 + ALF).                                *
*BRTOT, N*SIGMA TRANSMUTATION (TH + U8 + PU0) / N*SIGMA.        *
*CAPT. (U3 + PU9 + PU1).                                        *
*****************************************************************
```

Table 2.16

```
                    UTMOST-302,NOPU.
                    ********************

   TITLE : SPHERIC GEOM.
   DATE  :  03/09/77

***************************************************
*                                                 *
*       ENTITY          SYMBOL   UNIT      VALUE  *
*                                                 *
*     GEOMETRY                                    *
*                                                 *
*   CORE RADIUS        R(1)      M          .954  *
*   WALL 1 THICKNESS   THICK(2)  M          .040  *
*   BLANKET THICKNESS  THICK(3)  M          .550  *
*   WALL 2 THICKNESS   THICK(4)  M          .040  *
*   HEIGHT OF CYLINDER HEIGHT    M          .000  *
*   HEIGHT/DIAM. RATIO HDRAT     -          .000  *
*   VOLUME OF CORE     VOLCOR    M**3      3.633  *
*   VOLUME OF BLANKET  VOLBL     M**3     11.299  *
*                                                 *
*     POWER                                       *
*                                                 *
*   POWER IN CORE      POINC     GW        2.732  *
*   POWER IN BLANKET   POINB     GW         .063  *
*   TOTAL POWER        POWTO     GW        2.795  *
*   POWER DENS. CORE   PODEC     GW/M**3    .752  *
*   POWER DENS. BLANKET PODEB    GW/M**3    .006  *
*   POWER RATING OF PU POWRAT    MW/KG     2.570  *
*                                                 *
*     INVENTORY                                   *
*                                                 *
*   TOTAL PU239 INVENT. P9TOT    KG      736.542  *
*   TOTAL PU INVENT.   PUTOT     KG     1063.152  *
*   U233 IN CORE       U3C       KG        1.035  *
*   U233 IN BLANKET    U3B       KG       47.307  *
*   TOTAL U238 INVENT. U8TOT     KG     6763.900  *
*   TOTAL TH INVENT.   THTOT     KG    23551.789  *
*   RATIO U38/(PU+U33) RATU8     -         6.362  *
*                                                 *
*     OTHER DATA                                  *
*                                                 *
*   BURN. RATIO (PU/U) BURNR     -        50.983  *
*   DOUBLING TIME      DTIME     YEARS     1.984  *
*   MEAN FLUX CORE     FLMEAN    NEUT/S*CM2 1.460E+16 *
*                                                 *
***************************************************
```

Table 2.17

```
***********************************************************
*                            TABLE                        *
*                           *******                       *
*          MATERIAL BALANCE OF THE BREEDER SYSTEM         *
*                  INCLUDING NEW REACTOR                  *
***********************************************************
*   *   THOR    *    PA     *   U233    *   U238    *   PU39    *
***********************************************************
*ORS   1.26E+04   6.83E+02   6.74E+02   4.24E+03   6.49E+02*
*SHL   2.36E+04   5.68E+01   4.73E+01                      *
*FHL   9.55E+00   9.17E+00   1.51E+01                      *
*FTR   6.83E+02   6.74E+02              6.69E+02           *
*SCO                         1.03E+00   6.76E+03   7.37E+02*
*FFP                                    2.78E+02   7.34E+02*
*FCB   6.94E+02                         9.47E+02           *
*FLR   2.36E+00   5.68E-03   4.73E-03   6.76E+00   7.37E-01*
*SRE   2.36E+00   5.68E-03   1.03E-03   6.76E+00   7.37E-01*
*FWA   6.94E-01              6.58E-01   6.76E-04   7.37E-02*
*FNW   1.19E+04              6.58E+02   3.29E+03           *
*SNW   2.36E+04              1.31E+03   6.53E+03           *
***********************************************************

WHERE

ORS = FLOW FROM THE ORE OR SYNTH. IN KG / YEAR
SHL = STEADY STATE IN THE BLANKET IN KG.
FHL = FLOW FISS. + PARAS.IN BLANKET IN KG / YEAR
FTR = FLOW FOR TRANSMUTATION IN KG / YEAR
SCO = STEADY STATE IN THE CORE IN KG
FFP = FLOW FISS.+ PARASITIC IN CORE IN KG / YEAR
FCB = FLOW TO THE CORE OR BLANKET IN KG / YEAR
FLR = FLOWTO REPROCESS AND RETURN IN KG / YEAR
SRE = STEADY STATE IN THE RE-PROCESSING IN KG
FWA = FLOW TO THE WASTE IN KG / YEAR
FNW = FLOW TO THE NEW IN KG / YEAR
SNW = IN THE NEW AFTER DTIME IN KG
```

Fig. 2.3 Fissile atom concentration in the core

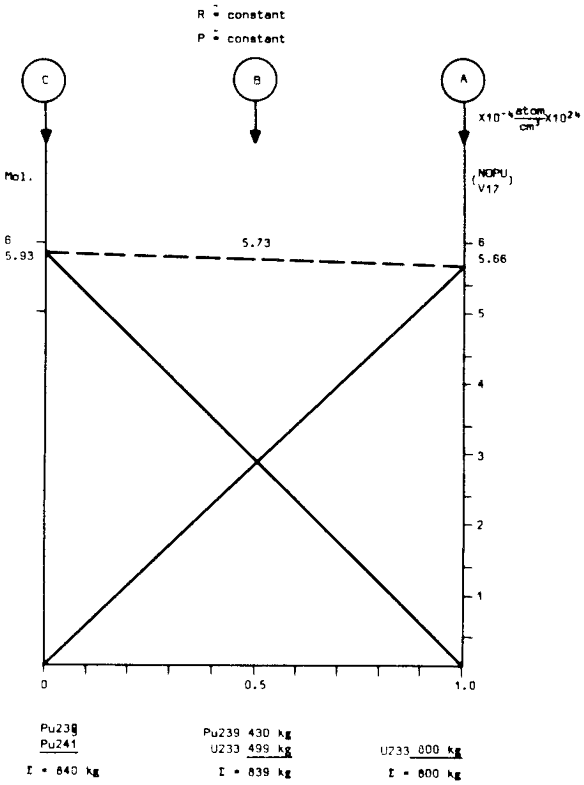


Fig. 2.4 Thorium and uranium-238 breeding
(NOPU-21)

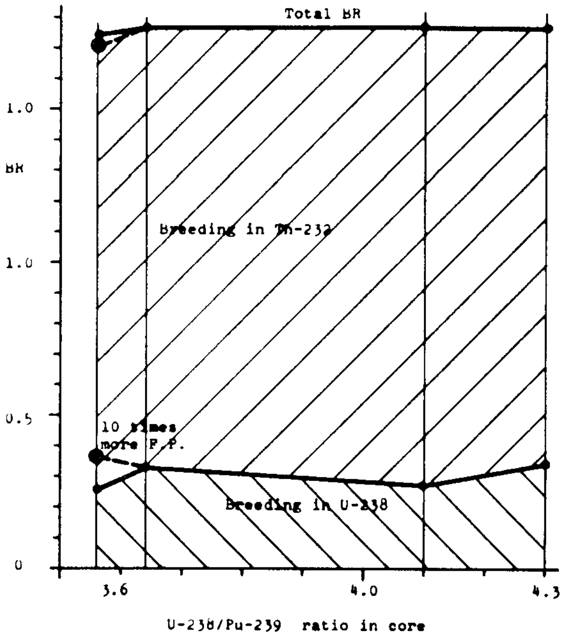


Table 2.18 Core Diameter = 0.95 m
Power = 2.8 GW(th)

		Build-up	Transient	Steady State
		A	B	C
Fissile	-	U233	U233, Pu239 + 241	Pu239 + 241
Core inventory	kg	800	409, 430	840
Fertile	-	U238	U238	U238
Core inventory	kg	6864	6978	6763
$\frac{\text{Fissile}}{\text{Fertile}}$ (enrichment)	%	10.4	10.7	11.0
Breeding ratio total	BR_{tot}	1.34	1.45	1.58
Mean neutron flux	$10^{16}\ \frac{\text{neutrons}}{\text{cm}^2\text{s}}$	1.26	1.33	1.46
Power density	$\frac{\text{GW(therm)}}{\text{m}^3\ \text{core}}$	0.75	0.745	0.75

It should be emphasised that the era when fusion reactors are replaced by other energy sources and hence the plutonium burners are shut-down, has not been dealt with.

Even here it is conceivable that a reactor design could be proposed which only consumes and thus be used in this shutdown phase. This however has not been calculated.

2.2 Impact of some parameters on the mixed zone two zone fast breeder

In this section some intermediate results are presented concerning the influence of selected parameters in this design, and giving the breeding capability of a two zone fast breeder reactor. (Table 2.19)

The influence of mutual displacement.

$$\Sigma(\mathrm{Pu}\ -239) + \Sigma(\mathrm{U}-233) = 9 \times 10^{-4}\ (10^{24}\mathrm{atom.\ cm}^{-3})$$

by five steps from A to E is given in Table 2.20 and Fig. 2.5 and 2.6.

The influence of the thickness of the external blanket, which contains thorium, on the total breeding ratio and the volume of the core for a given case of the following concentrations of fissile nuclides

Pu-239 3.5×10^{-4} (10^{24}atom. cm^{-3})

U-233 10.0×10^{-4} (10^{24}atom. cm^{-3})

is given in Table 2.20.

For the same case the partial BR's and BR total are shown in Fig. 2.7 and Fig. 2.8

In addition the following 4 reactor designs have been calculated having the following arrangement

Core Pu-239 $1.1 \times 10^{4} \longrightarrow 1.8 \times 10^{4}$ ($\times 10^{24}$atom. cm^{-3})

U-233 $3.4 \times 10^{-4} \longleftarrow 5.5 \times 10^{-4}$ ($\times 10^{24}$atom. cm^{-3})

U-233/Pu-239 = 3.1

Blanket thickness 100 cm.

The data are given in Tab. 2.21. Other results are found in Fig. 2.9, 2.10 and 2.11.

Table 2.19

OBJECT: Plutonium burning *in situ*: some parameters.

REACTOR TYPE : Power, Breeder, Two zones
GEOMETRY: INTERNAL ZONE : Spherical core, ∿ 65 cm
WALL : ∿ 4 cm
INTERMEDIATE ZONE : -
WALL : ∿ 4 cm
EXTERNAL ZONE : Blanket: 100 cm
WALL, REFLECTOR : Iron,....

POWER (GW thermal) : 3 GW
POWER DENSITY (GW therm/m^3CORE) :
NEUTRON FLUX, MEAN (n/cm^2s) : 2

FISSILE NUCLIDE : Core - Pu239
: Out blanket - U-233
FERTILE NUCLIDE : Core - U238 Out, Blanket Th232
DILUTENT : Chlorides
COOLING SYSTEM : External to core
BREEDING RATIO :

PARAMETER STUDIED : Ratio U238/PU239 in core
: Ratio PU239/U233
: FP Concentration
: Wall (Molybdenum) thickness
: Blanket thickness

METHOD OF NEUTRONIC :
CALCULATION : see 1.10 (page 17)

Table 2.20 Mixture of Pu-238 and U-233 in the core

Pu/U3 Ratio	A Pu only	B Pu > U3	C Pu ≈ U3	D Pu < U3	E U3 only
Fission/sec	2.74×10^{20}	2.6×10^{20}	2.51×10^{20}	2.41×10^{20}	2.32×10^{20}
Pu-239 } core U-233 $\times 10^{24}$ at/cm^3)	9.10^{-4} 0.0	6.10^{-4} 3.10^{-4}	4.10^{-4} 5.10^{-4}	2.10^{-4} 7.10^{-4}	0.0 0.10^{-4}
BR total	1.761	1.64	1.56	1.49	1.42
Partial BR U/Pu	0.353	0.312	0.289	0.271	0.259
Partial BR Th/U-233	1.408	1.33	1.27	1.21	1.16
Flux tot. (ncm^{-2}s^{-1})	2.05×10^{16}	1.99×10^{16}	1.94×10^{16}	1.86×10^{16}	1.76×10^{16}
Radius, (cm)	67.28	66.22	65.72	65.5	65.8
Volume, (cm)	1.28	1.22	1.19	1.18	1.19
Spec. power (GW m^{-3})	2.34	2.46	2.52	2.54	2.52
Ratio of partial breeding ratios	3.99	4.26	4.39	4.46	4.48
Fission ratios $\frac{\text{U-233}}{\text{Pu-239}}$	0	0.89	2.28	5.40	$\propto$

Fig. 2.5 **Thorium and uranium-238 breeding (NOPU-22)**

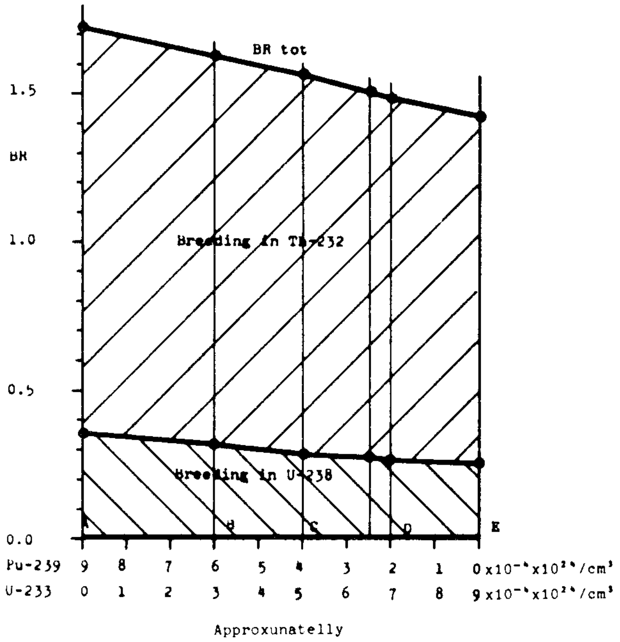


Fig. 2.6 Ratio of breeding ratios (NOPU-22)

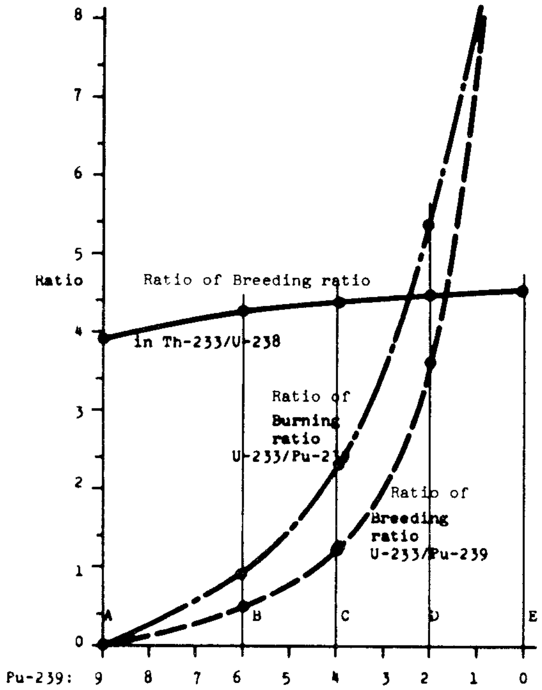


Fig. 2.7 **Radius of core versus radius of blanket (NOPU-23)**

$\left.\begin{array}{l}\text{Pu} = 3.5\times10^{-4}\\ \text{U33} = 20.0\times10^{-4}\end{array}\right\}\times10^{24}$atom/cm^3

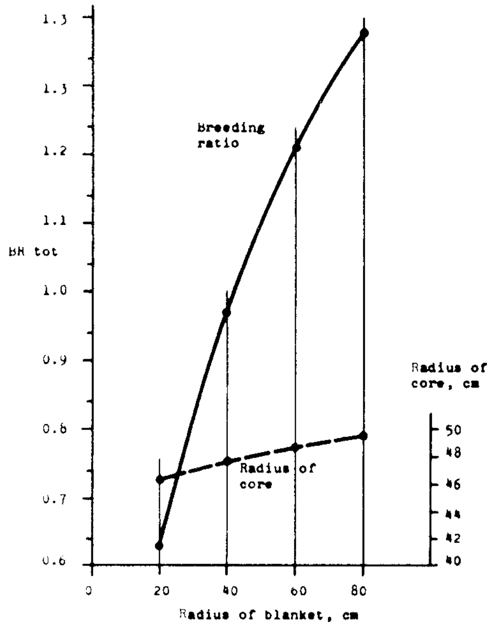


Fig. 2.8 **Partial breeding ratio (NOPU-23)**

$\left.\begin{array}{l}\text{Pu} = 3.5\times10^{-4}\\ \text{U33} = 10.0\times10^{-4}\end{array}\right\}\times10^{24}$atom/cm^3

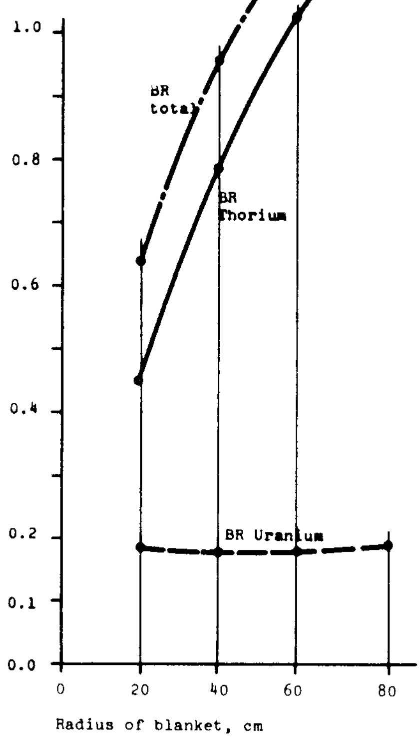


Table 2.21 Influence of Pu/U33 ratio

Parameters	A	B	C	D
U-238	1.0	0.85	0.85	0.85
Pu-239 $\times 10^{24}$ atom/cm^3	1.8×10^{-4}	1.5×10^{-4}	1.3×10^{-4}	1.1×10^{-4}
Pu-240	2.5×10^{-5}	2.1×10^{-5}	1.8×10^{-5}	1.55×10^{-5}
U-233	5.5×10^{-4}	4.7×10^{-4}	4.0×10^{-4}	3.4×10^{-4}
U3/Pu9 ratio	3.06	3.13	3.08	3.09
BR total	1.51	1.51	1.52	1.528
Partial BR thorium	1.12	1.04	0.95	0.852(see fig.2.9)
Partial BR uranium	0.39	0.47	0.564	0.677
$\frac{Th}{U}$ breeding ratio	2.37	2.21	1.68	1.26(see fig.2.10)
Flux tot (ncm^{-2}s^{-1})	1.3×10^{16}	0.92×10^{16}	0.62×10^{16}	0.463×10^{16}
Radius of core, cm	78.6	91.4	107.7	122.0
Volume of core, m^3	2.03	3.2	5.23	7.61
Spec. power (GW m^{-3})	1.48	0.938	0.574	0.39
Thickness of blanket m	1.0	1.0	1.0	1.0
Volume of blanket m^3	80.6	93.4	109.7	124.0
Pu-39 in core, atoms	3.65×10^{26}	4.8×10^{26}	6.80×10^{26}	8.37×10^{26}
mol	609	800	1130	1400
kg	146	191	271	333
U.33 in core, atoms	1.12×10^{27}	1.5×10^{27}	2.09×10^{27}	3.59×10^{27}
mol	1860	2510	3490	4310
kg	434	584	812	1000
Th in blanket, atoms	1.01×10^{29}	1.21×10^{29}	1.49×10^{29}	1.86×10^{29}
mol	169000	202000	248000	293000
kg	39200	46800	57600	68000
U-238 in core, atoms	9.54×10^{27}	1.5×10^{28}	2.46×10^{28}	3.58×10^{28}
mol	15900	25100	41000	596000
kg	3780	5970	9750	14200
$\frac{Th-32}{U-8}$ inventory ratio	10.6	8.1	6.0	4.9
Burning rate				
Pu-239 in 10^{17}atom/s	1.44	1.39	1.39	1.36
U-233 in 10^{17}atom/s	6.08	6.11	6.08	6.09
Ratio $\frac{U3}{Pu}$ burning		4.36		

Fig. 2.9 **Partial breeding ratio**
(NOPU-24)

Concentration ratio $\frac{U\text{-}233}{Pu\text{-}239} \simeq 3.1$

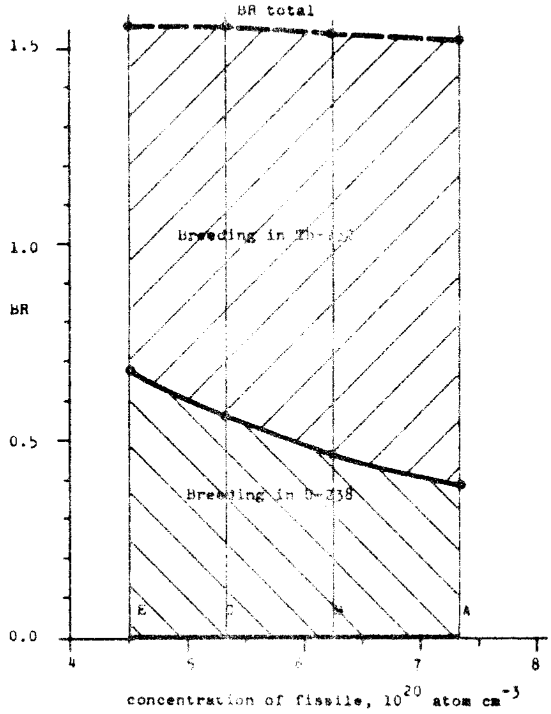


Fig. 2.10 **Ratio of burning rate and ratio of breeding rate**

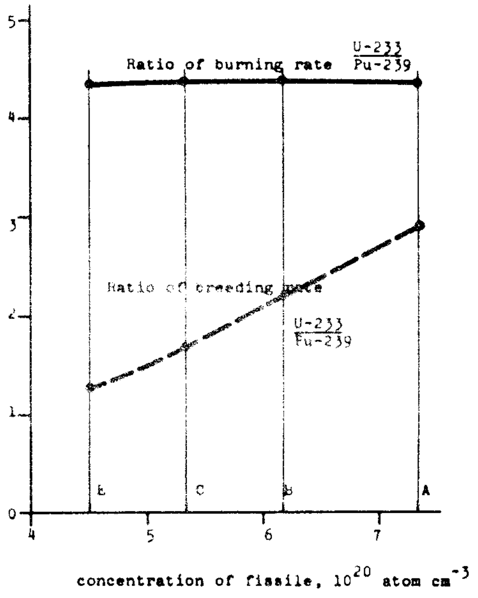
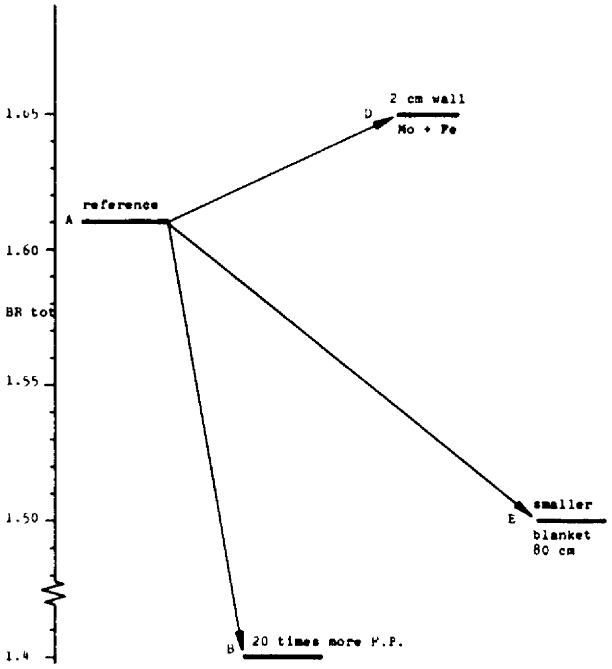


Fig. 2.11 Impact of some selected parameters (NOPU-26 reference)



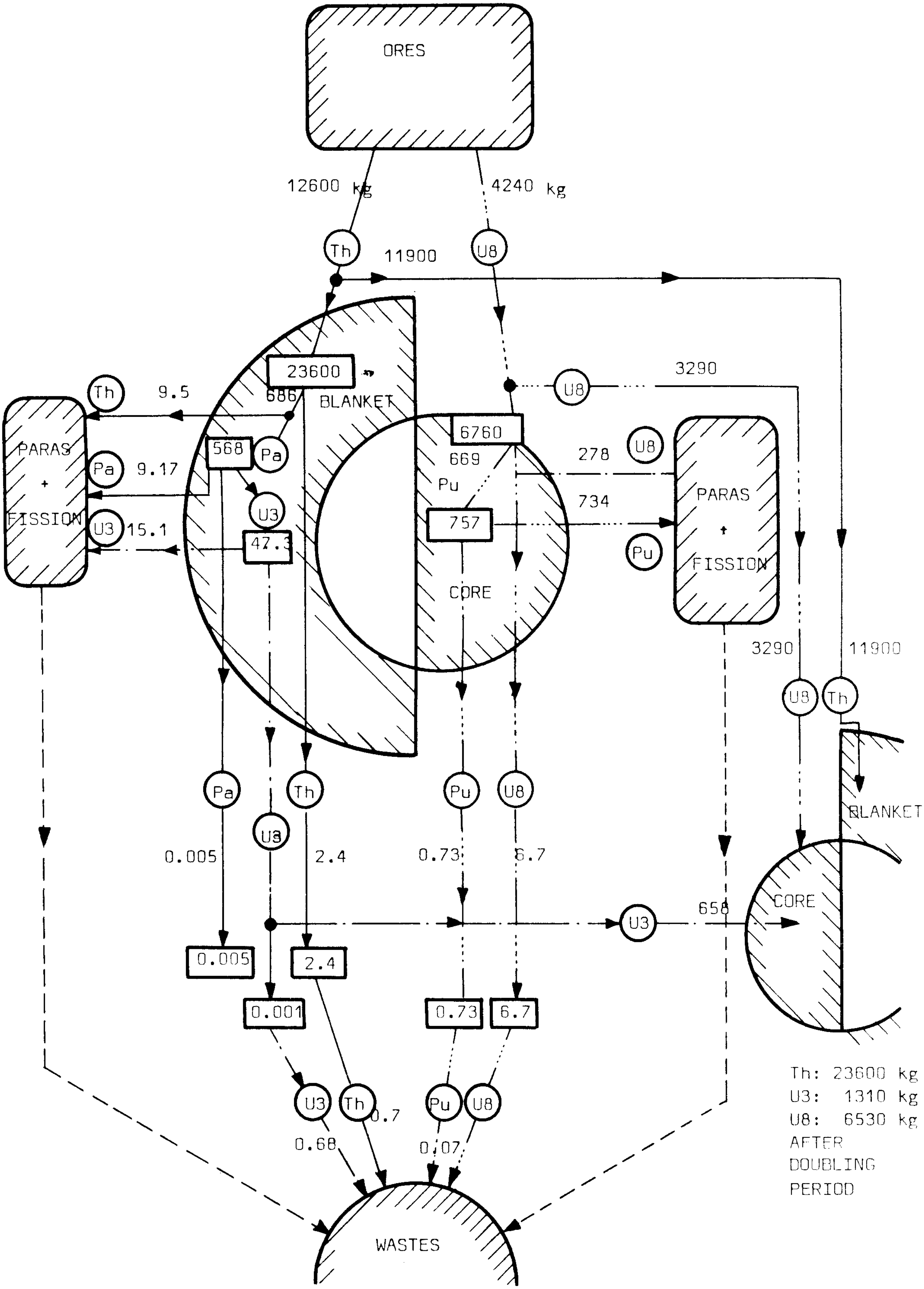
2.3 Material balance of the state reactor

The most important feature of this reactor is the *in situ* consumption of plutonium. The studies show this to be feasable. Fig. 2.12 gives the resulting material balance for the steady state reactor.

2.4 Conclusions

The molten chlorides fast breeder is ideally suited to the *in-situ* burning of plutonium matching the current requirement for a "proliferation safe" concept.

Fig. 2.12



Th: 23600 kg
U3: 1310 kg
U8: 6530 kg
AFTER
DOUBLING
PERIOD

3.1 THE THREE ZONE REACTOR

3.1.1 Introduction

The reactor discussed now is rather unconventional because of its three zones (see Fig. 1.2)

- internal blanket zone
- fuel zone
- external blanket zone

This concept has been compared with the more conventional type. Holding most of the parameters the same the breeding gain comes out about equal. However, one large difference is that although the total power is the same the specific power changes by more than one magnitude being higher in the conventional central fuel zone reactor. Also the mean neutron flux increases from 1.2×10^{16}n cm^{-2}s^{-1} for the non-conventional central blanket zone to 2×10^{17}n cm^{-2}s^{-1} for the conventional central fuel zone.

Since the specific power and intensity of neutron flux is clearly a major problem form the point of view of the engineering design of the reactor (cooling, radiation damage of structural material and fuel), both systems have been studied, that is without a central blanket region and with a radius up to 110 cm. The results are given below for fuel without uranium and with uranium fuel for both cases: no internal blanket zone and with an internal blanket zone.

3.1.2 The three zone breeder with thorium/uranium-233

The first case is the fast breeder molten salt reactor with uranium-233 as fissile and thorium-232 as fertile material with a three region layout.

- internal fertile zone	1, 50, 90, 110	cm
- wall	2	cm
- spherical shell core	19	cm
- wall	3	cm
- external fertile zone	110	cm
- reflector	40	cm

Table 3.1 gives the main details of the reactor.

3.2 A Three zone breeder reactor with a mixed fuel cycle U-238/Pu-239 plus Th-232/U-233

The next step is a study of a mixed fuel cycle in the three zone fast breeder reactor. This problem has received much attention and the published papers given in list D should be referred to.

A three zone fast breeder reactor with the geometry shown in table 3.1 has been calculated.

The range of variations covered include

Pu-239 or U-233 in the core
U-238 Th-232 in the blanket

For a given geometry and concentration of ertile and fissile nuclides the influence of the fission products, when the concentrations are increased by a factor 10 results in a reduction in the breeding ratio by 5%. An increase by a factor 10 in proactinium decreases the breeding ratio by only 2% (Fig. 3.4).

For the three zone reactor calculations have also been made for mixed fuel cycles. For the fissile materials Pu-239 and U-233 and for fertile materials with U-238 in the core only and Th-232 in the blanket only.

Table 3.1

OBJECT: Three zones thorium cycle

REACTOR TYPE : Power, Breeder
GEOMETRY : INTERNAL ZONE : Spherical, fertile material
(M) WALL : Metallic
INTERMEDIATE ZONE : Shell; fissile material, active core
WALL : Metallic
EXTERNAL ZONE : Shell, fertile material
WALL, REFLECTOR : Metallic

POWER (GW thermal) : 2.6
POWER DENSITY (GW therm/m^3core) : 0.78 - 9.52
NEUTRON FLUX, MEAN (n/cm^2s) : $\sim 10^{16}$

FISSILE NUCLIDE : Internal Blanket
: Core - Pu + U233
Outer Blanket U233
FERTILE NUCLIDE : Internal blanket Core U238
Outer Blanket - Th 232
DILUTENT : Chlorides
COOLING SYSTEM : External
BREEDING RATIO : 1.09 - 1.14

PARAMETER STUDIED : Thickness of blanket
: Thorium concentration in balnket material
FP and PA concentration
:
:

METHOD OF NEUTRONIC : ANISN, S4
23 Groups
CALCULATION : 80 spatial positions
P_1 approxim.
: GGC-3 code
ENDF/B-1 and B-2 Data

Table 3.2 (THOC-300)

Three zone reactor. Volumes and breeding ratio

Geometry:	internal blanket, radius	110 cm
	wall, width	3 cm
	core, width	27 cm
	wall	3 cm
	external blanket, width	100 cm
	wall reflector	140 cm

Variable				Results		
Case	Internal fertile (cm)	Internal wall (cm)	U-233 concen. in core ($10^{24}/cm^3$)	Volume of core (m^3)	Specific power (GW m^{-3})	Breeding ratio, total
B	1.0	1.0	0.0012	0.783	3.36	1.09
C	50.0	2.0	0.0012	1.66	1.57	1.10
D	90.0	3.0	0.0012	3.73	0.70	1.13
E	110	3.0	0.0012	5.62	0.46	1.11
A		3.0				
reference	110		0.0018	3.13	0.80	1.16
F	110	3	0.0008	9.52	0.28	1.14

see Fig. 3.1

(THOCL-300)

Three zone reactor: atomic composition (atomic concentration × 10^{24})

Internal fertile zone	Wall	Core	Wall	External blanket	Reflector
Th 4.5×10^{-3} Pa 1×10^{-4} U-33 1×10^{-4} Cl 2.2×10^{-2} Na 4.5×10^{-3} variable radius (see Fig. 3.1)	Fe 7×10^{-2} Mo 1×10^{-2}	Th 2.5×10^{-3} Pa 1×10^{-4} U-33 1×10^{-3} U-34 1×10^{-4} variable U-233 concentration (see Table above).	Fe 7×10^{-2} Mo 7×10^{-2}	the same as internal blanket	Fe 7×10^{-2} Mo 1×10^{-2} (remark: wrong reflector with Mo!)

Fig. 3.1 Impact of the radius of the internal blanket (THCL-300) on the breeding ratio

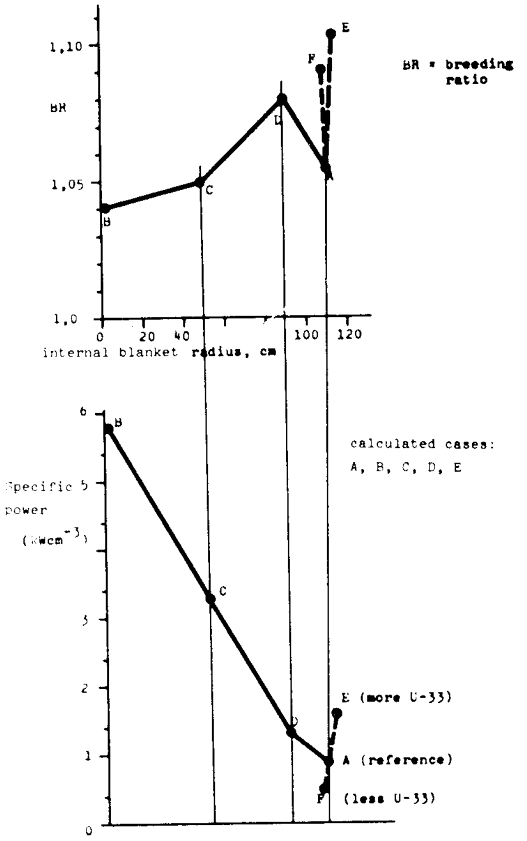


Fig. 3.2 Concentration of thorium and uranium-233 (in 10^{24}/cm^3) (THOCL-301)

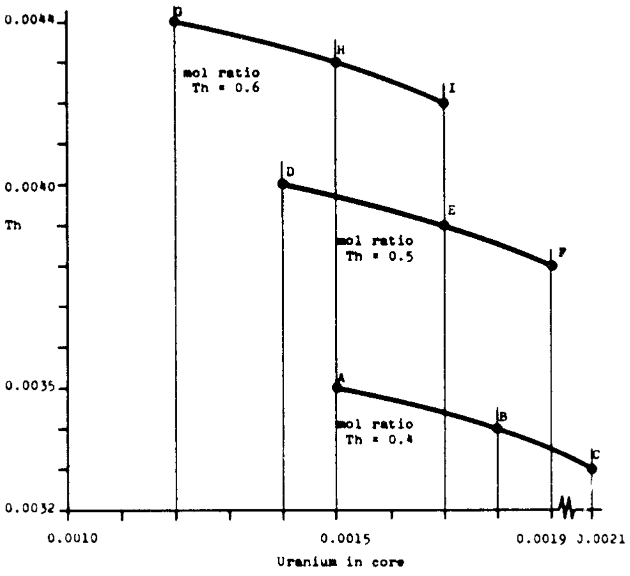


Fig. 3.3 Uranium concentrations for given geometry (THOCL-301) for A, B, C, D, E, F, G, H, I,

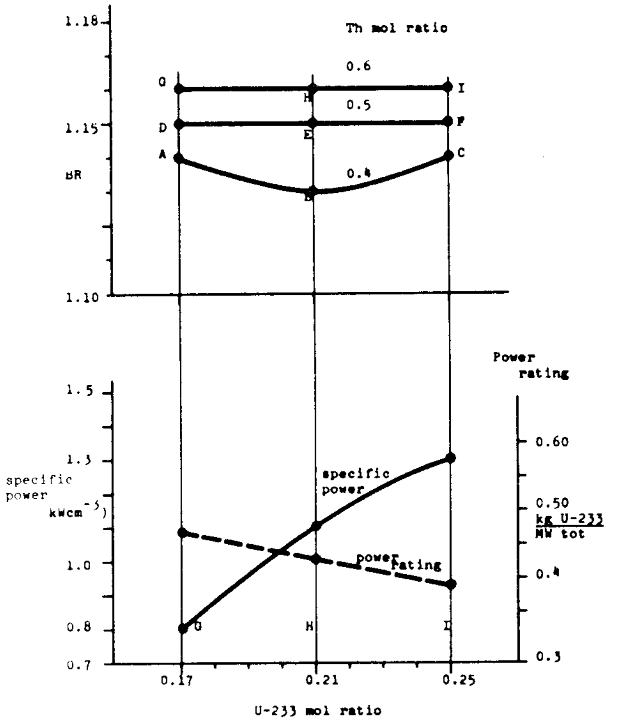


Fig. 3.4 Three zones, mixed fuel cycle
Impact of fission products and protactinium
(THOCL-305)

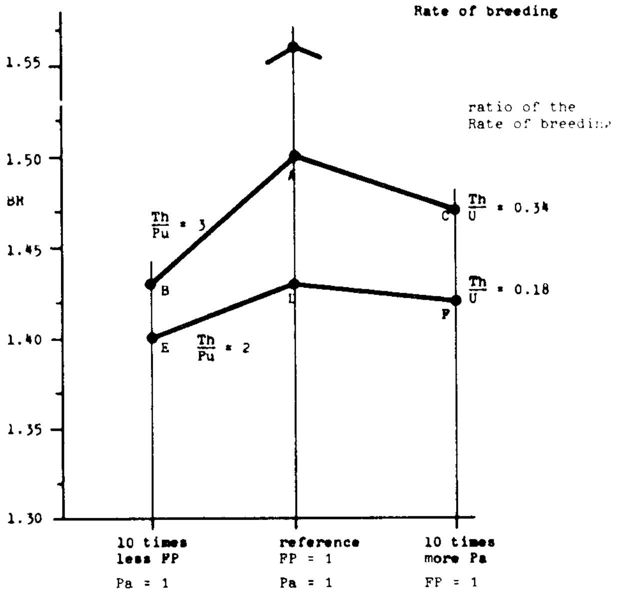


Table 3.3 (THOCL-302)

Influence of FP and Pa concentration

geometry	internal blanket	wall	core	wall	external balnket	reflector
	110 cm	3 cm	19 cm	3 cm	100 cm	40 cm

Case	Fission product content $\times 10^{24}$ cm^{-3}	Protactinium concentration corresponds to dwelling time, days.			Breeding ratio
		2	6	18	
A	3×10^{-5}, dwelling time = 3 days	1×10^{-5}			1.119
B			3×10^{-5}		1.124
C				9×10^{-5}	1.123
D	9×10^{-5}, dwelling time = 10 days	1×10^{-5}			1.1238

see also Fig. 3.4

Remark Table 3.4 canceled

The influence of parameter variations is given in Fig. 3.5. The following results can be noted:

the breeding ratio increases (under the given conditions) when

- the concentration of U-233 in the core decreases and U-238 increases
- when the internal wall thickness is halved (note this is very sensitive due to the presence of 15 atom% of molybdenum)
- when the outer fertile radius increases
- when the inner fertile zone radius increases.

3.3 The three zone reactor - uranium-plutonium fuel cycle

The reference reactor is described in table 3.5 and 3.6. The thermal flux in all three zones, the external breeding zone, the fuel and the internal breeding zone is only 10^{-8} of the total flux and in the external blanket reaches 10^{-6} of the total flux. The total flux has a relatively flat distribution and even in the fuel region the max. to mean ratio is only about 1.13 (Fig. 3.6 and Fig. 3.7).

The neutron flux is rather hard and the mean neutron energy (calculated as the mean of the no. of fissions) is around 370 keV (see Fig. 3.8). In a typical LMFBR and in a gas cooled fast breeder this value is 120 keV and 176 keV respectively.

A good illustration of the influence of the most important papameters on the breeding ratio is given in table 3.7. The differences between these calculations and the computur output is approx 8%.

Fig. 3.5 Three zones; mixed fuel cycle
Impact of selected parameters on the breeding ratio
(NOPU-1)

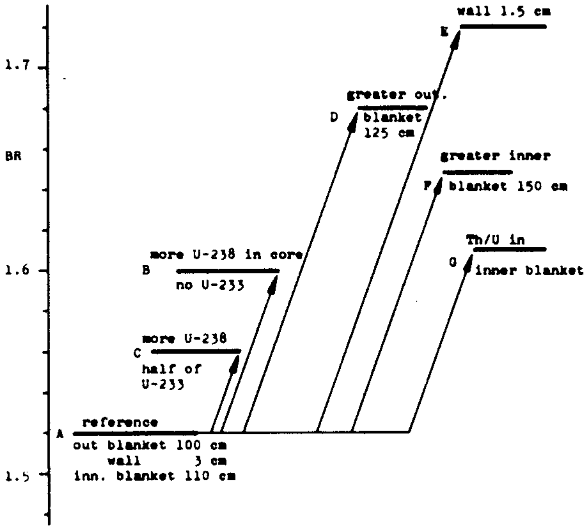


Fig. 3.6 Three zones: uranium-plutonium fuel cycle
Neutron Flux in reference reactor

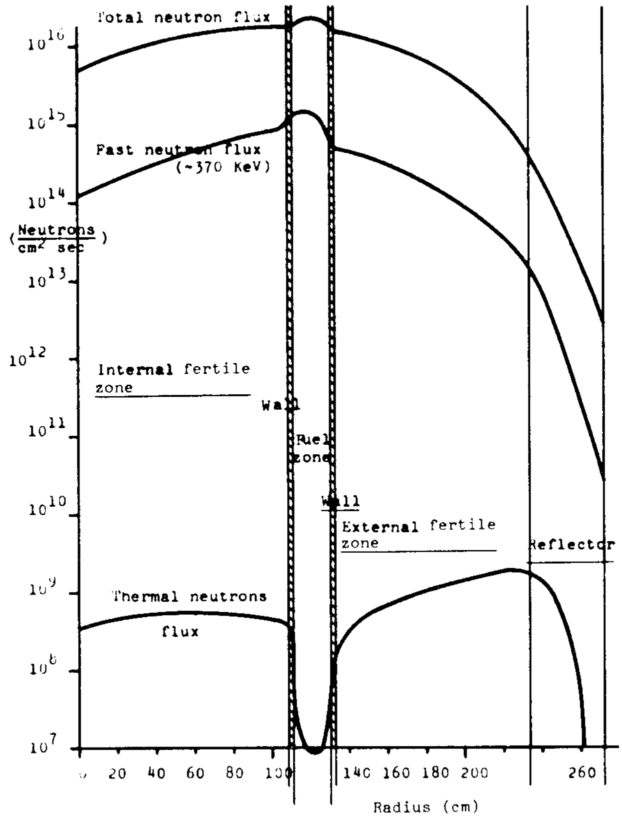


Table 3.5

OBJECT : Uranium-plutonium cycle - Maximum breeding ratio in three zones

REACTOR TYPE : Optimised breeder
GEOMETRY : INTERNAL ZONE : Spherical fertile zone
(M) WALL : Iron, Molybdenum
INTERMEDIATE ZONE : Core, Fuel
WALL : Iron, Molybdenum
EXTERNAL ZONE : Fertile zone
WALL, REFLECTOR : Iron

POWER (GW thermal) : ∿6
POWER DENSITY (GW therm/m^3 CORE) : 1.1
NEUTRON FLUX, MEAN (n/cm^{2}s) : ∿10^{16} - 10^{17}

FISSILE NUCLIDE : Internal Blanket: small amounts of Pu239, U238
: Core Pu 239, Pu240, Pu241, 0.7: /.2: 0.1:
External Blanket: small amounts of Pu239, U238
FERTILE NUCLIDE : Internal Blanket U238
DILUTENT : Chlorides, Sodium
COOLING SYSTEM : Outer
BREEDING RATIO : 1.64 - 1.95

PARAMETER STUDIED : Plutonium-uranium ratio
: With and without uranium
: Reflector, Fe, Pb
:

MEHTOD OF NEUTRONIC : see chapter 1
CALCULATION :
:

Table 3.6 (200/C)

Three Zones reactor: uranium-plutonium fuel cycle.

Radius cm	Width of zone cm	Zone	Composition atoms/10^{24}cm^3	Flux $\frac{\text{thermal;}}{\text{total,}}$ Breeding ratio	Specific power GW/m^3, Temperature:
0	110.0	I Central fertile zone	U-238 6.4×10^{-3} Pu-239 6.0×10^{-5} F.P. 2.0×10^{-5} Na 3.4×10^{-3} Cl 2.27×10^{-3}	$\frac{1.05\times10^{16}}{3.7\times10^{8}}$ BR 0.490	$T_{inlet}=700^{0}C$ $T_{outlet}=800^{0}C$
110.0	3.0	II Wall	Fe 7×10^{-2} Mo 1×10^{-2}	$\frac{1.15\times10^{16}}{9\times10^{7}}$	$850^{0}C$
113.0	17.9	III Fuel zone	Pu-239 1.3×10^{-3} Pu-240 4.2×10^{-4} Pu-241 2.1×10^{-4} U-238 4.2×10^{-3} F.P. 2.0×10^{-5} Na 3.4×10^{-3} Cl 2.6×10^{-2}	$\frac{1.02\times10^{16}}{6.6\times10^{7}}$ BR 0.22	1.1GW/m^3 $T_{inlet}=750^{0}C$ $T_{outlet}=1050^{0}C$
130.9	3.0	IV Wall	Fe 7.0×10^{-2} Mo 1.0×10^{-2}	$\frac{8.24\times10^{15}}{2.4\times10^{8}}$	$850^{0}C$
133.9	100.0	V External fertile zone	the same as central fertile zone, I	$\frac{3.9\times10^{14}}{1.9\times10^{9}}$ BR: 1.040	$T_{inlet}=700^{0}C$ $T_{outlet}=800^{0}C$
233.9	40	VI	Fe 8.0×10^{-2}	$\frac{5.2\times10^{12}}{5\times10^{4}}$	
273.9					

Fig. 3.7 Three zones: uranium-plutonium fuel cycle
Total flux in the fuel zone

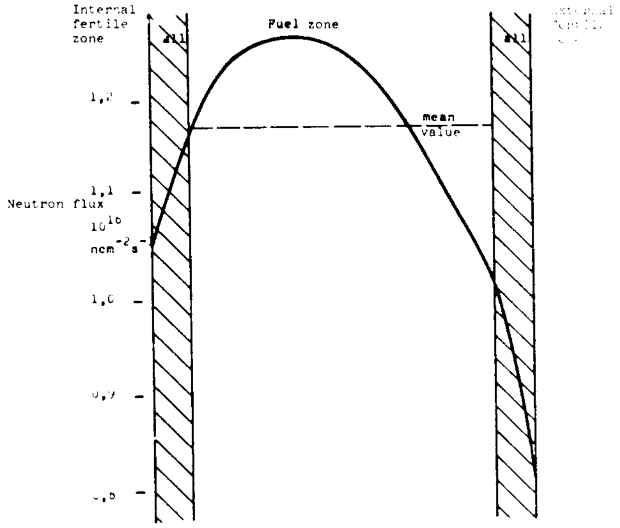


Fig. 3.8 Three zones: uranium-plutonium fuel
Spectrum in fuel zone

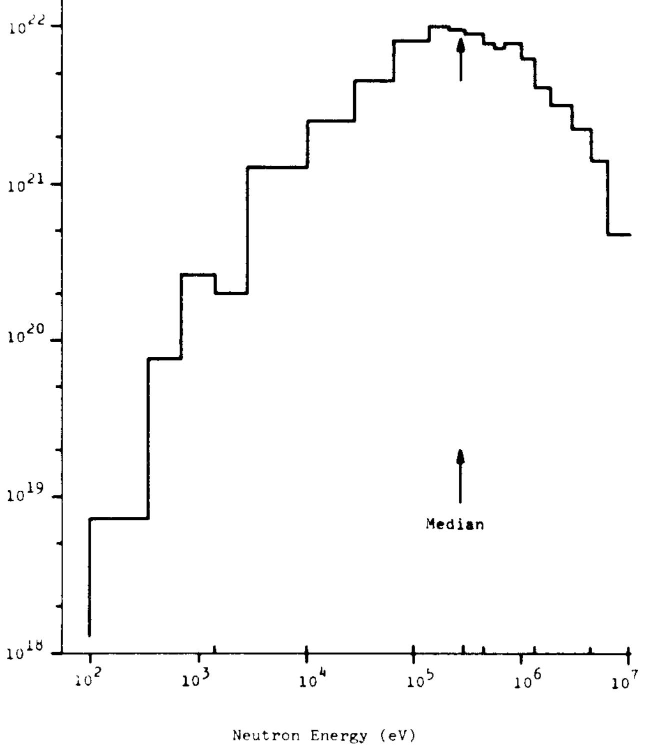


Table 3.7 Three Zone reactor: uranium-plutonium fuel

Simplified calculation of the breeding ratio and neutron balance.

	Parameter		Component of breeding ratio
Median energy	keV	370	
Pu-239	σ_f(barn)	1.83	
(from computer output)	σ_c(barn)	0.180	
	ν	2.95	
	α	0.0984	
Breeding potential	$(\eta-1)$	1.6857	1.685
Fission ratio fertile/fissile	δ	0.37	
Fast bonus	$\frac{(\nu'-1)}{1+\alpha}$	0.539	0.539
Total positive			2.225
Losses (absorption in FP, Cl, Na, Mo, Fe)		0.160	
Leackage (arbitrarily)		0.10	
Total losses	$\frac{\text{Loss}+\alpha}{1+\alpha}$	0.32	0.320
Calculated BR (micro)			1.890
Computed BR (macro)			1.752

3.4 The three zone breeder reactor: Very high breeding gain

One of the most important factors in achieving a very high breeding ratio is the hardness of the neutron flux. This is strongly influenced by the fuel composition.

In this case the fuel is postulated to be a mixture of

$a\ PuCl_3 \cdot b\ NaCl \cdot c\ UCL_3$ where

$a = 0.1 - 0.2, \quad b = 0.7 - 0.8, \quad c = 0.1 - 0.2$

Unfortunately not all data are available for this system. (see Fig. 6.9)

The rough calculations on changing the concentration of $PuCl_3$ in the melt of NaCl (Fig. 3.9) shows a rather sharp decrease of breeding gain (BG) for decreasing plutonium concentration, especially when the plutonium molar ratio to sodium is lower than 0.25.

In spite of these uncertainties of the $PuCl_3$-NaCl-UCl_3 system, the influence of the U-238 in the fuel has also been calculated. For a constant $PuCl_3$ concentration with a simplified assumption for the NaCl concentration the results are given in Fig. 3.10 and 3.11.

Increasing the ratio of uranium to plutonium in the fuel from 0 to 3 causes the total breeding gain to increase from 0.65 to 0.95. This is rather clear and thus the reference reactor concept includes uranium in the fuel in a ratio of 2 : 1 to plutonium.

Such a high breeding gain is a special feature of this type of reactor for producing large quantities of fissile material.

Fig. 3.12 and 3.13 give the results of calculation when the radius of the central fertile zone is varied. Table 3.8 shows the simplified calculation of central and external fertile zone breeding ratios.

Fig. 3.9 Impact of Plutonium Concentration

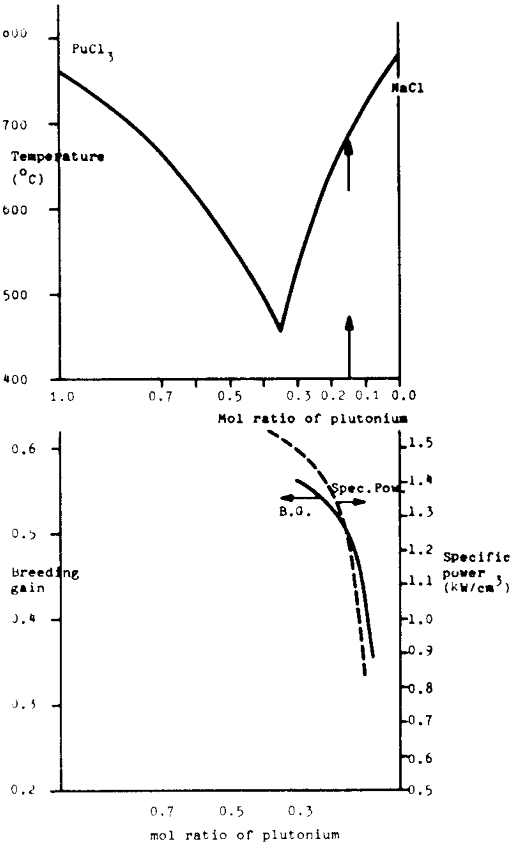


Fig. 3.10 Three zones: uranium-plutonium fuel cycle
Impact of U-238 concentration in the fuel
Pu-Concentration: 0.0021×10^{24} atom/cm^3

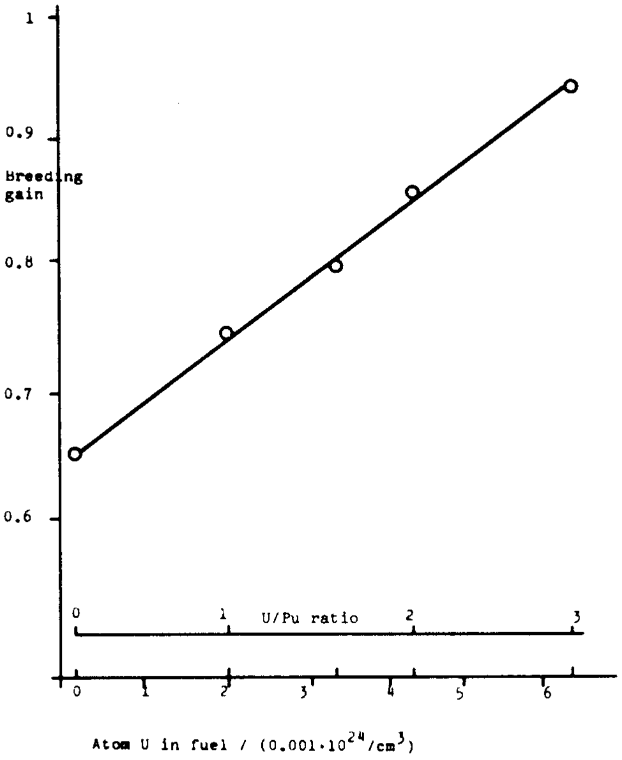


Fig. 3.11 Three zones: uranium-plutonium fuel cycle
Impact of Uranium concentration in the fuel
(202/201) versus central fertile zone

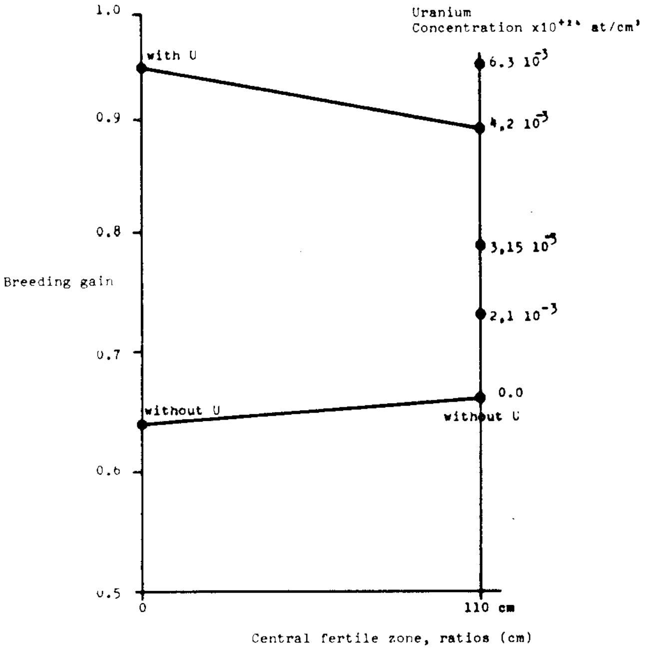
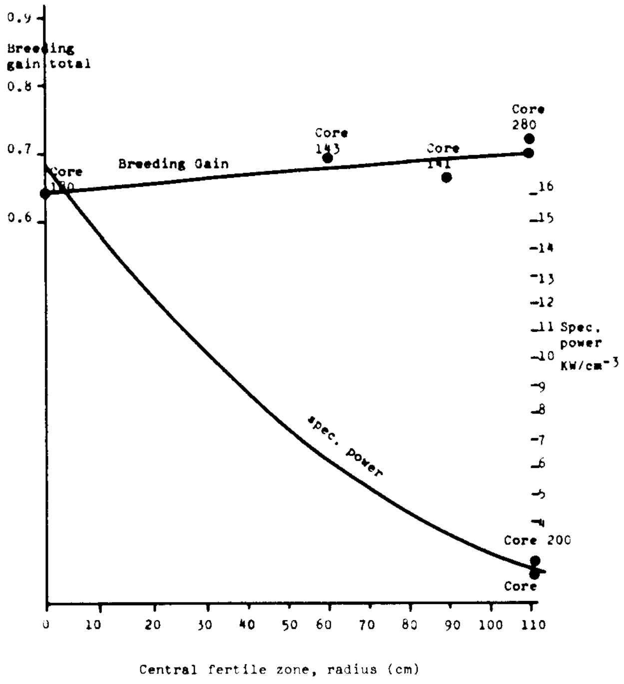


Fig. 3.12 Three zones: uranium-plutonium fuel gele
Impact of central fertile zone radius
(No U in fuel)



From these results the following conclusions can be drawn

- increasing the raius of the internal fertile zone up to 110 cm increases the breeding gain for a given type of fuel. The effect of wall and fertile material changes are insignificant
- at the same time the specific power decreases dramatically
- increasing the U/Pu ratio from 2 to 3.6 does not influence the total breeding gain (see Fig. 3.13)
- the FP concentrations play a rather significant role (Fig. 3.14)

Table 3.8

	Internal fertile zone			Fuel zone		External fertile zone			Total breeding ratio
Case (Number)	Pu-239 $\sigma \times \nu$	U_{fiss} $\sigma \times \nu$	U_{cap}	Pu-239_f $\sigma \times \nu$	Pu-241_f $\sigma \times \nu$	Pu-239_f $\sigma \times \nu$	U_{fiss} $\sigma \times \nu$	U_{cap}	
three zones (200)	0.14	0.308	0.8	3.05	0.351	0.30	0.47	1.83	1.70
two zones (180)	-			3.63	0.367	0.04	0.46	2.50	1.63

The influence of the 40 cm reflector if changed from iron to lead is not very great as shown in table 3.9

Fission product concentration however plays a very important role. For a given reactor design and given fuel and fertile compositions, increasing the concentration of fission products (simulated here with Cs-133 only) from 2×10^{-5} to 2×10^{-4} (in $10^{24}/cm^3$) decreases the breeding gain from 0.65 to 0.38 when the specific power decreases less than a factor two.

Table 3.9 Three zone reactor: uranium-plutonium fuel

Central fuel (Core 180)
(wall 2.5 cm Pu = 2.1×10^{-3} x atom/10^{24}cm^3)

Case	A	B	C
Uranium 238 in fuel	no	yes 4.2×10^{-3} $\times10^{24}$atom/cm^3	no
Reflector 40 cm: material	Fe	Fe	Pb
Volume fuel x 10^5cm^3	2.95	2.40	2.97
Spec. power in fuel kW/cm^3	18.4	23.0	18.3
Breeding ratio total	1.64	1.94	1.66
Total flux of fuel zone n/cm^{2}s	1.18×10^{17}	1.25×10^{17}	1.187×10^{17}

Fig. 3.13 Three zones: uranium-plutonium fuel
Breeding ratio versus radius of internal fertile zone

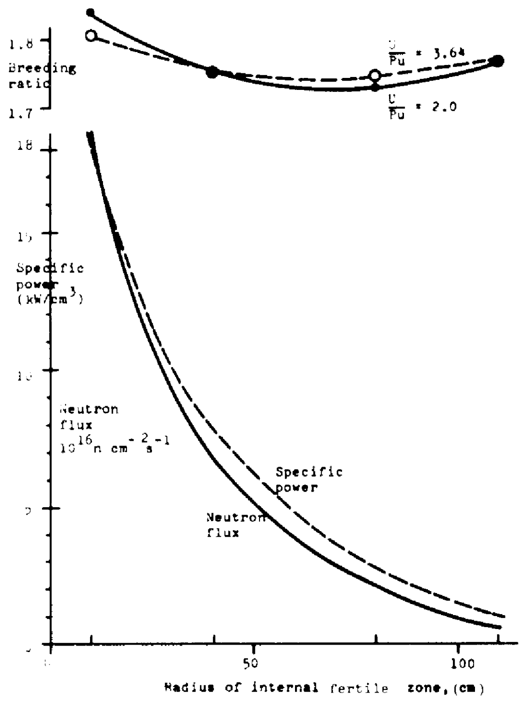
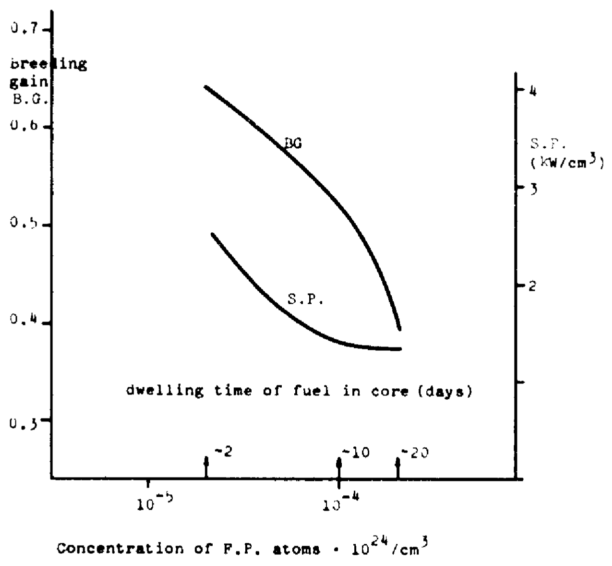


Fig. 3.14 Three zones: uranium-plutonium fuel gele
Impact of Fission Products Concentration in fuel
(very simplified, from different calculations)



In the steady state reactor a concentration of 2×10^{-5} atoms F.P. $\times 10^{24}/\text{cm}^3$ for a fuel having 2.1×10^{-3} atoms $\text{Pu} \times 10^{24}/\text{cm}^3$ is reached for a specific power of 2 GW/m³ after a time t of

$$t = \frac{(2\times 10^{-5}) \times 10^{24}}{(2\times 10^{3})\ (3.1\times 10^{10}) \times 2} = 1.61 \times 10^{5} \text{ s}$$

that is after 1.87 days. The higher fission product concentration - that is 2×10^{-4} corresponds to 18.7 days of mean dwell time f or the fuel in the reactor.

The influence of chlorine-37 separation may now be looked at. The influence of each absorber on the breeding ratio is given by

$$B = \frac{A + D + L + \alpha}{1 + \alpha}$$

B = decrement of breeding ratio

A = absorbtion rate in a given absorber

D = absorbtion rate in rest of absorbers

L = leakage

$\alpha = \sigma_c / \sigma_f$

It can be postulated that for a strong absorber in a hard (fast) spectrum that

$A = 0.15$

$D + L = 0.15$

$\alpha = 0.15$

The relative influence on the rather high breeding ratio of 1.6 results in a case where the profit of the separation factor will be for example 0.9 then

$$\Delta B \ \frac{0.9 \times 0.15}{1.15} = 0.12$$

and in relation to the breeding gain

$$\Delta G \ \frac{0.12}{0.6} = 0.20$$

which results in an increase in doubling time of

$$\frac{T_2}{T_2} = \frac{G}{G+\Delta G} = 0.83$$

It can be seen that introducing three zones does not result in any significant increase in the breeding gain (table 3.10). Therefore the two zone reactor must be preferred for its simpler layout.

Table 3.10 Fuel in central zone (2 zone reactor) versus fuel in middle zone (3 zone reactor)

Core, Case Number		Two zones (conventional) (100)	Three zones (nonconventional) (200)
Geometry	Central Zone	Fuel 100 cm	Blanket 110 cm
	Middle Zone	---	Fuel ~18 cm
	Outer Zone	Blanket 100 cm	
Thermal power, GW		6	6
Pu/FP ratio		$2.1\times10^{-3}/2\times10^{-5}$	$2.1\times10^{-3}/2\times10^{-5}$
Spec. power, kW/cm^3		17.7	1.41
Power in fuel, %		90.9%	76.2%
Flux total left } in fuel right	boundary	$\frac{2.04\times10^{17}}{1.15\times10^{17}}$	$\frac{1.2\times10^{16}}{1.08\times10^{16}}$
Flux in left } outer blanket	boundary	8.99×10^{16}	9.7×10^{15}
blanket right		2.16×10^{15}	1.5×10^{14}
Breeding gain		0.63	0.70
Median energy (group)		~9	10

3.5 The two zone fast breeder. Fuel of uranium plutonium fluorides

3.5.1 Introduction

The aim of this section is to give a rough idea of a fast breeder power reactor having the fuel in form of plutonium trifluoride in the molten state instead of molten chloride. (Table 3.11)
The earier suggestions for a reactor of this type came from A.M. Weinberg.

The first attempts at carrying out calculations on a reactor of this type were not successful because a fuel was chosen having a high concentration of light metals, lithium and beryllium.

A very rough attempt by J. Ligou and the author (1972) shows the possibility for a fast breeder reactor with molten plutonium fluoride where the light metals were eliminated and the melting point increased.

P. Faugeras (Fautenay aux Roses) claimed that the fluoride of U-233 and Th-232 can be used for non-thermal reactors.

Some preliminary results for a three zones reactor are given in a short form in Table 3.12. The neutron flux remains rather hard (Fig. 3.15).

3.5.2 Arbitrary assumptions and uncertainties

The fuel composition has been arbitrarily chosen since the appropriate data is lacking in the literature. In most cases the following fuel composition has been used

$1\ PuF_3\ /\ 1.2\ NaF$ $\quad (T_{melt} \cong 727^{\circ}C)$
$+\ 2.4\ NaF\ /\ 1\ Zr\,F_4$ $\quad (T_{melt} \cong 510^{\circ}C)$

another alternative would be

$PuF_3\ /\ 2\ NaF$
$CaF_2\ /\ 3\ NaF$ $\quad (T_{melt} \cong 820^{\circ}C)$

Table 3.11

OBJECT : Fast Breeder, Molten Fluoride, Three zones

REACTOR TYPE	:	Power
GEOMETRY : (M)	INTERNAL ZONE =	Fertile zone
	WALL	Iron, Graphite
	INTERMEDIATE ZONE :	Fuel, fluoride
	WALL :	Iron, Mo, Graphite
	EXTERNAL ZONE :	Fertile zone
	WALL, REFLECTROR :	Iron

POWER (GW thermal) : 6
POWER DENSITY (GW therm/m^{3}CORE) : 3.5
NEUTRON FLUX, MEAN (n/cm^{2}s) : 10^{16}

FISSILE NUCLIDE : Plutonium. Fuel composition PuF_3, NaF, ZrF_4
:
FERTILE NUCLIDE : U-238
DILUTENT : NaF, ZrF_4
COOLING SYSTEM : External
BREEDING RATIO : 1.38 (up to 1.51 with higher specific power)

PARAMETER STUDIED : Wall structural material: graphite
: Wall: beryllium, Iron, thickness
: Fission product concentration
:

METHOD ON NEUTRONIC CALCULATION : FLUORIDE+ recalculated from Hansen-Roach
: Fission product only as Cs-133

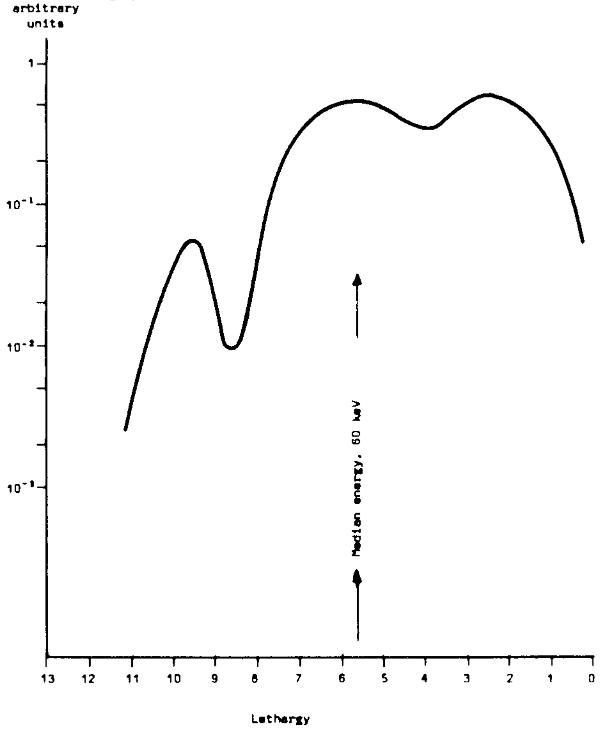
ANISN
Regions 6
Meshes 110
Order of quadrature S_4
Anistropy P_1
23 neutron groups incl. thermal ENDF B/III

Table 3.12 Design of fast breeder molten fluoride reactor
(Three zone: 6 GW(thermal))

Power rating, total = 1 kg Pu/MWth, Doubling time = 6.5 years.

Radius cm	Zone		Components molecules per $cm^3 \times 10^{24}$		Flux $\frac{\text{total}}{\text{thermal}}$	Breeding ratio
0	I	Internal Blanket liquid state	UF_4 NaF PuF_3	6×10^{-3} 6×10^{-3} 6×10^{-5}	$\frac{1.8\times10^{16}}{5.8\times10^{12}}$ T_{mean} = 800°C	0.42
80.0	II	Wall	Fe Mo (graphite is also possible)	7×10^{-2} 1×19^{-2}	$\frac{1.86\times10^{16}}{2.2\times10^{12}}$	--
81.0	III	Fuel liquid state	Pu-239 F_3 Pu-240 F_3 Pu-241 F_3 NaF ZrF_4 F.P. (Cs133)	1.47×10^{-3} 4.2×10^{-4} 2.1×10^{-4} 7.5×10^{-3} 5.1×10^{-3} 0.2×10^{-3}	$\frac{1.57\times10^{16}}{1.00\times10^{16}}$ T_{inlet} = 750°C T_{out} = 1050°C	fertile material Pu-240 0.356
98.2	IV	Wall	Fe Mo (graphite is also possible)	7×10^{-2} 1×10^{-2}	$\frac{1.37\times10^{16}}{5.2\times10^{12}}$	--
100.2	V	External blanket liquid state	UF_4 NaF PuF_3	6×10^{-3} 6×10^{-3} 6×10^{-5}	$\frac{8.8\times10^{13}}{2.5\times10^{11}}$ T_{mean} = 800°C	0.889
200.0	VI	Reflector			$\frac{3.5\times10^{11}}{5.3\times10^{6}}$	--
240.0					Total	BR = 1.38

Fig. 3.15 Neutron spectrum in molten fluoride fast breeder (roughly)



The results obtained, in spite of the uncertainties are encouraging. The simplified breeding ratio calculation gives a BR_{tot} of 1.51 and the computed value is BR_{tot} = 1.465 (table 3.13).

Table 3.14a shows that the influence of fission products is very significant (see also Fig. 3.16). However the change of structural material of the wall form beryllium to iron has only a small effect on the breeding ratio and specific power (table 3.14b and Fig. 3.17).

In the calculations for three zones studies here, increasing the radius of the internal fertile zone from 60 cm to 80 cm, that is a volume increase of 2.37 has little effect on the total breeding ratio in spite of changes in the regional breeding ratios (table 3.15a and Fig. 3.18).

Altering the small amounts of plutonium in the fertile material as a result of reprocessing efficiences also has only a small effect on the breeding ratio and specific power (table 3.15b and Fig. 3.19).

Becouse of the good experience of American and French groups using graphite as a structural material for molten fluordie thermal breeders, calculations have been made using graphite for separating walls for fast breeders. Graphite 2 cm thick as the wall material was chosen.

The results are rather encouraging. The breeding ratio using graphite is still very high, even slightly higher and the specific power in the fuel is lower (table 3.16a and Fig. 3.20).

Changing from the complicated design of three zones with fuel in the intermediate shell, to the "classical" two zone design having fuel in the central region results in a dramatic increase in specific power to a prohibitive 26 $kW.cm^{-3}$ (table 3.16b, Fig. 3.21, table 3.17).

Table 3.13 Simplified calculation of the breeding ratio

Median energy (from computer calculation)	keV	60
Pu-239	σ_f (barn)	1.459
	σ_c	0.2328
	ν	2.899
	α	0.18
	η	2.4568
	$\eta - 1$	1.4568
U-238 (arbitrary)	ν'	2.69
Fast fission of ertile component (from computer output)	δ	0.25
Fast bonus	$\frac{(\nu'-1) \times \delta}{1+\alpha}$	0.34
Total positive	Σ positive	1.79
Losses by absorption (from computer)	L_{ab}	0.1
Leakage (assumption)	L_{leack}	0.05
Total losses	L_{tot}	0.15
Total negative	$\frac{L_{tot+\alpha}}{1+\alpha}$	0.28
Breeding ratio = total positive - total negative	BR (micro)	1.510
Breeding ratio (from computer output)	BR (macro)	1.465

Table 3.14a Three zones reactor: fluoride fuelled fast breeder reactor

Influence of Fission Products.

BR = Breeding ratio, total

Core Number	Wall fuel/external blanket 2 cm	Fission products (Cs-133) atoms/cm^3 x 10^{24}	B.R.	Specific power GW/m^3
163	Be	0.0001	1.437	4.33
164	Be	0.0002	1.427	4.24
166	Be	0.0040	1.14	2.32
167	Fe	0.0040	1.12	2.12

(see also Fig. 3.16)

Table 3.14b Three zones reactor: fluoride fuelled fast breeder reactor

Influence of the beryllium-moderator

F.P. = 0.0002 atom x 10^{24}/cm^3

Core Number	Material 2 cm	Material 1.5 cm	B.R.	Specific power GW/m^3
164	Be		1.427	4.24
162		Be	1.464	4.19
165	Fe		1.38	3.45
161		Ce	1.37	3.47

(see also Fig. 3.17)

Fig. 3.16 Impact of fission products

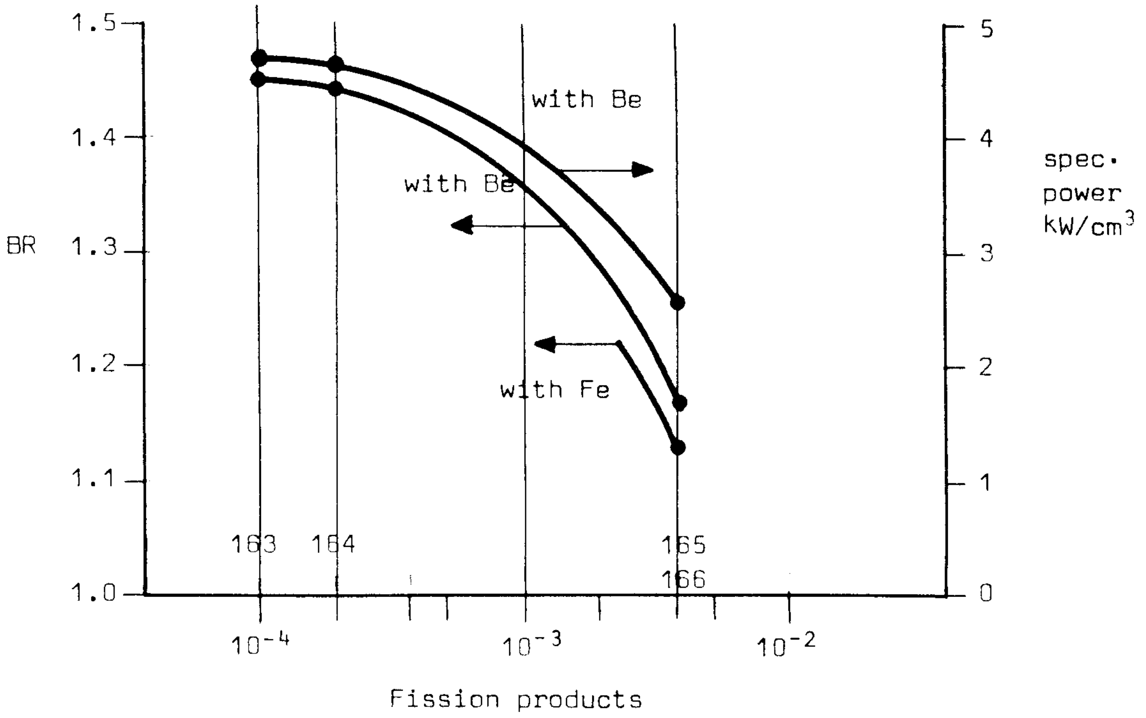


Fig. 3.17 Impact of the beryllium and iron moderator on BR and spec. power

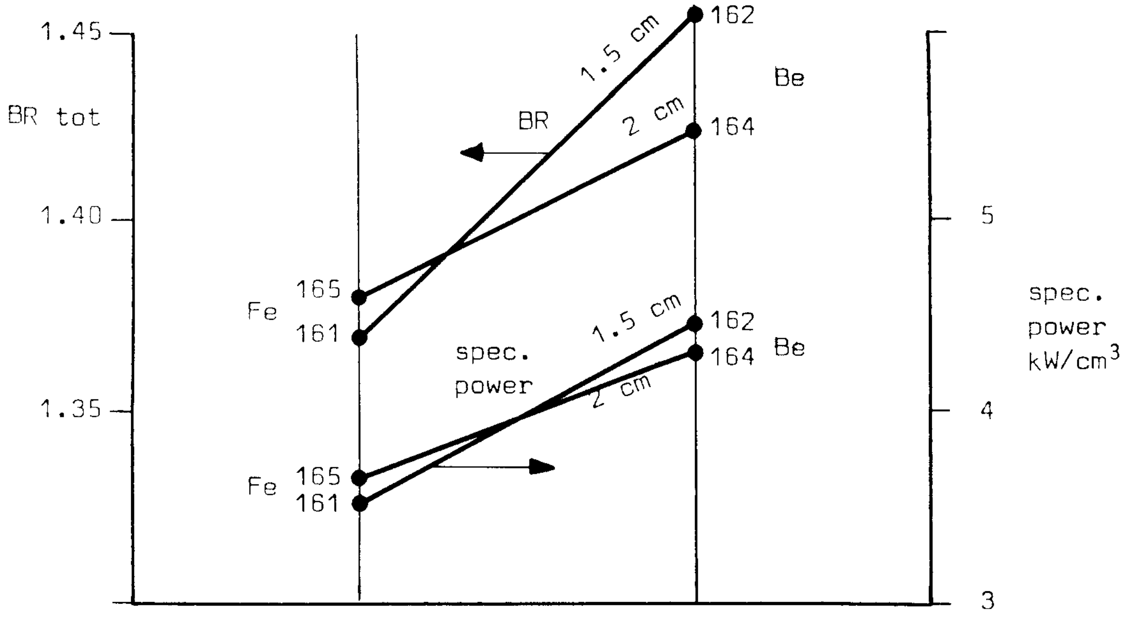


Table 3.15a Three zone reactor: fluoride uranium-plutonium fuel

Influence of the radius of the internal fertile zone

Number of Core	Radius of internal fertile zone, cm	Breeding ratio			
		internal fertile zone	fuel	external fertile zone	total
165	80	0.42	0.056	0.889	1.364
169	60	0.34	0.053	0.99	1.379
Ratio of cores $\frac{165}{169}$	Ratio of volume 2.37	1.21	1.05	0.898	0.989

(see Fig. 3.18)

Table 3.15b Three zone reactor: fluoride uranium-plutonium fuel

Influence of Pu-239 in the fertile material

Number of core	Pu-239 in fertile material	F.P. concentration	B.R. tot	Spec. power GW/m^3
168	0.001	2×10^4	1.49	3.79
161	0.01	2×10^{-5}	1.37	3.471
Ratio of cores $\frac{168}{161}$	8.1	10	1.09	1.09

(see Fig. 3.19)

Fig. 3.18 Three zones fluoride uranium-plutonium fuel
Impact of radius of internal blanket

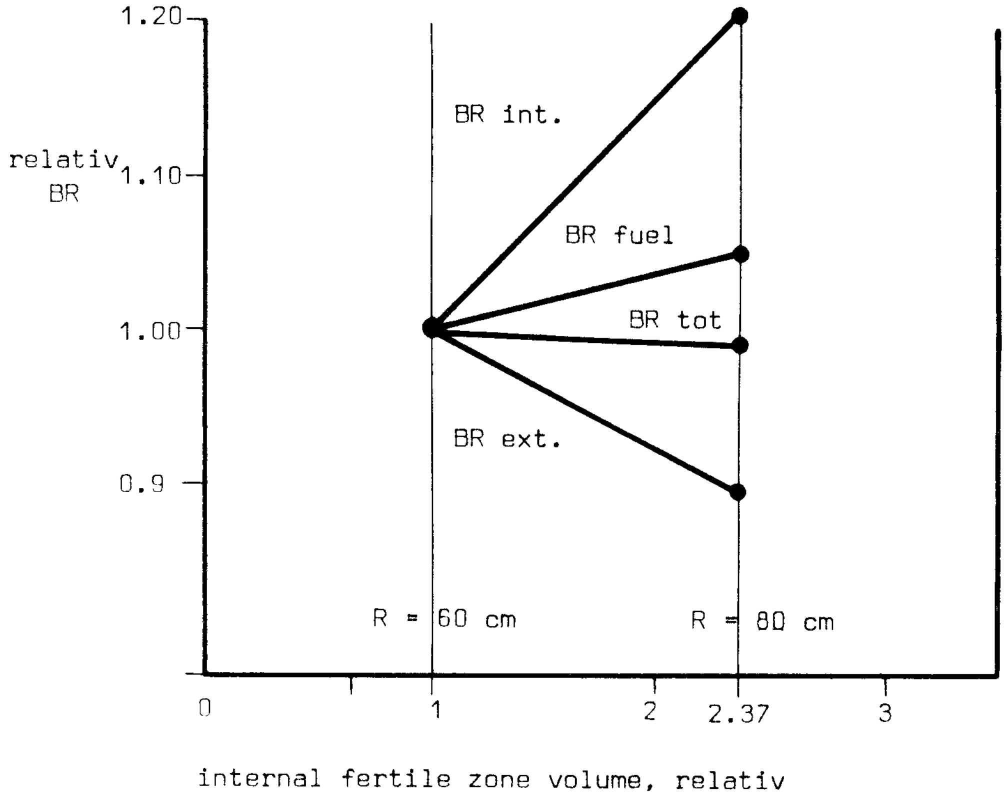


Fig. 3.19 Three zones: fluoride uranium-plutonium fuel
Impact of Pu-239 in fertile material

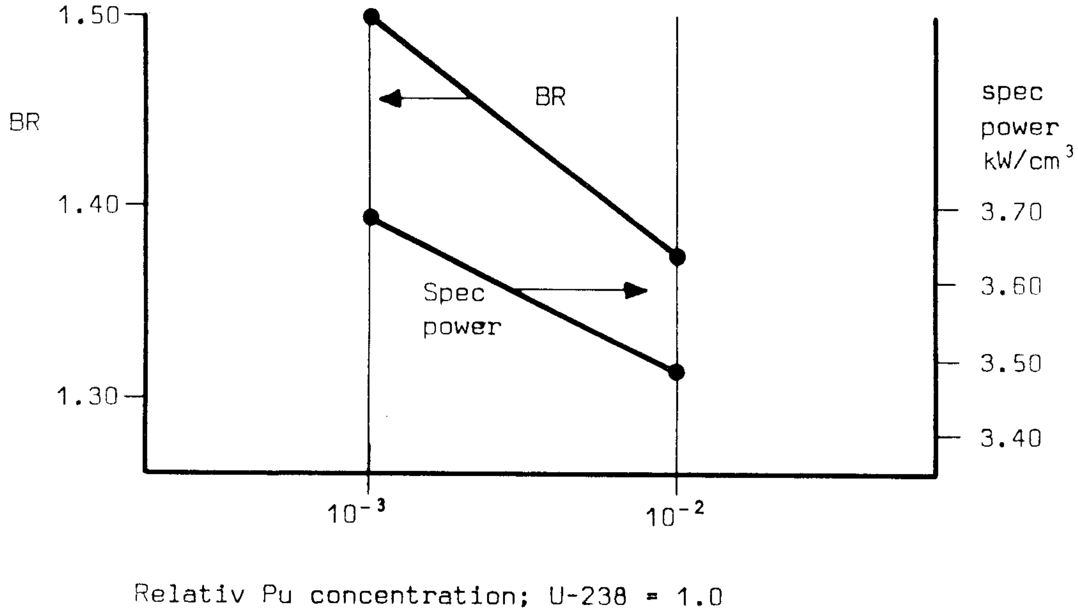


Table 3.16a Three zones reactor: fluoride uranium-plutonium fuel

Influence of graphite as structural material

Number of core	1st wall between internal fertile zone and fuel 2 cm	2nd wall between fuel and external fertile zone 2 cm	B.R. Total	Spec. power; GW/m^3
164	Fe, Mo	Be met	1.427	4.24
171	C graphite	C graphite	1.45	3.65
Ratio of cores $\frac{171}{164}$	$\frac{\text{graphite}}{\text{Fe}}$	$\frac{\text{graphite}}{\text{Be}}$	1.02	0.86

(see Fig. 3.20)

Table 3.16b Two zones reactor: fluoride uranium-plutonium fuel

Influence of the geometry of the reactor

Number of core	Structure			B.R. total	Spec. power GW/m^3
	Internal zone	Intermediate zone	External zone		
160 two zone reactor	Fuel	Blanket zone	Cooling zone	1.424	26.7
161 three zone reactor	Blanket	Fuel cooled out of core	Blanket	1.37	3.47
Ratio of cores $\frac{160}{101}$				1.04	7.69

(see Fig. 3.21)

Fig. 3.20 Three zones-fluoride uranium-plutonium fuel
Graphite instead of beryllium as a structural material

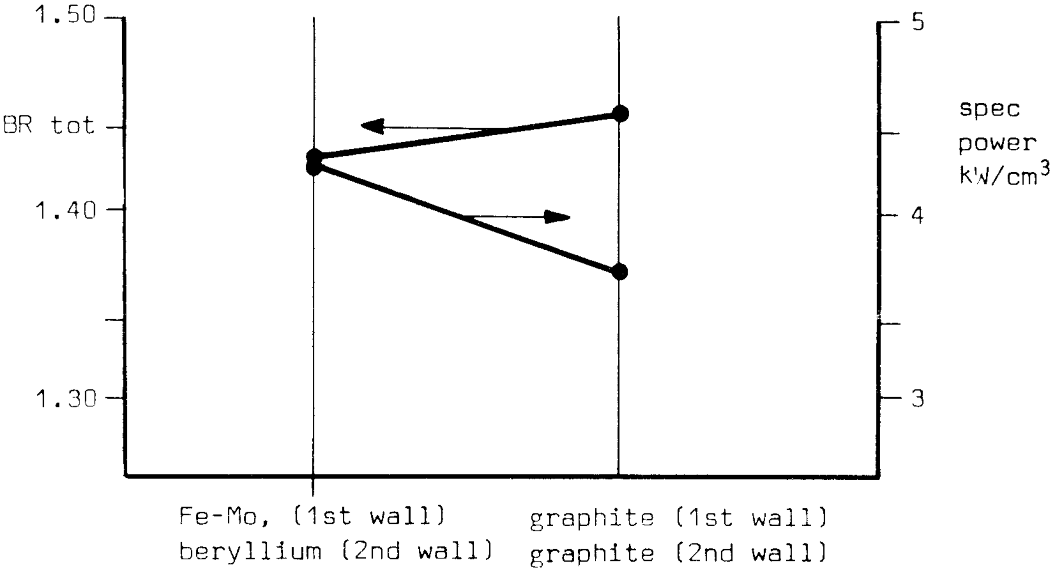


Fig. 3.21 Two zones: fluoride uranium-plutonium fuel
Impact of geometry

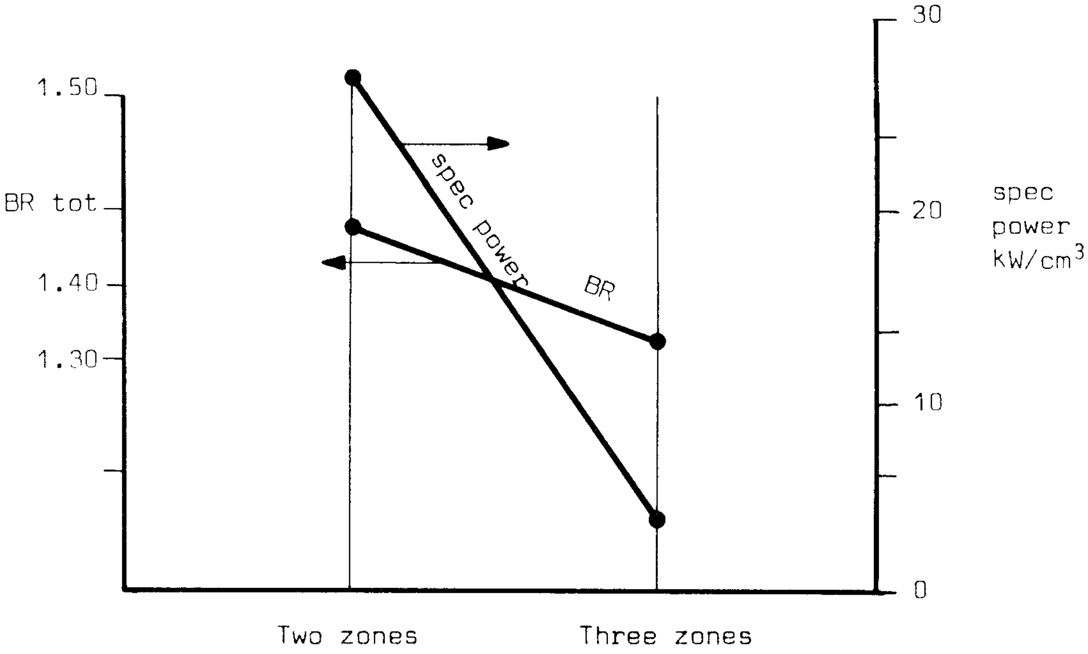
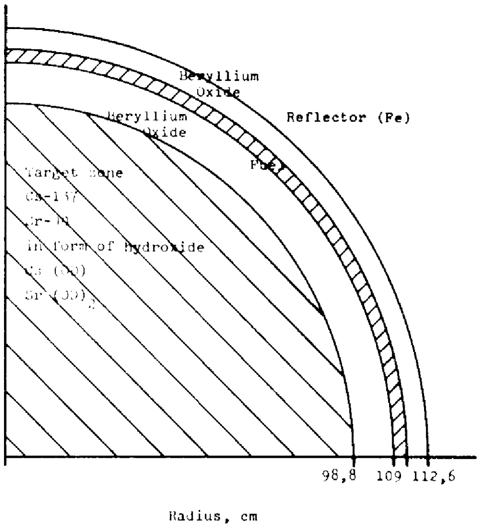


Table 3.17 Three zones reactor: fluoride uranium-plutonium fuel

Influence of the plutonium concentration

Number of Core	Pu total in fuel Mol %	$\frac{\text{Atoms}}{cm^3 x 10^{24}}$	B.R. tot.	B.G. gain	Spec. Power GW/m^3
165	14	0.0022	1.38	0.38	3.45
172	7	0.0011	1.05	0.05	4.05
Ratio of cores $\frac{172}{165}$	0.5	0.5	0.76	0.13	1.17

Fig. 4.12 Burner reactor with molten fluoride fuel
Fuel: PuF_3-3.54 NaF-2.41 ZrF_4
Specific power ~20 kWcm^{-3}
Total power 11 GW (thermal)



3.6 High Flux Reactor with Fluoride Fuel

Can a fluoride fuelled burner, as opposed to a chloride fuelled reactor be considered as a reactor for transmuting fission products? (See chapter 4, for the high flux transmutation reactor).

In such a reactor a fuel made up of $PuF_3/5.4\ NaF/2.4\ ZrF_4$ has been assumed. The calculations have been carried out for a larger bruner of 11 GW(th) and the fission products are assumed to be generated in a total system of 55 GW(th).

The results are not encouraging in spite of the fact that the neutron lfux in the target region was 1.05 times higher for the fluoride fuelled reacotr as opposed to the chloride fuelled reactor (Table 3.18, Fig. 3.22).

The effective half life of both fission product nuclides was (in years)

	In fluoride fuelled reactor	In chloride fuelled reactor (reference)
Cs-137	8.57	8.93
Sr-90	1.73	1.83

These "benefits" must be balanced against a specific power which is twice as high as the reference case. In the fluoride core this is 19.9 $GW.m^{-3}$ and for the chloride reactor 10.1 $GW.m^{-3}$. Such a high specific power is not realisable.

In addition since graphite might be used in place of beryllium oxide as moderator (possible for a neutronic viewpoint) a significant improvement in corrosion problems is obtained. This has been proved by the excellent experience of Oak Ridge National Laboratory with one proviso - at ORNL the fuel was $LiF - BeF_2 - ThF_4 - UF_4$.

Table 3.18 High-flux burner reactor with fluoride fuel

Total power 11 GW(th)
Burning fission products form a total system of 55 GW(th)

Zone Radius (cm) Volume (m^3)	Components (atom $10^{24}cm^{-3}$)	Neutron flux $10^{16}cm^{-2}s^{-1}$ $\frac{\text{total}}{\text{thermal}}$	Specific power ($GW\ m^{-3}$) Transmutation rate (s^{-1})
I 0 - 98.8 cm Target zone Vol: 4.1 m^3	Cs-137 0.0116 Sr-90 0.0016 Oxygen 0.0145 Deuter 0.0145	$\frac{4.01}{2.21}$	Cs-137 : 1.8×10^{-9} Sr-90 : 1.2×10^{-8}
II 98.8 - 109 cm Moderator, wall	Be 0.060 Oxygen 0.060 with thin graphite layer	$\frac{5.09}{1.83}$ $\frac{5.25}{0.239}$	
III 109 - 112.6 cm Fuel zone Vol: 0.55 m^3	Pu-239 0.0017 Pu-240 0.00042 Pu-241 0.00021 Na 0.0075 Zr 0.0051 F 0.0340	$\frac{5.02}{0.0457}$	19.9 $GW\ m^{-3}$
IV 112.6 - 118.6 cm wall	Be 0.060 Oxygen 0.060	$\frac{4.87}{0.034}$ $\frac{3.9}{0.041}$	
V 118.6 - 218.0 cm Reflector zone	Fe 0.08	$\frac{0.0023}{2.8 \times 10^{-9}}$	

4. A HIGH FLUX BURNER REACTOR FOR TRANSMUTATION

4.1 Need for fission product transmutation

4.1.1 Introduction

The problems associated with the management of highly radioactive fission product waste has been intensively and extensively discussed (Fig. 4.1).

Here only the transmutation of fission products (F.P.) is dealt with. The recycling of the actinides is not treated. Transmutation occurs by using neutron irradiation in a fission reactor.

A short outline of this chapter can be presented in the form of the following questions

- why, contrary to many assertions, is neutron transmutation in a fusion reactor not feasable? - this in spite of the fact that the fusion machine has often been proposed for this purpose
- why are recent opinions concerning transmutation in fission reactors rather pessimictic?
- could transmutation in a fission reactor be possible taking into account the neutron balance in a breeding system?
- which fission products are candidates for irradiation in a fission reactor?
- is the rate of transmutation sufficiently high in a fission reactor?
- in what type of reactor is the transmutation physically possible?
- what are the limiting parameters for transmutation in a solid fuelled fission reactor?
- is a very high flux fission reactor possible if the fuel is in the liquid state instead of the solid state?

Fig. 4.1 POSSIBILITIES FOR TRANSMUTATION OF ^{90}Sr AND ^{137}Cs (see also table 4.1)

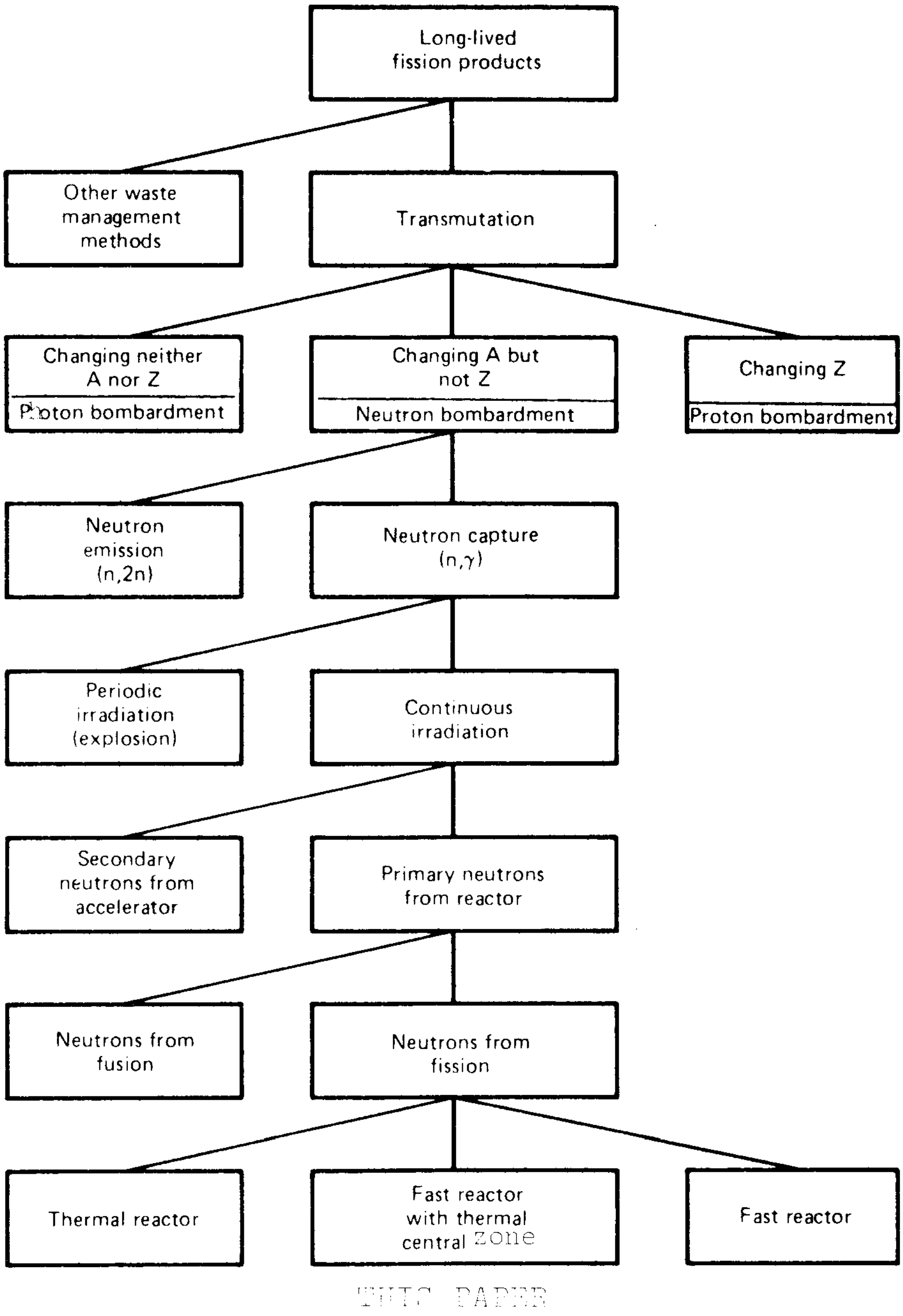


Table 4.1 Transmutation possibilities for different devices

Machine	Flux/ Energy	Reactions, and remarks of authors of original reports.
Accelerator of medium and high energy protons	Protons 100 MeV	Reaction p,xn) not promising. Ruled out on basis of energy balace criteria.
	Protons 1.10 GeV with Cs-137 as target and/or thermalised flux of neutrons	Spalation (p,xn) and (n,2n) (n,γ) secondary neutron flux Not feasible within limits of current technology. The capital cost is prohibitive. (see table 4.2)
Fusion (thermonuclear) reactor in all cases with wall	Fast flux of 14 MeV neutrons from (D-T) $\Phi = 5 \times 10^{14} n\ cm^{-2} s^{-1}$	Neutron reactions (n,2n) and (n,γ). Fast Flux of $5 \times 10^{15} n\ cm^{-2} s^{-1}$
	Thermalised flux in beryllium trap	Practically only (n,γ) Thermal flux $6.7 \times 10^{15} n\ cm^{-2} s^{-1}$
		Attractive transmutation rate has not been demonstrated but possible to transmutate all Cs-137 and Sr-90 created by fission reactors
Nuclear	Fissile explosive or thermonuclear explosive	Technically not feasible. No. of explosions per year very high. Appr. 3900 per year each of 100 kton. (For USA in year 2000 Cs-137 and Sr-90) Probably not ecceptable to public!
Fission reactor		(see Fig. 4.2)

Fig. 4.2 FISSION PRODUCTS

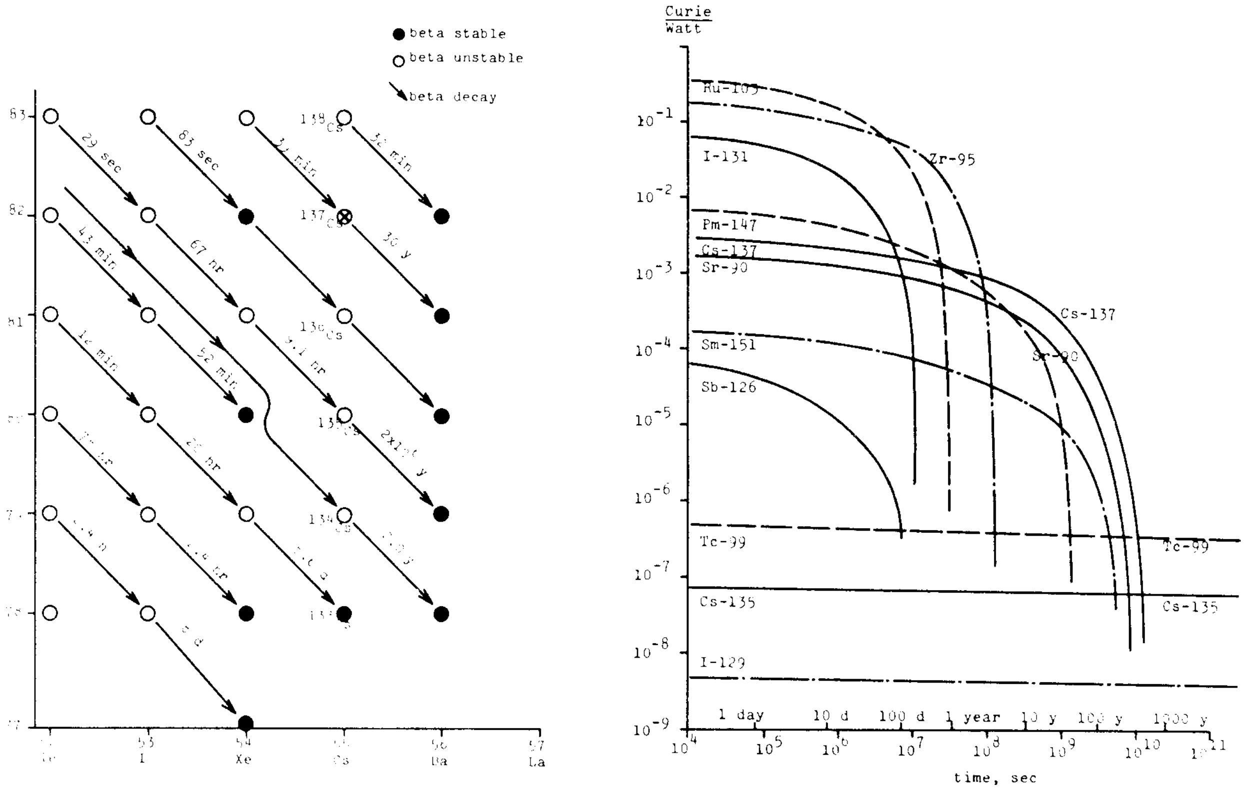


Fig. 4.2 FISSION PRODUCTS

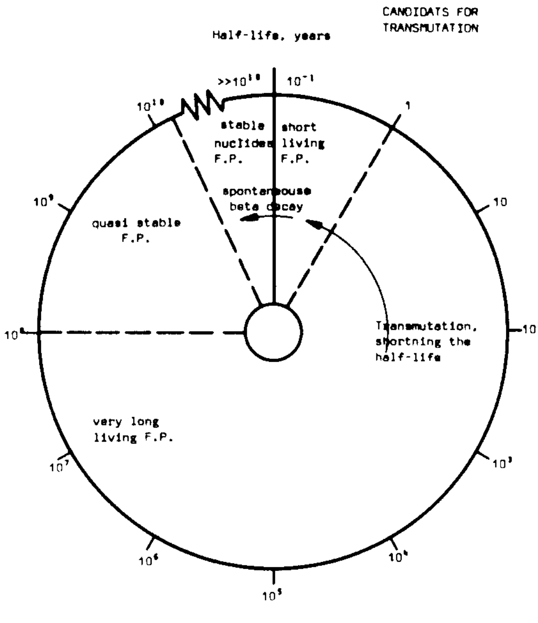


Table 4.2 Possibility for transmutation of F.P. - particularly Cs-137 and Sr-90 in a fission reactor according to BNWL - 1900

	Reactor	Reference	Flux	Remarks
Thermal	power reactor	Steinberg Wotzak Manowitz, 1964		The authors use a wrong value: Kr-85 with large σ = 15 barns instead of σ = 1.7 barns. Isotopic separation of Kr-isotopes.
			3 x 10^{13} thermal	Only I-129 can be transmuted.
	high flux (trap)	Steinberg, 1964	10^{16} in the trap smaller in the presence of the F.P. target.	An equal or greater no. of F.P. would be formed in the fission process per transmutation event.
		Claiborne, 1972	2 x 10^{15} thermal	This reactor does not meet the criteria of overall waste balance and of total transmutation rate.
Fast	liquid metal fast breeder	Claiborne, 1972	1 x 10^{15} fast	Neutron excess 0.15 - 0.3 at the expense of being no longer a viable breeder of fissile material. Also this flux does not allow the attainment of a sufficiently high transmutation rate and is, therefore, not a feasible concept.
Fast with thermal	liquid fuel fast reactor with thermal column.		this paper	

- how could such a high flux reactor with circulating liquid fuel and a thermal column operate as a "burner" for some F.P. (Cs-137, Sr-90 etc.) transmutations? (Fig. 4.3)

- is such a system feasable?

Comment

In BNWL-1900 it was noted that the calculation (in a moderating blanket of the CRT) represents a more realistic blanket configuration with a neutron wall loading of 10 MW/m^2 (This is still a very optimistic value. M.T.).

In this case the following data have been obtained for a thermalised neutron flux from a CTR with a 10 MW/m^2 wall loading. (Tab. 4.3)

Table 4.3

For 80% fraction	Φ thermal ($n.cm^{-2}s^{-1}$)	$\Phi \cdot \sigma$ (n,γ)	$\Phi \cdot \sigma$ $(n,2n)$	effective $t_{1/2}$ effective
∿291 kg Cs/yr	6.71 x 10^{15}	$\sigma_{(n,\gamma)} = 0.117$ barn 7.91 x 10^{-10}	$\sigma_{(n+2n)} = 0.10$ barn 7.0 x 10^{-10}	$\lambda = 22.2 \times 10^{-1} s^{-1}$ 9.9 years

The conclusions of this study are that useful quantities of Cs-137 could be transmuted under the projected CTR blanket loading conditions. The reduction in Cs-137 "toxicity" is still expected to be at most a factor 3 down. In addition a study of the build-up of fission product nuclei in order to establish the requirements of periodix chemical processing and associated costs has not been carried out.

Fig. 4.3 Accelerator for transmutation
BALANCE FOR 1 FISSIONED ATOM

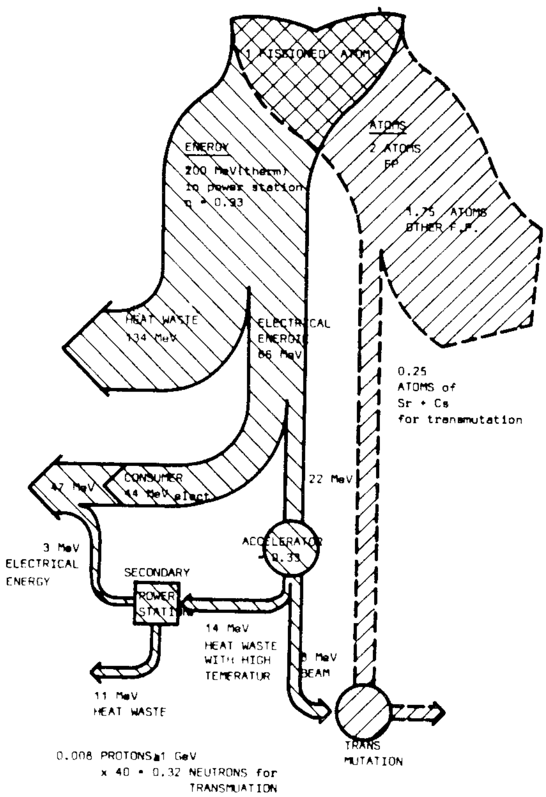
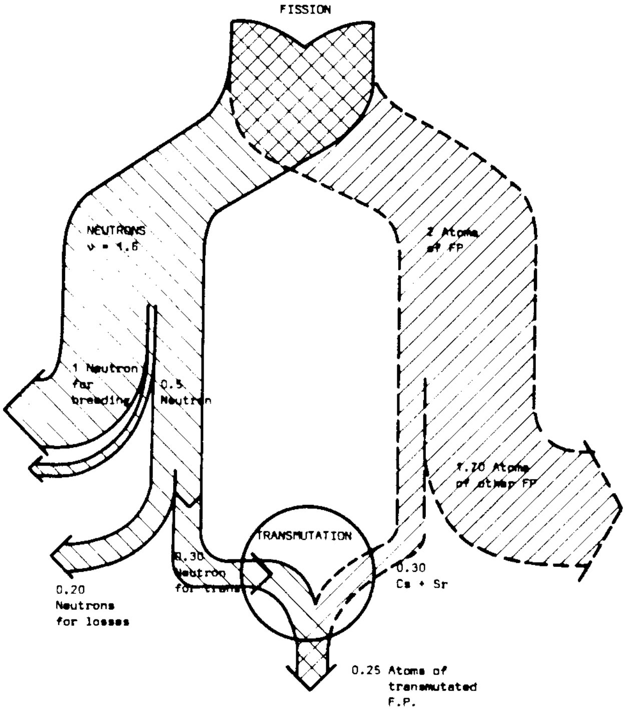


Fig. 4.4 Neutron balance



Comment

H.W. Lefevre (appendix to BNWL-1900) makes an interesting comment on the study of the transmutation of Cs-137 and Sr-90 in CTR: "Everyone knows that a CTR will be "clean". Don't spoil that illusion. I think that I would worry some about a CTR loaded with 50 kg of Cs-137".

4.1.2 Why some opinions concerning transmutations in a fission reactor are rather pessimistic

A recent and most intensive study of the use of a fission reactor for the transmutation of fission products has been published by Claiborne (1972). He writes:

"The problem fission products cannot be eliminated by any system of fission power reactors operating in either a stagnant or expanding nuclear power economy since the production rate exceeds the elimination rate by burnout and decay. Only at equilibrium will the production and removal rates be equal, a condition that is never attained in power reactors. Equilibrium can be obtained, however, for a system that includes the stockpile of fission products as part of the system inventory since the stockpile will grow until its decay rate equals the net production rate of the system. For the projected nuclear power economy, however, this will require a very large stockpile with its associated potential for release of large quantities of hazardous radio-isotopes to the environment. It is this stockpile that must be greatly reduced or eliminated from the biosphere. A method suggested by Steinberg et al. is transmutation in "burner reactors", which are designed to maximize neutron absorption in separated fission products charged to a reactor. If sufficient numbers of these burners are used, the fission products inventory of nuclear power system can then reach equilibrium and be maintained at an irreducible minimum, which is the quantity contained in the reactors, the chemical processing plants, the transportation system, and in some industrial plants.

If the assumption is made that burner reactors are a desirable adjunct to a nuclear economy, what are the design requirements and limitations: it is obvious that they must maximize (with due regard to economics) the ratio of burnout of a particular fission products to its production rate in fission reactors, and the neutron flux must be high enough to cause a significant decrease in its effective half-life. Of the fission types, the breeder reactor has the most efficient neutron economy and in principle would make the most efficient burner if all or part of the fertile material can be replaced by a Sr-Cs mixture without causing chemical processing problems or too large a perturbation in the flux spectrum because of the different characteristics of these fission products. The cost accounting in such a system would set the value of neutrons absorbed in the fission product feed at an accounting cost equal to the value of the fuel bred from those neutrons.

The maximum possible burnout of fission products would occur when the excess neutrons per fission that would be absorbed in a fertile material are absorbed instead in the fission product feed. The largest possible burnout ratio would then be the breeding ratio (or conversion ratio for nonbreeders) divided by the fission product yield. The estimated breeding ratio for the Molten Salt Breeder Reactor (MSBR), a thermal breeder, is 1.05 and for the Liquid Metal Fuelled Fast Breeder Reactor (LMFBR), 1.38. The yield of ^{137}Cs + ^{90}Sr is 0.12 atom/fission, but a number of other isotopes of these elements are produced which would also absorb neutrons. However, if the fission product waste is aged two years before separation of the cesium and strontium, the mixture will essentially be composed of about 80% ^{137}Cs + ^{90}Sr and 20% ^{135}Cs (which will capture neutrons to form ^{136}Cs that decays with a 13-day halflife consequently the maximum burnout ratio for ^{137}Cs + ^{90}Sr will be decreased by 20%. This leads to a maximum possible burnout ratio of about 7 for the MSBR and about 9 for the LMFBR. Unfortunately, however, the neutron fluxes in these designs are well below 10^{16} n cm^{-2}s^{-1}. Any modifications of these designs to create high neutron fluxes will increase the neutron leakage and decrease the burnout ratios significantly". (Claiborne 1972)

Comment

It is not clear why Claiborne claimed that after 2 years ageing and separation of strontium and caesium the isotope composition will be

80% Cs-137
Sr-90

20% Cs-135

From Crouch (1973) the fission products of U-235 have the following composition (2 years ageing) (in at % per fissioned nucleous). (see Table 4.4)

Sr-88	(stable)	3.63	
Sr-90	(28 years)	4.39	
Cs-133	(stable)	6.57	
Cs-134	(2 years)	3.5	(7.09 0.5 from independent yield)
Cs-135		6.26	
Cs-137		5.99	
Subtotal		30.34	

The realistic data are unfortunately more than twice those cited by Claiborne.

The same negative opinions concerning the use of Fission Reactors for F.P. - transmutation are given by the following authors:

- A.S. Kubo (BNWL - 1900):
  "Fission products are not conductive to nuclear transformation as a general solution to long term waste management".
- BNWL - 1900, itself:
  "In summary it is improbable that transmutation of fission products in fission reactors could meet any of the technical feasibility requirements for the production of stable daughters".
- Claiborne (1972):
  "Developing special burner reactors with the required neutron flux of the order of 10^{17}n $cm^{-2}s^{-1}$ is beyond the limits of current technology".

4.1.3 Which fission products are suitable candidates for transmutation and in what quantities?

A simplified breakdown of neutrons and fission products produced by fission of 1 fissile plutonium atom is given in Fig. 4.4

From this it must be clear that only a very limited amount of fission products can be irradiated by neutrons of the whole system to retain a good breeding gain and doubling time - in other words a s elf sustaining and expanding breeding system. For further consideration it is postulated that the maximum number of transmutable nuclides equals T = 0.3.

The proposed system for the transmutation includes two types of reactor:

- power reactors in the form of fast breeder reactors with a total power of three to four times that of:
- a high flux burner reactor.

The crucial F.P. nuclides are characterised in table 4.4 together with others. The data available now makes it possible to estimate the number of candidates for transmutation in our breeder/burner system, using the tollowing criteria

- the total amount of all nuclides to be transmuted cannot be greater than the estimated value of T = 0.30, that is 30 atoms of F.P. nuclides for each 100 fissioned nuclides.
- the order of priority taken from this table is given as Cs > Sr> I > Te
- in the first instance no isotopic separation process is postulated.

Table 4.4 shows the F.P. nuclides selected for transmutation.

Table 4.4 The priorities for the transmutation of fissioned products

Selected	Yield for fission of 100 atoms of Pu-239		Atom/100 atom Pu-239 Subtotal	Assuming isotopic separation atoms/100 atoms Pu-239	
Cs-133 (stable)	6.91	21.140	6.91	0.14	14.37
Cs-135	7.54		14.450	7.54	
Cs-137	6.69		21.140	6.69	
Sr-90	2.18	2.209	23.32	2.18	2.209
Sr-88 (stable) (2% isotopic separation efficiency)	1.44 x 0.02 = 0.029		23.349	0.029	
I-129	1.17	1.55	24.519	1.17	1.18
I-127 (stable)	0.38		24.899	0.01	
Tc-99	5.81	5.81	30.709	5.81	5.81
Kr-83 (stable)	0.36	2.474			
Kr-84 (stable)	0.56				
Kr-85	0.672			0.67	0.71
Kr-86 (stable)	0.882		33.183	0.04	
Total			33.183		24.28

4.1.4 In what way could a burner reactor be coupled to a system of breeders?

The aim of the calculations used here is to show

- that given a system containing some breeding power reactors with a breeding gain of G^B
- the fission products from all of these reactors can be tansmuted in the high flux burner reactor, which includes of course the fission product transmutation for the burner reactor itself.

The calculation of the ratio of breeder power to burner power is as shown here

Transmutation rate (atoms s^{-1}),	T
Yield of fission products (atoms/fission),	Y
Effectiveness of transmutation device	ε

$$T = \varepsilon^{-1}\ (Y(^{90}Sr) + Y(^{137}Cs) + Y(\text{other F.P.}))$$
$$Y(^{90}Sr) = 0.041; \qquad Y(^{137}Cs) = 0.064$$

Breeding gain for the total system without transmutation
G^B = 0.375 (arbitrary)

Breeding gain for the total system with transmutation G^T
Ratio of fission to total capture: α = 0.24 (arbitrary)

$$G = G^B - T/(1+\alpha)$$

Taking a numerical example with the same values arbitrarily chosen (including also another fission product with a yield of 0.1)

$$T = 0.5^{-1}\ (0.041 + 0.064 + 0.1) = 0.40$$
$$G^T = 0.375 - \frac{0.40}{1+0.24} = 0.05$$

which is sufficient for a power system having a slowly increasing capacity with a doubling time of 100 yr. This corresponds to the near steady state case.

To determine X, the number of power reactors

$$X \cdot G^B - 1 = (X+1)\ G^T$$

$$X = \frac{G^T+1}{G^B-G^T} = \frac{0.05+1}{0.375-0.05} = 3.23$$

The correspondence ratio of the power of the breeder reactors and the burner reactor in this case equals

$$X = \frac{30-7}{7} = 3.3$$

This means for example for 8 power reactors each of 3 GW(th) (i.e. a system total of 30 GW(th) can deal with the transmutation of the fission products chosen here. The electrical output assuming an efficiency of 40% is (3x8 + 1x7)x0.40 = 12 GW(e) (Fig. 4.5).

4.1.5 Is the rate of transmutation sufficient?

It is clear that the rate of radioactive nuclide removal in a field of particles is given by

$$\lambda_{eff} = \lambda_{decay} + \lambda_{transmutation} = \frac{\ell n2}{t_{1/2(eff)}}$$

where

λ_{decay} = constant of radioactive decay (s^{-1})

$\lambda_{trans} = \sigma_{trans} \cdot \Phi$

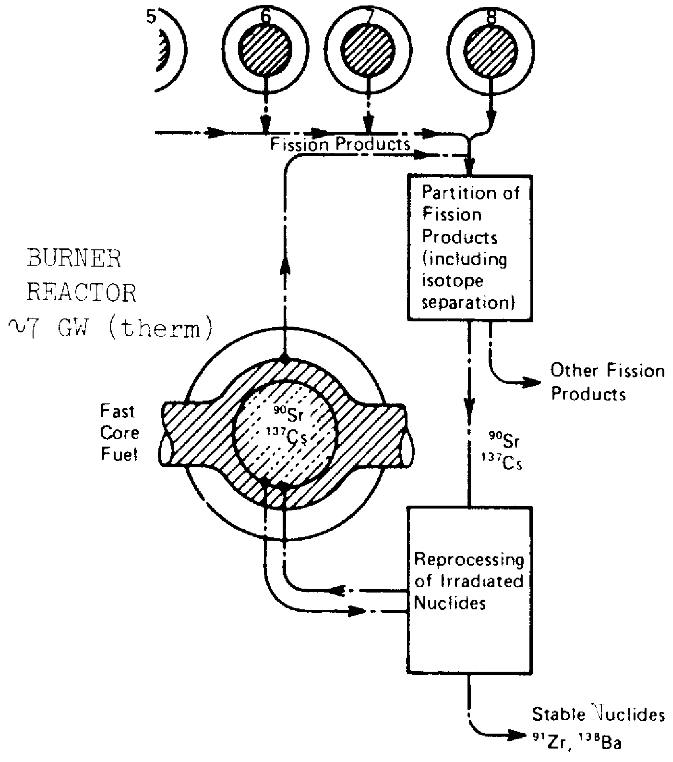
σ = cross section (cm^2) for a given reaction

Φ = flux of the reacting particles (particles $cm^{-2}s^{-1}$)

this value of λ_{eff} will be used later for the calculations of the neutron flux required to permit the transmutation rate to match.

Fig. 4.5 Scheme of the proposed breeding power system with "self-cleaning". (For the sake of simplicity, only the routes of the fission products ^{90}Sr and ^{137}Cs are shown).

8 Power Reactors, each GW (thermal) total $\sim$24 GW (thermal)



Let us assume that the energy production is based on a set of n burners and nX breeders. At a time t_n (see Fig. 4.5) when it is decided to stop the use of fission energy production in favour of other sources the total amount of a selected fission product present is

$$(1) \quad N(t_n) = (X+1)\ n\cdot\frac{k}{\lambda_{eff}}$$

with K = Y•P/E
Y = yield of the selected F.P.
P = power per burner (or breeder) (Watt)
E = energy per fission (Joule)

This amount of F.P. is located only in the burners, therefore each burner can receive

$$(X+1)\ \frac{k}{\lambda_{eff}}$$

although then any production should only represent

$$\frac{k}{\lambda_{eff}}$$

in the steady state.

At time t_n the nX breeders are shut down and only n burners are in operation. Later on (time t_{n-1}) the nuclide removal is such that a re-arrangement is possible and one burner can be stopped, ist F.P. contents will be loaded in the remaining burners etc. At the beginning of each time step t_p the p burners which are still working contain the maximum possible amount of F.P.

$$(X+1)\ \frac{k}{\lambda_{eff}}$$

$$(2)\quad \frac{N(t_n)}{n} = \frac{N(t_{n-1})}{n-1} = \frac{N(t_p)}{p} = \frac{N(t_{p-1})}{p-1} = \frac{N(t_2)}{2} = \frac{N(t_1)}{1} =$$

$$= (X+1)\ \cdot\ \frac{k}{\lambda_{eff}}$$

where N(t) represents the total amount of the selected F.P.

One could imagine other schemes: for example the rearrangement could be made only when 2 burners are to be shutdown. From the reactivity point of view this solution is worse than the one proposed. Coming back to the original solution, one has still to solve at each time step $(t_p,\ t_{p-1})$ the burn-up equation.

$$(3)\quad \frac{dN}{dt} + \lambda_{eff}\cdot N = K\cdot p$$

where the right hand side is the F.P. production.

then the solution is

$$(4)\quad N(t) = \frac{K\cdot p}{\lambda_{eff}} + \left(N(t_p) - \frac{K\cdot p}{\lambda_{eff}}\right)\ \cdot\ \exp(-\lambda_{eff}\cdot(t-t_p))$$

Using (2) the time needed to go from p burners to (p-1) can be deduced

$$(5)\quad \lambda_{eff}\cdot(t_{p-1}-t_p) = \sum_{p=2}^{p=n} \ln\ \frac{1}{1-\frac{X+1}{p\cdot X}}$$

with a summation one obtains the time t_1 after which one burner only is in operation.

$$(6) \quad \lambda_{eff} (t_1 - t_n) = \sum_{p=2}^{p=n} \ln \frac{1}{1 - \frac{X+1}{p \cdot X}}$$

A more direct evaluation can be obtained if n is so large that the number of operating burners changes continously with time (p = n(t)) then by a single elimination of p between (2) and (3) one gets

$$\frac{dN}{dt} + \lambda_{eff} N = \frac{N \lambda_{eff}}{X+1} \qquad \text{or}$$

$$(4') \quad N(t) = N(t_n) \cdot \lambda_{eff} \frac{X}{X+1} (t - t)_n)$$

$$(6') \quad \lambda_{eff} (t_1 t_n) = \frac{X+1}{X} \ln \frac{N(t_n)}{N(t_1)} = \frac{X+1}{X} \ln (n)$$

The two approaches give similar results except at the end when few burners are in operation.

For times longer than t_1 only one burner is operated and the amount of F.P. would decrease from

$$(X+1) \frac{K}{\lambda_{eff}} \text{ to } \frac{K}{\lambda_{eff}}$$

We shall postualte that it has no sense to operate this last burner when the amount of F.P. is only 1.2 times longer than the asymptotic value which requires a new time interval (eq. 4 p = 1).

(7) $\lambda_{eff}(t_o - t_1) = \ln 5 \cdot X$

The total time $t_o - t_n$ will be the sum (6) + (7) which corresponds to the reduction factor

$$\frac{n(X+1)}{1.2} \cdot$$

Further reductions can only be obtained by natural decay ($t > t_o$).

Numerical application: with $X = 4, n = 100$ which means the economy was based before t_n on 400 breeders, the initial F.P. amount is reduced 415 times when the last burner is shutdown. Then the required time is defined by $_{eff}(t_o - t_n) = 8.93$ (8.76 with the approx expression). If this time is to be less than say 60 years 2 reactor generations) then $\lambda_{eff} = 4.7 \times 10^{-9} s^{-1}$ ($t_{1/2}$ eff = 4.7 years).

Now the problem of the intensity of the neutron flux desired for transmutation arises. Since the most hazardous F.P. nuclides are those which apart from their high metabolic activity and high retention in living organismus also have a half life of the same order as a human life span of 60-70 years we arrive at the following list of hazardous isotopes which are the most important for transmutation.

Kr-85 $t_{1/2} = 10$ years, $\lambda_{dec} = 20.9 \times 10^{-10} s^{-1}$

Sr-90 $t_{1/2} = 28.2$ years, $\lambda_{dec} = 7.76 \times 10^{-10} s^{-1}$

Cs-137 $t_{1/2} = 30$ years, $\lambda_{dec} = 7.32 \times 10^{-10} s^{-1}$

desired 'half life' = 4.7 years, $\lambda_{desired} = 4.7 \times 10^{-9} s^{-1}$

as we know $\lambda_{desired} = \lambda_{decay} + \lambda_{transmutation}$

The most important problem arises from the fact that the two nuclides Sr-90 and Cs-137 have very small cross sections for neutron absorption in both the thermal and fast regions.

	Cross section, σ(barn = 10^{-24}cm^2)		Ratio
	thermal	fast	therm./fast
Sr-90	0.8 barns	0.0076 barns	$\sim$ 100
Cs-137	0.11 barns	0.0137 barns	$\sim$ 8

therefore to achieve $\lambda_{desired} = 4.7 \times 10^{-9} s^{-1}$ the neccessary fluxes should be:
fast flux for transmutation of

$$\text{Cs-137: } \Phi_{fast} = \frac{\lambda_{desired} - \lambda_{decay}}{\sigma \text{ (Cs-137 fast)}} = \frac{4 \times 10^{-9}}{\sim 0.01 \times 10^{-24}}$$

$$\Phi_{fast} \cong 4.0 \times 10^{17} (n \text{ cm}^{-2} s^{-1})$$

thermal flux for transmutation of

$$\text{Cs-137: } \Phi_{th} = \frac{4 \times 10^{-9}}{0.11 \times 10^{24}} = 3.2 \times 10^{16} \ (n \text{ cm}^{-2} s^{-1})$$

thermal flux for transmutation of

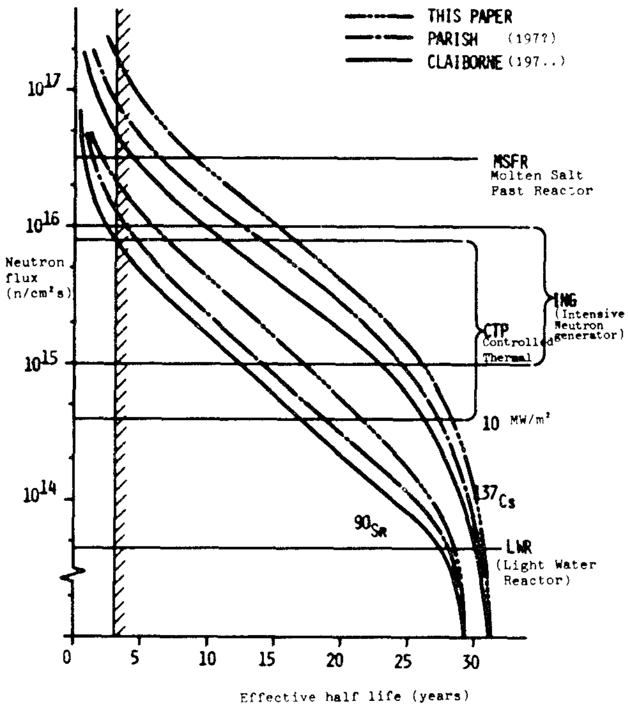
$$\text{Sr-90: } \Phi_{th} = \frac{4.0 \times 10^{-9}}{0.8 \times 10^{24}} = 5.0 \times 10^{15} \ (n \text{ cm}^{-2} s^{-1})$$

(see also Fig. 4.7).

The question then arises, in what device are such fluxes possible - a fast flux of 4×10^{17} of a thermal flux 6×10^{16}. It is interesting to point aout that during the period of 60 years which provides the reduction factor of 415 (if the $\lambda_{eff} = 4.7 \times 10^{-9} s^{-1}$ can be achieved) the natural decay of Cs-137 would have reduced it only by a factor 4 which demonstrates the efficiency of the burner. Also the burning which occures during the first period (t - tn) reduces the amount of

$$\text{F.P.} \frac{\lambda_{eff}}{\lambda_{decay}} \text{ times} = 6.7 \text{ times for Cs-137}$$

Fig. 4.7 Effective half life and neutron flux for transmutation



4.1.6 In what reactors are the transmutations possible?

From the point of view of this paper the most important process is the transmutation of some of these nuclides by neutrons in a fission reactor. The criteria given in chapter 2 limit the choice of system. That is

- the number of F.P. nuclei cannot be too large in relation to the number of fissioned atoms in the burner reactor (reactor for transmutation) because the latter process also produces new fission products.

- the fission reactor should be self-sustaining - that is a breeding system.

- the specific power of the reactor is proportional to the neutron flux. High neutron flux means high specific power which is controlled by the effectiveness of the core cooling.

- the specific power P and the neutron flux Φ are coupled by the fission cross section and the concentration of fissile nuclide (N_f)

$$P = N_f \cdot \sigma_f \cdot \Phi$$

For thermal neutrons σ_f is approx. 700 barns and for fast neutrons only 1.8 barns, that is 400 times smaller.

For the given total power and the same specific power the product $N_f \cdot \Phi$ for the thermal reactor must be approx. 400 times smaller than for a fast reactor. Since the critical concentration of fissile nuclides in a thermal reactor can only be 10 times smaller than for a fast reactor then for a given specific power the neutron flux in a fast reactor can be about 40 times higher than that of a thermal reactor.

The cross section for thermal neutrons for the nuclides considered here is from 3 to 10 times larger than in a fast flux and this must be taken into account.

All these factors bring us to the following solution of the problems under discussion.

- the highest specific power and hence the highest neutron flux is possible if the cooling process is carried out by the fuel itself and not by a separate cooling agent only.
  This directs our interest towards a reactor with molten fuel in spite of the exotic nature of this solution.

- the high flux reactor must be a fast reactor (small for fast fission)

- because $\sigma_{th} > \sigma_{fast}$ the thermalisation of the high flux in a internal thermal zone is postulated, then it is possible that

$$\Phi^{\text{zone}}_{\text{therm.}} > \Phi^{\text{core}}_{\text{fast}}$$

- the first approximation is made for an isotopically pure radionuclide e.g. Cs-137 without Cs-133 (stable) and Cs-135 and also Sr-90 without Sr-88 (stable).

The discussion then results in:

- transmutation of Cs-137 (and some other nuclides) in a thermalised central trap of high flux neutrons:

$$\Phi_{\text{therm}} = 5\times10^{16}\ \text{n cm}^{-2}\text{s}^{-1}$$

- production of a high flux of fast neutrons 5×10^{16} n $\text{cm}^{-2}\text{s}^{-1}$ and the high specific power of 15 kW cm^{-3} is achieved by means of liquid fuel circulating through an external cooler.

- transmutation of other selectred fission products in an external thermalised zone with a thermal flux of 5×10^{15} or 1×10^{15} n $\text{cm}^{-2}\text{s}^{-1}$.

- coupling of one burner - high flux fast burner reactor with a system of 'normal' power breeder reactors.

4.1.7 What are the limitations of a solid fuelled reactor?

Can the desired specific power of 15 kWcm^{-3} be achieved in a solid fuel reactor? These are the self-evident limits in this case.

- the rate of burning of fissile nuclides is limited due to depletion of fissile or an increase of F.P. nuclides

- the heat transfer limitation of fuel/clad to coolant

- the temperature and temperature gradients in the fuel and cladding (melting, mechanical properties)

- the boiling of the coolant
- the limitation of coolant velocity, pumping power, stability.

Now we discuss these limitations in more detail

- the dwell time in a solid fuelled reactor in core for the fissile nuclides must not be too short.

$$t_{dwell} = \frac{\text{concentration of fissile nuclide} \cdot \text{maximum burn-up}}{\text{fission rate}}$$

We could write:

$$t_{dwell} = \frac{N \cdot b}{R}$$

where N = concentration of fissile nuclide

$$N = \frac{R \cdot f}{\sigma_f \cdot \Phi} \text{ and } f = 3.1 \times 10^{10} \text{ fissions/joule}$$

b = maximal burn-up

P = power (watts)

R = fission rate (fission s^{-1})

$R = P \cdot f$

from this:

$$t_{dwell} = \frac{P.f.b}{\sigma \cdot \Phi \cdot P.f} = \frac{b}{\sigma_f \cdot \Phi}$$

$b = 0.03$ (corresponds to 30.000 MWd/t)

$\sigma_{fiss}^{ther} = 700 \times 10^{-24}\ cm^2$

$\Phi = 5 \times 10^{16}\ n\ cm^{-2} s^{-1}$

$t_{dwell} = 850\ s = 14.3$ minutes

but also for $b = 0.10$ we achieve $t_{dwell} = 47.6$ minutes

For a fast reeactor (some arbitrary values)

$b = 0.10$

$\sigma^{fast}_{fiss} = 1.8 \times 10^{-24}$ cm^2

$\Phi = 5 \times 10^{16}$ n cm^{-2}s^{-1}

$t_{dwell} = 1.1 \times 10^{6}$ s = 12.9 days

Conclusion:

- the dwell time in a thermal reactor is prohibitively short,
- in a fast reactor it is more reasonable but still very short, especially in the case of a solid fuel reactor
- the limitation of specific power by heat transfer is as follows: Specific power, P_{spec}, in a "good" 3 GW_{th} power reactor and with the appropriate flux taken from literature is:

thermal P_{spec} = 0.05 kW/cm^3; Φ_{th} = 5×10^{15} n cm^{-2}s^{-1}

fast P_{spec} = 1 kW/cm^3; Φ_{fast} = 5×10^{15} n cm^{-2}s^{-1}

In a high flux reactor: (see also Fig. 6 1.8)

thermal: P_{spec} = 2.0 kW/cm^3; Φ_{th} = 3×10^{15} n cm^{-2}s^{-1}

P_{spec} = 1.5 kW/cm^3; Φ_{th} = 3×10^{16} n cm^{-2}s^{-1}

fast: P_{spec} = 1.0 kW/cm^3; Φ_{fast} = 1.5×10^{16} n cm^{-2}s^{-1}

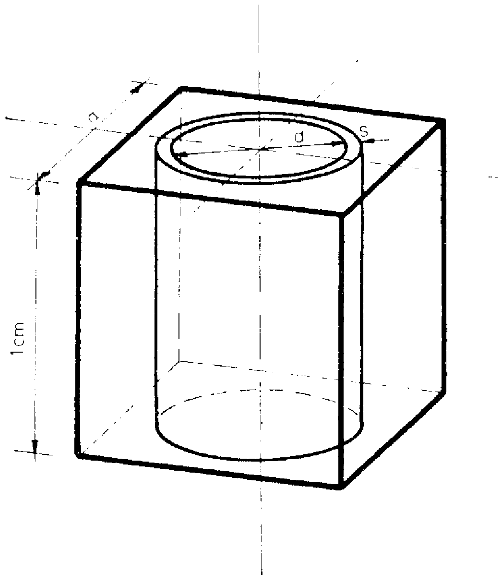
With the same geometry the very high flux reactor desired here would have the following flux for Cs-137 transmutation:

for Φ_{th} = 5.0×10^{16}, the specific power P_{th} = 22.5 kW/cm^3

for Φ_{fast} = 4.0×10^{17}, the specific power P_{fast} = 20 kW/cm^3

For a solid fuel we postulate the following "unit-cell"

	Dimension	Volume	Cross-section area	Surface-area
Cell:	0.9x0.9x1.0 cm	0.81 cm^3	0.81 cm^2	3.60 cm^2
Fuel:	= 0.60 cm	0.283 cm^3	0.283 cm^2	1.885 cm^2
Cladding:	diam = 0.63 cm			
	wall: s = 0.03 cm			
	diam = 0.60 cm	$0.568 \times 10^{-2} cm^3$	$0.568 \times 10^{-2} cm^2$	1.904 cm^2
Coolant:		0.521 cm^3	0.521 cm^2	



In this specified cell of a "desired" high-flux-reactor, we would achieve a heat-flux, per unit fuel element surface area: (for both bypes of reactors, thermal and fast)

$$H_{fs} = \frac{21\ kW/cm^3 \cdot 0.81\ cm^3}{1.885\ cm^2} = 9\ kW\ cm^{-2}$$

Using now a simplified model for the first guess of the temperature gradient we can say: the amount of heat generated in the fuel must be the same as that leaving the surface of the cladding material.

$$\Delta T_{clad} = H_{fs} \cdot \frac{s}{\lambda}$$

Where s = wall thickness and λ heat conductivity $(W \cdot cm^{-1} \cdot K^{-1})$ an optimistic value for stainless steel is $\lambda = 0.4\ W \cdot cm^{-1} \cdot K^{-1}$.

$$\Delta T_{clad} = 9000 \cdot \frac{0.03}{0.4} = 675^{0}C$$

It is evident that this result is <u>not realistic.</u>

The solution of this problem may be the thermalisation of neutrons in a high flux fast core and the irradiation of Cs-137 in a thermal central zone.

In such a thermal central zone we postulate (and this must be based later on core calculations)

$$\Phi_{th} = 1.2 \cdot \Phi_{fast}$$

to reach $\Phi_{th} = 6.0 \times 10^{16}$ we require $\Phi_{fast} = 5.0 \times 10^{16}\ n\ cm^{-2}s^{-1}$. For this fast flux the specific power can be assumed, if we take into account the effective increase of the fission cross section because of the influence of the thermal trap. The simplified calculation results in a specific power of 10 kW cm^{-3}.

The corresponding heat-flux is therefore reduced to

$$H_{fs} = \frac{P_{spec} \cdot V_{cell}}{A_{fs}} \quad 4.3\ kW\ cm^{-2}$$

and the temperature gradient to

$$\Delta T_{clad} = 4300\ (W \cdot cm^{-2})\ \frac{0.03\ (cm)}{0.4\ (W \cdot cm^{-1}K^{-1})} = 323^{0}C$$

This value is still rather high. A rough estimate of the thermal stress in the wall can be taken as

β = coefficient of linear expansion (K^{-1})

$$\sigma = \frac{3}{4} \cdot \beta_{th} \cdot \Delta T \cdot E;$$

E = modulus of elasticity ($Kp \cdot cm^{-2}$)

and the corresponding values for stainless steel (19-9 DL)

$$\sigma = \frac{3}{4} \cdot 15 \cdot 10^{-6} (K^{-1}) \cdot 323 \ (K) \cdot 2.5 \cdot 10^{6} (Kp \cdot cm^{-2}) = 5500 \ Kp \cdot cm^{-2}$$

It is also evident that this result is not realistic!

The resulting thermal stress in the cladding wall coupled with the high flux is prohibitive.

Impact of the thermal flux tail in the fast neutron solid fuel core

Hiere, the most important problem is the local overheating by the thermal flux present at the interface of the thermal column and the solid fuel core. If, in the fast region near to the interface approx. 20% of the enrgy comes from the thermal neutrons, the following ratio of fluxes must be achieved:

$$\sigma_{th} \cdot \Phi_{th} \ (\sigma_{fast} \cdot \Phi_{fast}) \ 0.2$$

$$\sigma_{th} \ \frac{\sigma_{fast}}{th} \cdot \Phi_{fast} \cdot 0.2 \ \frac{1.8}{700} \cdot 0.2 \cdot \Phi_{fast}$$

$$\Phi_{th} = 5 \cdot 10^{-4} \cdot \Phi_{fast}$$

That the thermal flux must be 2000 times lower than the fast, would seem very difficult to realise.

4.1.8 The liquid-fuelled fast reactor with central thermal zone

A much better solution is using a liquid-fuel. The transfer of the heat generated is done, by pumping and cooling the liquid fuel out of core.

For a unit cell of 1 cm^3, which in this case consists of fuel only, we can write the following heat-balance.

$P_{spec} = (\rho \cdot c) \cdot w \cdot \frac{T}{l}$; $(\rho \cdot c)$ = heat capacity $(J \cdot cm^{-3} \cdot K^{-1})$

w = Velocity of fuel (cm/s)

T/l = temperature increase per unit cell $(K \cdot cm^{-1})$

P_{spec} = Specific power $(W \cdot cm^{-3})$

If we allow in the core a temperature increase of $\Delta T/l \sim 3\ deg \cdot cm^{-1}$

$$w = \frac{21.10^3\ (W \cdot cm^{-3})}{2.0\ (J \cdot cm^{-3} \cdot K^{-1}) \cdot 3\ (K \cdot cm^{-1})} = 35\ m.s^{-1}$$

This velocity appears to be within the practical limits proposed for a fuel-velocity of 40 $m.s^{-1}$ for a reactor with 10 $kW.cm^{-3}$ specific power. This point however has to be seriously investigated, as erosion due to high velocities is a problem.

For a 7000 MW(th) core with a specific power of 21 $kW \cdot cm^{-3}$ the fuel volume is about 0.330 m^3.

The target volume, that is the volume of irradiated (transmuted) fission products e.g. Cs-137, is postulated as 1.3 m^3. The diameter of a spherical core is therefore 146 cm. The temperature increase of the unit cell of fuel, in one pass through the core, equals approximately

$$T_{fuel} = 3\ (deg \cdot cm^{-1}) \cdot 146\ (cm) = 438^0C$$

and for a fuel inlet temperature of 550^0C we reach an outlet temperature of approx. 988^0C.

The idea of a liquid fuel high flux reactor has been discussed for some years. Lane (1971) for example writes about high flux fas reactors:

"As an alternative some consideration has been given at Oak Ridge to the possibility of using a molten salt reactor as a fast flux test facility. The primary virtue of this approach includes the ability to achieve very high power densities and at the same time eliminate the down time associated with refuelling the reactor. A fast spectrum molten salt reactor however requires a high fissile concentration (i.e. 300 to 500 g ^{235}U/litre) in order to get mean neutron energies in the range of 10 to 50 keV. Switching from an NaF - UF_4 salt (on which the energy range just mentioned is based) to a <u>chloride-salt reactor</u> would permit a higher mean energy for the same fuel concentration but would require the development of a new technology associated with the use of chlorides. Since the fast flux level is largely determined by the power density a flux of the order of 10^{16} or more corresponds to a peak power density in the fuel salt in a range of 5 to 10 MW/litre and to a power level of about 1000 MW(th). This means that there will be only 100 to 200 litres in the core: however the external volume would be about 10'000 litres".

4.2 The Neutron-physical Aspects of the High Flux Reactor

4.2.1 Introduction

The idea of destroying the beta active long lived radionuclides is based on the following:

$$\begin{tikzcd}
{}^{A}_{Z}FP \arrow[rr, "\beta^-", "\substack{\text{spontaneous decay}\\ t_{1/2}\sim 10\text{ years}}"'] \arrow[d, "(n,\gamma)\quad\text{neutron irradiation}"'] & & {}^{A}_{(Z+1)}E \\
{}^{(A+1)}_{Z}E^{*} \arrow[rr, "\beta^-", "\substack{\text{spontaneous decay}\\ t_{1/2}\text{ is very short}}"'] & & {}^{(A+1)}_{(Z-1)}E
\end{tikzcd}$$

FP = primary fission product

E* = FP after irradiation and is unstable

A = atomic mass

Z = atomic number

E = stable nuclide

Fig. 4.10 gives some of the given transformations which may occur under a high flux. (Remark: Fig. 4.8 and 4.9 omited)

In this system the following simple assumptions are made

- the amount of fission products come from both the fast power reactor breeders and the thermal burner.
- the fuels and materials are continually reprocessed
- the irradiated fission products are continously (or periodically) separated in order to eliminate the daughter stable nuclides (e.g. Zr-90 and Zr-91) from the decay and burning of Sr-90
- the amounts of transmutated nuclides in the steady state (SS) irradiation are calculated by the obvious relationships for the i-th nuclide.

4.2.2 Neutronic calculations

A reference burner reactor concept is shown in Fig. 4.11. The flux trap is surrounded by a BeO beryllium oxide spectrum convertor, a critical fuel thickness and an outer wall (see Table 4.5 and Table 4.6). Fig. 4.12 shows the calculated flux distribution. The total flux in the fuel is similar to that in the flux trap (Fig. 4.13)

4.2.3 Moderation requirements

To form a thermal neutron flux trap one must naturally use neutron moderating materials. As is well known, light materials can scatter neutrons past the neutron-absorbing intermediate-energy resonance region. $^{1}_{1}H$ is the most efficient nuclide in this respect but als exhibits appreciable thermal absorption. Deuterium $^{2}_{1}D$, beryllium $^{9}_{5}Be$ and arbon $^{12}_{6}C$ are usual alternatives. Oxygen $^{16}_{3}O$ is rather heavy though frequently already present in a molecular combination. Other light nuclides have unacceptable nuclear or physical limitations.

Fig. 4.10 Transmutation of selected Nuclides-fission products

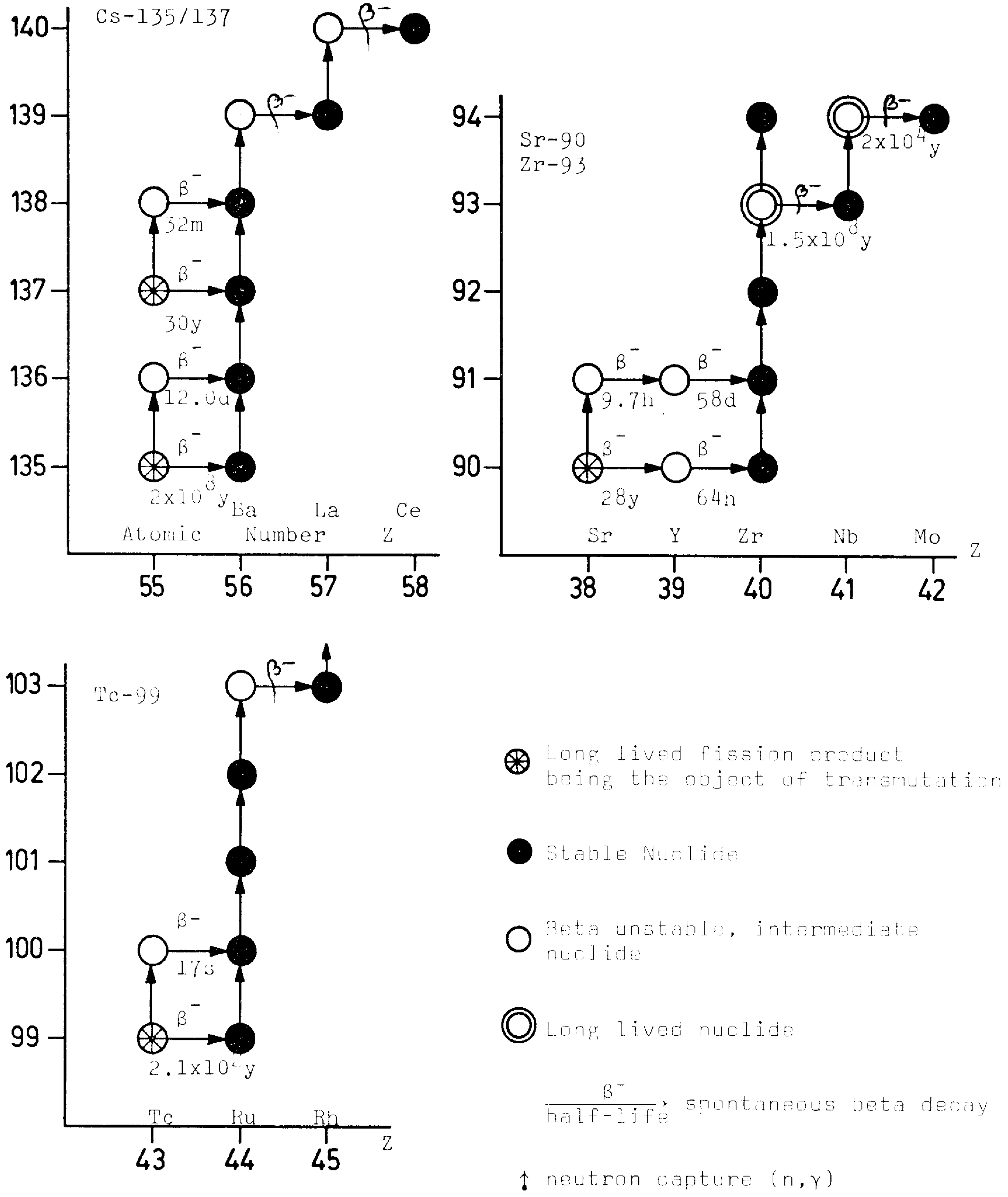


Table 4.5

OBJECT: HIGH FLUX BURNER REACTOR WITH THERMAL ZONE

REACTOR TYPE :
GEOMETRY: INTERNAL ZONE : THERMAL, HIGH FLUX
WALL : IRON, GRAPHIT
INTERMEDIATE ZONE: CORE, FUEL
WALL : IRON
EXTERNAL ZONE : -
WALL, REFLECTOR : IRON

POWER (GW thermal) : 7
POWER DENSITY (GW therm/m^3 core): ∿ 10
NEUTRON FLUX, MEAN (n/cm^2s) : ∿ 4×10^{16}

FISSILE NUCLIDE: Pu-239
DILUTENT : NaCl
COOLING SYSTEM : out of core
BREEDING RATIO : -

PARAMETER STUDIED: Moderators in thermal zone
: Wall, thickness, beryllium, graphite
: Volume, specific power
: cooling parameters

METHOD OF NEUTRONIC: ANISN S4
CALCULATION : 23 Groups
P_1 APP.
: GCC3 CODE
END

Fig. 4.11 GEOMETRY OF THE HIGH FLUX REACTOR

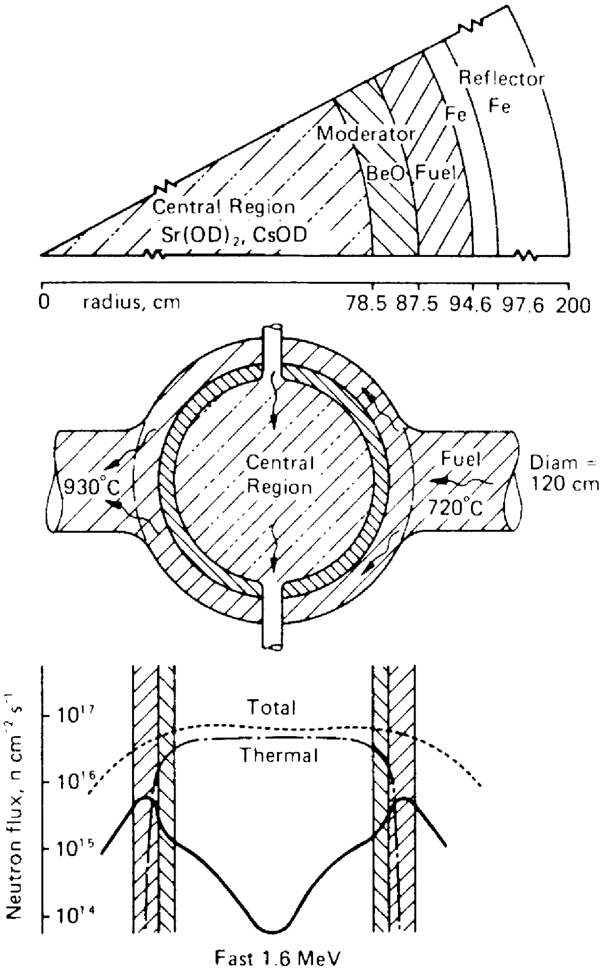


Table 4.6 Geometry and Neutronics of the High-Flux Burner Reactor

(Total power = 7 GW(th): fission products from the total system of 30 GW(th)).

Region	Radius (cm)		Components (atoms 10^{-24}cm^{-3})		Neutron Flux (n cm^{-2}s^{-1}) $\frac{\text{total}}{\text{thermal}}$
	Inner	Outer			
Cetral for transmutation	0.0 Volume 2.1 m^3	78.5	^{137}Cs ^{90}Sr O D (Cesium and strontium deuteroxide)	0.0116 0.0016 0.0145 0.0145	$\frac{3.83}{2.05} \times 10^{16}$
Wall, moderator	78.5 Volume 0.82 m^3	88.0	Be O (Beryllia: 8 cm, graphite: 0.5 cm)	0.060 0.060	$\frac{4.48}{1.79} \times 10^{16}$
Core (fuel)	88.0 Volume 0.67 m^3	94.6	^{239}Pu ^{240}Pu ^{241}Pu Na Cl (Plutonium, sodium chlorides: $PuCl_3 \cdot 6\ NaCl$)	0.0014 0.0004 0.0002 0.012 0.018	$\frac{4.03}{0.0156} \times 10^{16}$
Wall	94.6	97.6	Fe	0.08	$\frac{4.00 \times 10^{16}}{5 \times 10^{11}}$
Reflector	97.6	200	Fe	0.08	Boundary flux $\frac{2.4 \times 10^{13}}{10^{5}}$

Fig.4.12 Neutron spectrum in the core
Total mean flux $4.03 \cdot 10^{16}$ n $cm^{-2}s^{-1}$
Specific power 10.1 $kWcm^{-3}$
Total power 7 GWth

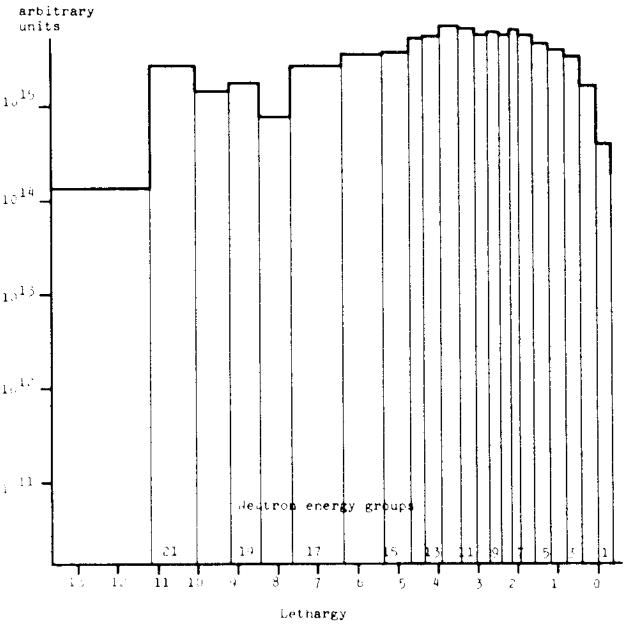
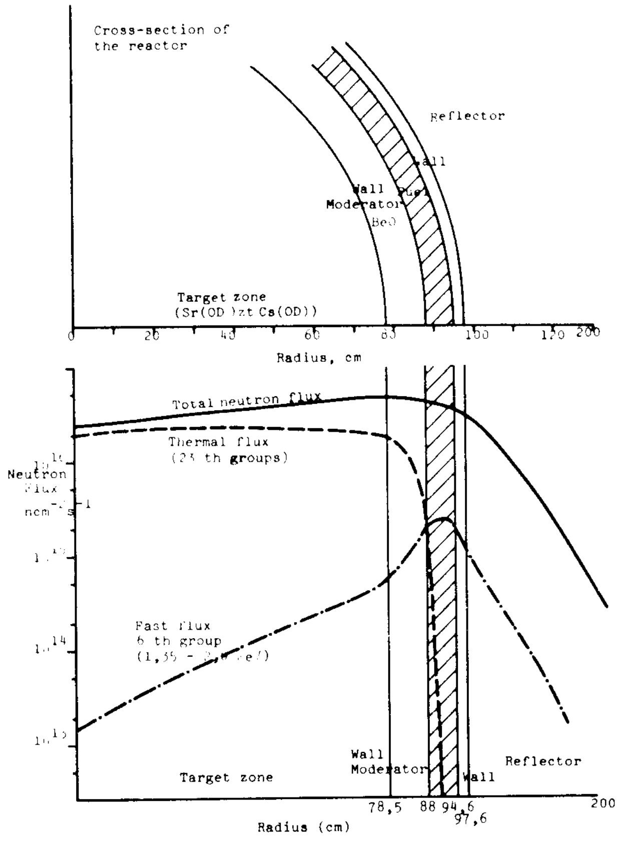


Fig. 4.13 High flux burner reactor



Considering chemical and physical properties, the logical materials to be used inside the lux trap are hydroxide and/or deuteroxide compounds of the FP. Fig. 4.14A shows that just a small proportion of H molar fraction has a large deleterious effect on the Cs-137 transmutation rate. This is due to the H absorption cross section. Therefore, CsOD and $Sr(OD)_2$ are preferred.

As Sr-90 and Cs-137 also have their fair share of resonances it is advantageous to thermalise the flux before reaching the flux trap region containing these targets. Therefore a spectrum converter between flux trap and fast fuel is needed. Bearing in mind the high temperatures to be obtained in this reactor and possible chemical reactions with molten salt, H_2O and D_2O are unacceptable. This leaves Be, BeO and graphite or some variant therefore for consideration. Be (and D) compounds, of course, have also to their advantage a relatively low (n,2n) threshold (1.67 MeV). Location nect to a fast region can therefore produce considerable very slow neutrons in the flux trap - which is a main objective of the burner reactor. Replacement of Be by C or Mo wall material should therefore lower the FP transmutation rate, and it does. (Table 4.7 Fig 4.14B) indicates an optimum thickness of about 5 cm Be. For the sake of safety and higher melting temperature, BeO is preferred over Be.

Table 4.7 Effect on Replacing Be Converter upon the Relative FP Transmutation Rates

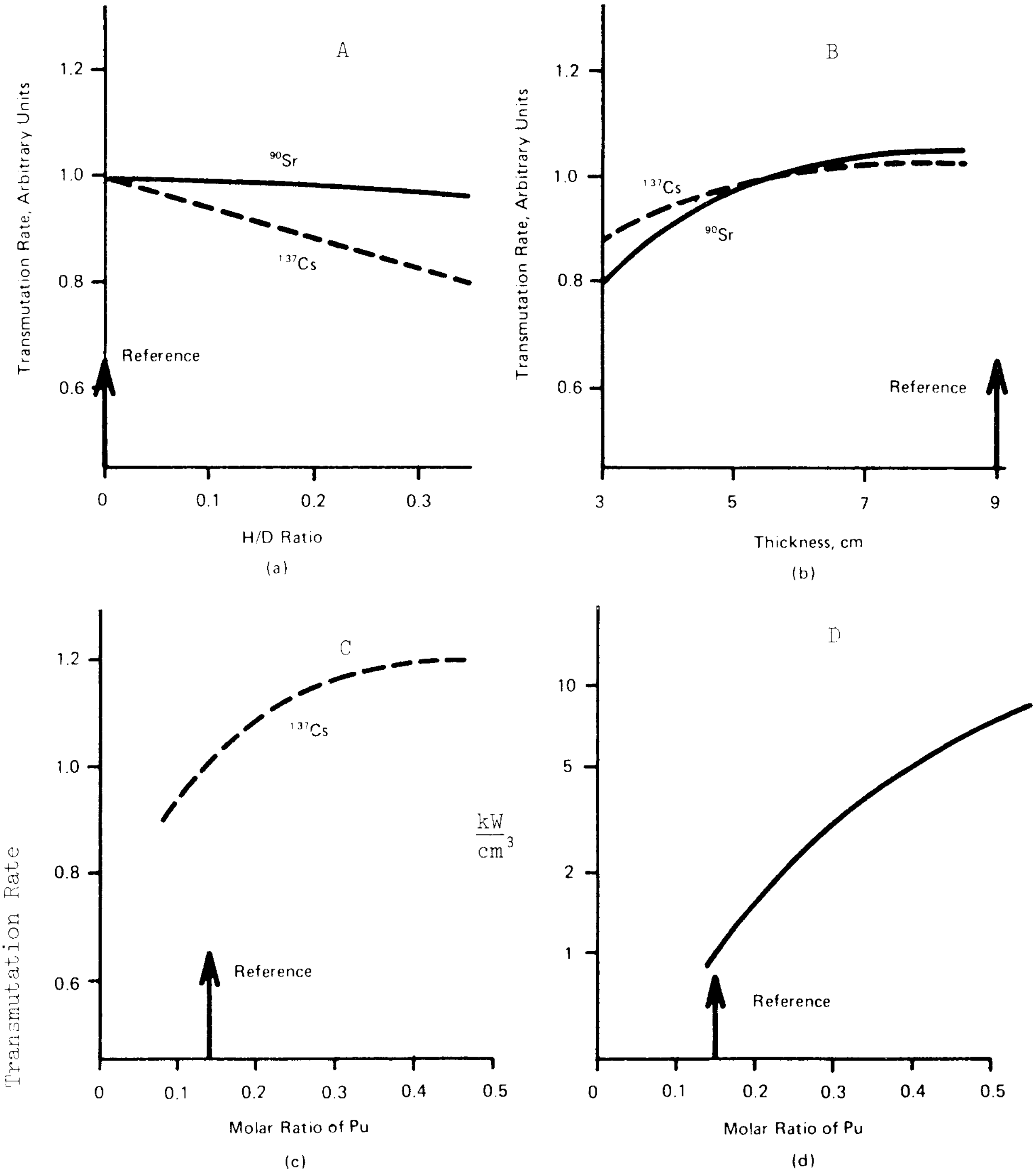
case	materials	$\lambda_{transm.}$ (Cs-137)	$\lambda_{transm.}$ (Sr-90)
1	Be, Be	1.0	1.0
2	C, Be	0.72	0.68
3	Be, Fe/Mo	0.84	0.82

(Note. transmutation rate in arbitrary units)

4.2.4 Influence of other parameters

The influence of plutonium concentration (Fig. 4.14C) is an important parameter of this reactor. Increasing the plutonium concentration significantly improves the transmutation rate but also increases the power density above a technically feasable level (Fig. 4.14D). See also Fig. 4.15.

Fig. 4.14 Impact of some selected parameter variations: (a) hydrogen/deuterium, (b) thickness of moderator (BeO), (c) plutonium concentration, and (d) power density, P (kW cm^{-3})



(a)

(b)

(c)

(d)

Increasing the reactor power (Fig. 4.16A) from 5 to 11 GW(th) improves the transmutation rate. However a power unit above 7 GW(th) seems to be beyond the technological limit even for the distant future. The reference case has been taken as 7 GW(th).

The use of beryllium (Fig. 4.16B and 4.16C) instead of iron in the reflector of the core improves the transmutation rate significantly. At the same time the power density increases prohibitively (Fig. 4.16D). The reference case contains iron in the reflector.

Molten fuel offers the only way of handling the very high power densities of 10 GWm^{-3}. In addition the very steep gradient of fission rate makes a molten fuel core essential since the local fission density can be one order of magnitude greater than the mean density. In a solid fuel core the high heat removal rates would not be achievable.

The use of boron to absorb the thermal neutrons results in a definite decrease in the rate of caesium transmutation (Fig. 4.17).

The large size of the thermal flux trap results in the fuel region approaching slab geometry with attendant high neutron leakage. To better economise on neutrons several possibilities may be tried

- use of an optimised reflector such as Fe, Ni, Cu or Be to minimise the critical mass
- use of the outer neutron leakage for breeding
- use of the neutron leakage for additional FP transmutation

To begin with a solid Fe reflector was assumed (Fig. 4.18).

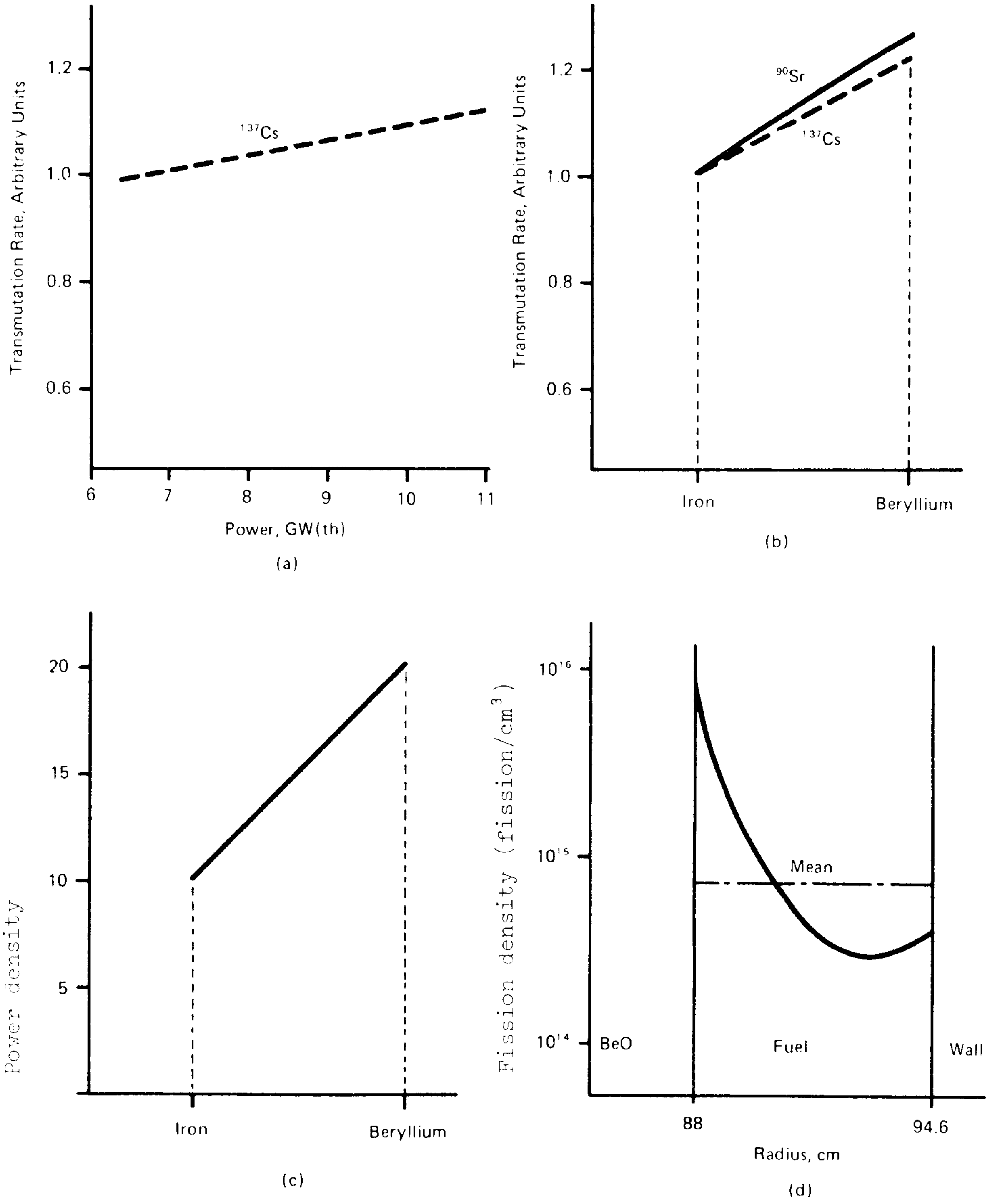
4.3 Thermohydraulic considerations

We now examine the thermohydraulic implications (for more detail see ch. 8).

The crucial parameter here is the core power density. The given value is high but still near the present state of the art (Table 4.8).

For comparison, power densities for some high flux reactors. (Table 8)

Fig. 4.16 Effect of variation in some selected parameters: (a) power, (b) power (b) reflector, (c) power density, P(GW m^{-3}), and (d) fission density in fuel, F (fission cm^{-3})



(a)

(b)

(c)

(d)

Fig. 4.15 Impact of plutonium concentration in the fuel

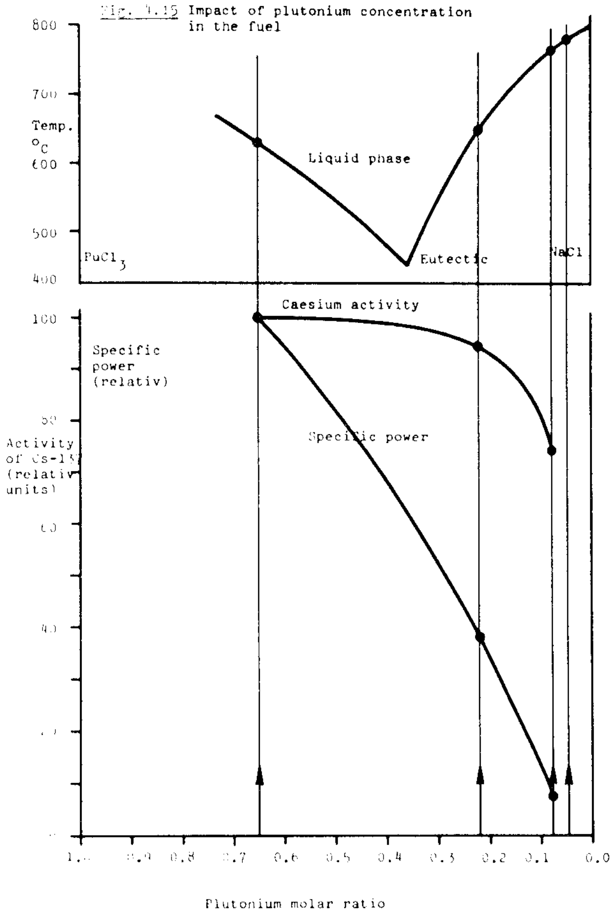


Fig. 4.17 Impact of natural boron

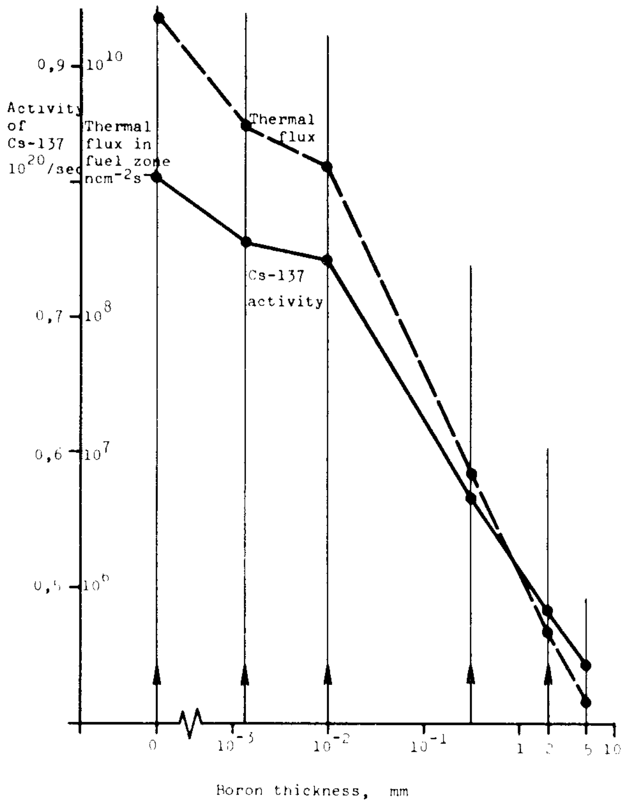


Fig. 4.18 Impact of reflector

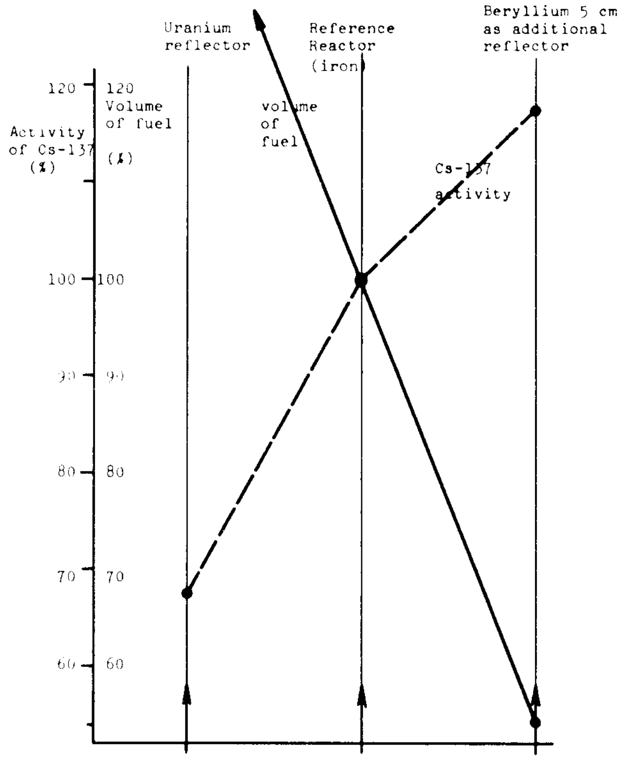


Table 4.8 Thermohydraulics of the High-Flux Burner Reactor

Parameter	Unit	Value
Total power	GW(th)	7.0
Core volume	m^3	0.69
Power density	$kW\ cm^{-3}$	10.5
Fuel density	$g\ cm^{-3}$	2.35
Heat capacity, mass	$J\ g^{-1}K^{-1}$	0.83
Heat capacity, volume	$J\ cm^{-3}K^{-1}$	1.95
Diameter of tube inlet	cm	120
Inlet velocity	$m\ s^{-1}$	15
Volumetric velocity	m^3s^{-1}	17
Heat capacity	$GW\ s\ K^{-1}$	0.033
Temperature increase (outlet-inlet)	^oC	210
Temperature of fuel inlet	^oC	720
Temperature of fuel outlet	^oC	930
Mean velocity of fuel in core	$m\ s^{-1}$	12
Cooling	---	Out of core in heat exchanger cooled by sodium

This seems to indicate that a total specific power rating of about 1 kgPu/MW(th) may be achievable.

Table 8 Power density in high-flux reactors

	Power density GW(th)/m^3	
	in core volume	in coolant volume
Feinberg, research reactor	3-5	8-10
Melekes CM-2 (Soviet Union)	2•5	5
FFTF (USA)	1•0	2
Lane (Molten chlorides)	5-10	5-10
HFIR (USA) mean	2	4
maximum	4•38	8•5
Phenix 250 (France)	0.46	1.0
This paper	10•9	10•9

The crucial problem will be the efficiency of the external heat exchanger. In the following example some typical heat exchanger characteristics are taken to demonstrate the possibilities (calculated for 11 GW(th).

Specific heat exchanger power (conservative data)	1 kW/cm^3
Total volume of heat exchanger for 11 GW(th)	11 m^3
Volumetric fuel ratio	0.3
Fuel volume in heat exchanger	3.3 m^3
Fuel in the core heat exchanger piping	1.0 m^3
Total fuel out of core	4.3 m^3
Fuel in core	1.0 m^3
Total fuel in system	5.3 m^3
Mean specific power of fuel in the whole system $\frac{11\ \text{GW(th)}}{5.3\text{m}^3}$	2.07 GW/m^3
Plutonium content of fuel	0.8 gPu/cm^3
Power rating of whole system	0.385 kgPu/MW(th)
The postulated power rating for the whole system	1 kgPu/MW(th)
For this case calculated the power rating in the breeder power reactors	1.15 kgPu/MW(th)

4.4 Some results

Parametric studies were made as variations around a reference system which assumed $P = 11$ GW(th) ($X = 2{\cdot}9$, $K = 4{\cdot}2$) and $R_{FT} = 78{\cdot}5$ cm. The flux trap is surrounded by 5 cm BeO converter, a critical fuel thickness of 6•6 cm and an outer wall. Fig. 4.19 shows the calculated flux distributions for such a burner reachtor. Note that the total flux in the fuel is similar to that in the flux trap. The calculated fluxes lead to the conclusion that

(total spectrum, flux trap) = $1/2{\cdot}(E{=}0.0253)$

The result indicates the relative effect of X on the ratio R of the FP transmutation rate to FP production rate for the reactor system. It can be seen that X should be kept as low as possible. Absolute results will depend on the Cs and Sr densities in the flux trap. A value of $R = 1$ was achieved for both FP nuclides at $X = 4{\cdot}5$. The F.P. atom ratio there was (Cs-137) / (Sr-90) = 7.25.

Another important problem is the relatively high flux in the outer zone or leakage from the core.

This flux can be used for two purposes:

1) for transmutation of other fission products which have rather a high absorption cross section e.g.

Tc-99 $\sigma^{th} = 22$ barns $t^{\beta}_{1/2} = 2.1 \times 10^5$ years

I-129 $\sigma^{th} = 28$ barns $t^{\beta}_{1/2} = 1.7 \times 10^7$ years

In both cases a flux of $10^{14} - 10^{15}$n cm^{-1}s^{-1} permits a rather effective transmutation rate of

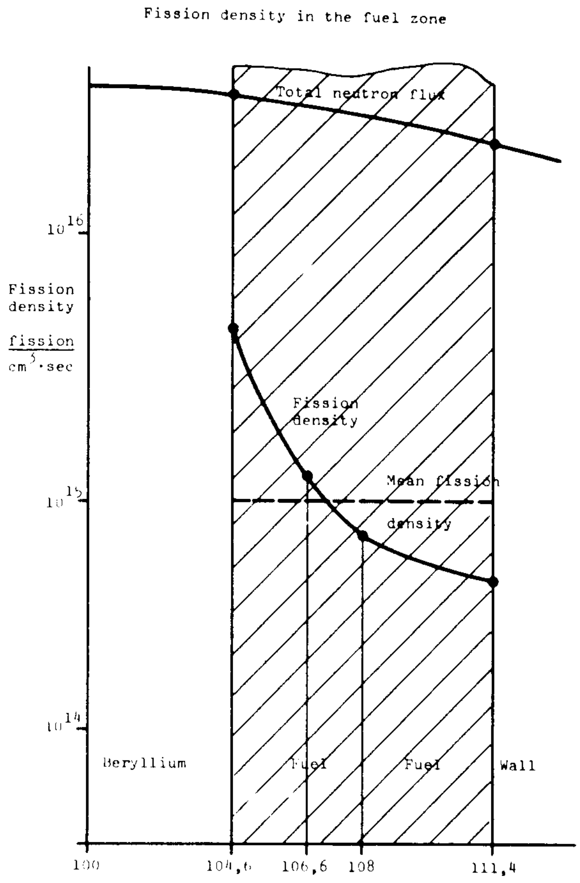
Tc-99 $\lambda = 7 \times 10^{-9}$s^{-1} $t_{1/2eff} = 3$ years

I-129 $\lambda = 7 \times 10^{-9}$s^{-1} $t_{1/2eff} = 2$ years

To still further improve these reactions the use of a beryllium moderator in the form of a 5 cm wall in the outer region of the core has been calculated. This gave an improvement in the transmutation rate of Cs-137 in the inner target region but also a very significant increase of specific power due to the scattering of the neutron flux in the fuel region.

The possibilities for transmutation of these two long lived fission products will be descussed further elsewhere.

Fig. 4.19 Fission density in the fuel zone



Remark: Fig. 4.20 omitted

The neutron flux outside the core may also be used for breeding in a uranium blanket. The breeding ratio may be higher than 1 but the decrease of the transmutation ratio is critical and seems to be too low for a burner reactor.

Nevertheless it could be useful to check these possibilities in more detail using some of the available neutrons for breeding in an external blanket region.

Table 4.9 summarises the data for the reference case. It can be shown that the transmutation rates obtained are equal:

$$^{90}\text{Sr:} \qquad \lambda_{eff} = \lambda_{tr} + \lambda_{\beta} = \sigma \cdot \Phi + \lambda_{\beta} = 1.19 \times 10^{-8} s^{-1}$$

$$t_{eff}^{1/2} = 1.85 \text{ yr}$$

$$t_{\beta}^{1/2} / t_{eff}^{1/2} = 15$$

$$^{137}\text{Cs:} \qquad \lambda_{eff} = 0.246 \times 10^{-8} s^{-1}$$

$$t_{eff}^{1/2} = 8.95 \text{ yr}$$

$$t_{\beta}^{1/2} / t_{eff}^{1/2} = 3.3$$

The results obtained are significant but rather pessimistic. To estimate the "profit" of the transmutation process, the hazard index (H) must also be taken into account. From Table 4.10 it can be seen that the total reduction in hazard from both fission products equals 13.5, which is a better indication.

4.6 Comments on hazard coefficients

It is perhaps valuable to estimate the usefulness of using the concept of hazard coefficients in fission product management. Table 4.10 gives some values for Sr-90 and Cs-137. From this it can be seen that in a steady state transmutations requiring the amount of hazardous substances is reduced by a factor ∿ 15 in relation to the steady beta-decay.

Table 4.9 The Transmutation Process for Selected Fission Products
(Total power $P = 30 \times 10^{9}$ W (burner reactor 7×10^{9} W); fission rate in
$= 9.3 \times 10^{20}$ fission/s.)

Property	Symbol	Unit	Data for	
			^{90}Sr	^{137}Cs
Yield of fission product[a]	Y	---	0.041	0.064
Production rate	$R_p = F \cdot Y$	atom s^{-1}	3.81×10^{19}	5.05×10^{19}
Concentration	C	atom/10^{24} cm^3 of target	0.0016	0.0116
Volume of target	V	cm^3	2.03×10^{6}	
Number of atoms	$N = V \cdot C$	atom	3.25×10^{27}	2.35×10^{28}
Decay constant	$\lambda\beta$	s^{-1}	7.86×10^{-10}	7.33×10^{-10}
Decay rate	$R = N\lambda\beta$	atom s^{-1}	2.55×10^{18}	1.72×10^{19}
Mean cross section	σ_c	10^{-24} cm^2	0.29	0.045
Total flux, mean	Φ	n cm^{-2} s^{-1}	3.83×10^{16}	
Transmutation rate	$R_{tr} = N \cdot \sigma_c \cdot \Phi$	atom s^{-1}	3.61×10^{19}	4.05×10^{19}
Total destruction rate	$R_t = R + R_{tr}$	atom s^{-1}	3.86×10^{19}	5.77×10^{19}
Destruction constant	$t^{1/2}_{eff} = \ln 2/\lambda_{eff}$	year	1.85	8.95
Efficiency of transmutation Inventory reduction ratio[b]	$E_{tr} = t^{1/2}_{\beta}/t^{1/2}_{eff}$	---	15	3.3
Steady-state equilibrium	$R_p \approx R$		3.8×10^{19}	5.8×10^{19}

[a] For the mean value of the yield, see Table 4.10

[b] See Fig. 4.21

Table 4.10 Hazard index for the Improved Reference Case*

Parameter	Symbol	Unit	^{90}Sr	^{137}Cs	Ratio $\frac{^{90}Sr}{^{137}Cs}$
Maximum permissible concentration					
water	H		3.7×10^{-1}	14.8	40
air	H		3.7×10^{-5}	2.2×10^{-3}	65
"mean"					50
Yield of fission products					
from ^{233}U	Y	atom/fission	0.062	0.066	
from ^{235}U	Y	atom/fission	0.051	0.0599	
from ^{239}U	Y	atom/fission	0.0218	0.0669	
Mean value for fission rate of ^{233}U:^{235}U:^{239}Pu 1:1:1	Y		0.041	0.064	
Hazard	H•Y	---	2.05	0.064	2.114
Efficiency of hazard reduction by transmutation	$H \cdot Y/E_{tr}$ (Table III)	---	0.137	0.019	0.156
Mean effective hazard reduction					$\frac{2.114}{0.156} = 13.5$

*The hazard coefficient (H) is defined as the amount of air and/or water needed to dilute the amount of a given nuclide present to levels proposed for the maximum permissible concentration.

Fig. 4.21 Inventory of Strontium-90 in the power reactors and in the burner reactor

Power of total system 55 GWth

Yield of Sr-90: 4,1 %

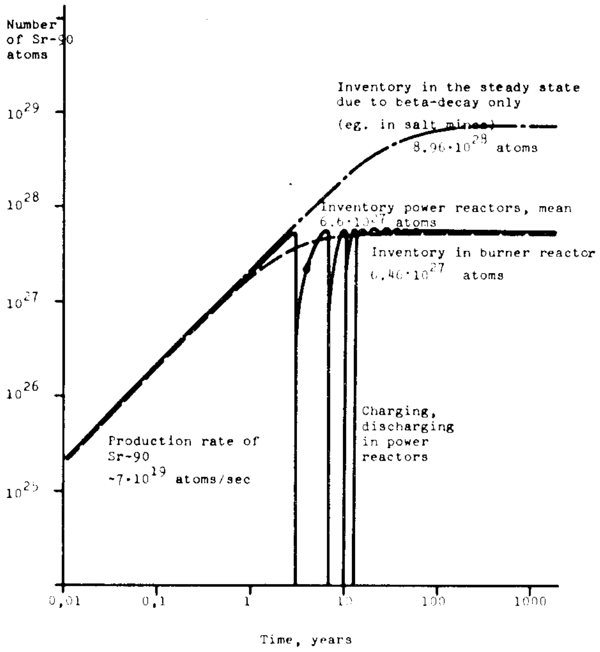
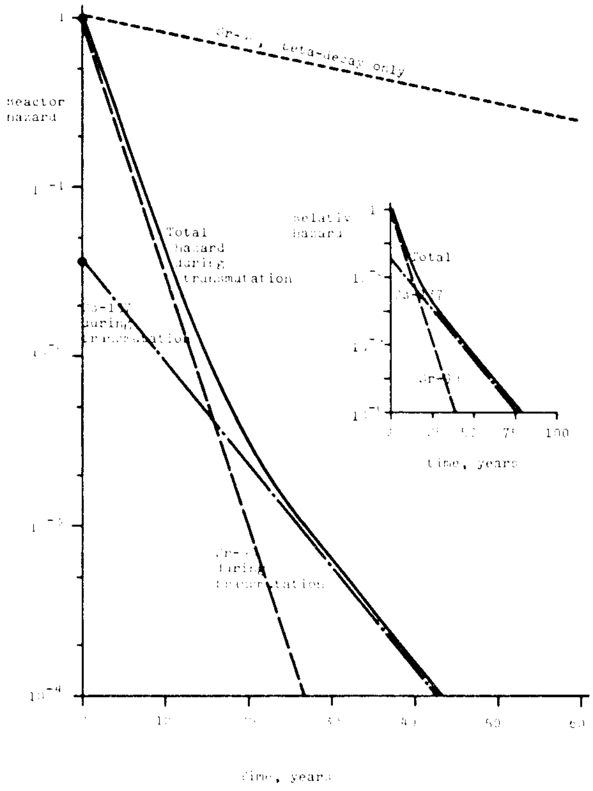


Fig. 4.22 Hazard of the both fission products: Sr-90 and Cs-137



The amount of strontium-90, the most hazardous nuclide, in the high flux burner is about the same as that found in the power reactors after 3 years of operation.

However the most impressive result comes from considering the end of the fission power area. Where by compared to storage (natural decay with a combined half life of 39 years) without transmutation, the transmutation case shows that the amount of nuclides remaining will be reduced by a factor of 1000. (Fig. 4.22).

In a high flux transmutation the reduction by a factor 1000 would be achieved with 26 years in the lifetime of one reactor generation.

4.7 Secondary processes

It must be remembered that the relatively high neutron flux results in the irradiation not only of the non-desirable radioactive nuclides but also the stable fission products, including the stable components of the fuel and structural material. A very short review of these partivular processes is given here.

Natural chlorine contains two stable isotopes (underlined)

$$\begin{array}{ll} {}^{35}_{17}\underline{\mathrm{Cl}}\ (n,\gamma)\ {}^{36}_{17}\mathrm{Cl} \xrightarrow[3.1\times10^{5}\ \text{years}]{\beta^-} {}^{36}_{18}\underline{\mathrm{Ar}} & \\ & \text{volatile} \\ {}^{37}_{17}\underline{\mathrm{Cl}}\ (n,\gamma)\ {}^{38}_{17}\mathrm{Cl} \xrightarrow[37{,}3\ \text{minutes}]{\beta^-} {}^{38}_{18}\underline{\mathrm{Ar}} & \end{array}$$

However not only (n,γ) reactions are important here. Much more important is the following raction

$${}^{35}_{17}\underline{\mathrm{Cl}}\ (n,p)\ {}^{35}_{16}\mathrm{S} \xrightarrow[8.7\ \text{days}]{\beta^-} {}^{35}_{17}\underline{\mathrm{Cl}}$$

(see ch. 7 for experiment results)

The presence of sulphur also influences the chemistry of the molten fuel (see ch.6).

Sodium having only one stable isotope is also transformed

$$^{23}_{11}\text{Na}\ (n,\gamma)\ ^{24}_{11}\text{Na} \xrightarrow{\beta^-} {}^{24}_{12}\text{Mg}\ (n,\gamma)\ ^{25}\underline{\text{Mg}}\ (n,\gamma)\ ^{26}\underline{\text{Mg}} \longrightarrow$$

$$\longrightarrow\ ^{26}\underline{\text{Mg}}\ (n,\gamma)\ ^{27}\text{Mg} \xrightarrow{\beta^-} {}^{27}\underline{\text{Al}}$$

This chain gives rise to increasing amounts of stable magnesium and over larger periods also stable aluminium.

4.8 Conclusions

The system proposed for the transmutation of ^{90}Sr and ^{137}Cs, fulfills the following criteria:

1. The energy balance is positive.

2. The hazardous waste balance is strongly negative. That is, the amount of hazardous material destroyed greatly exceeds the amount of freshly produced, e.g. tritium, ^{10}B, and the activation products of the structural material.

3. The rate of destruction (transmutation) is approximately at least one order of magnitude greater than that due to spontaneous beta decay.

4. The period in which a thousand-fold reduction in the hazard can be achieved is the same as the lifetime of one reactor, that is, 20 to 40 yr.

5. The neutron balance of the system is positive. That is, it permits breeding to occur along with the transmutation.

6. The weakest feature is shown in the relationship between the probability of catastrophic release to the environment for the transmutation operation, P_{trans}, to the probability of a similar event in the case of storage, P_{store}, where it is desired that

$$\frac{P_{trans}}{P_{store}} < \frac{trans}{store} = \frac{\sigma \cdot \Phi + \lambda_{\beta}}{\lambda_{\beta}}$$

where λ_{β} is the decay constant.

7. Further optimisations of the system are possible.

8. The proposed system, a molten-salt fast reactor, while rather exotic from a technological point of view, is not as far removed from the present state of technology as some other transmutation proposals (e.g. high-flux high-energy accelerators, controlled thermonuclear reactors) may be.

5. An internally cooled breeder with uranium-plutonium fuel

5.1 Design features and objectives

In this chapter a further variant of the molten salt breeder is described.

The core is cooled by a cooling medium circulating in tubes. The molten fuel is intensively miced. It remains in the core and no fuel other than that being drawn off for reprocessing is present outside the core.

The unique feature of this concept is the use of a molten fertile material as the cooling medium for the core.

Such a dual function for the fertile coolant results in some unusual properties for this reactor type. The study here is concerned with a molten chloride breeder reactor. The most important features are: (see table 5.1)

- thermal power: 2.05 GW - 1.94 GW in core + 0.11 GW in the blanket

- electrical power: 0.85 GW in the optimum case (η_{eff} = 0.4)

- molten fuel consisting of (in mol %)

15% $PuCl_3$ (of which Pu-239 + Pu-241 = 80% and Pu-240 = 20%)

85% NaCl (no $^{238}UCl_3$ in fuel)

and fission products in the form of chlorides or in an elementary state.

- molten fertile material (in mol %)

65% $^{238}UCl_3$

35% NaCl

and newly bred $PuCl_3$ and fission products

- coolant flowing in tubes: made up of the fertile material above. No other coolant in core.

- blanket material: also made up of the fertile material above

- the core is internally cooled. There is no circulating fuel outside the core.

Table 5.1

OBJECT: INTERNAL COOLED FAST BREEDER POWER REACTOR

REACTOR TYPE :
GEOMETRY: INTERNAL ZONE : CORE, Fuel
(M) WALL : Iron, Molybdenum
INTERMEDIATE ZONE:
WALL : cooling tube with fertile material
EXTERNAL ZONE : Fertile zone
WALL, REFLECTRO : Iron

POWER (GW thermal) : 2.1
POWER DENSITY (GW therm/m^3 core) : 0.22
NEUTRON FLUX, MEAN (n/cm^2s) : 7 x 10^{15}

FISSILE NUCLIDE: Pu-239/241
DILUTENT : NaCl, UCl_3
COOLING SYSTEM : Internal, in tubes by fertile material
BREEDING RATIO : 1.38

PARAMETER STUDIED : Design
: Tube materials
: Pu/U ratio
: Temperature

METHOD OF NEUTRONIC : ANISN S4
CALCULATION : 23 Groups
: P_1 APP.
: GCC3 CODE
END

- the fuel and coolant flow concurrently (Fig. 5.1)
- the reprocessing plant is in close proximity to the reactor ("under same roof")
- the fuel in the core, and the coolant is pumped with a velocity of 2 and 9 $m.s^{-1}$ respectively.
- structural material: possibly molybdenum along with small amounts of other metals e.g. Ni, Fe.

The advantages of the proposed reactor are as follows -

- no separate coolant, no "foreign" cooling agent (e.g. sodium, helium etc.) in the core which results in a more satisfactory system with improved neutron balance.
- the fuel inventory is very small due to lack of a separate cooling system and the small out of core inventory due to the directly coupled reprocessing plant.
- the fuel contains only plutonium and no uranium which simplifies the processing technology and removes the danger of uranium trichloride oxidation which also improves the corrosion properties of this medium
- the high velocities of both fuel and coolant, significantly reduces the temperature gradients in the equilibrium state and reduces the mass transport mechanisms. These are very sensitive to temperature gradients and play a large role in corrosion.

However the diadvantages are numerous:

- the first and most improtant is of course corrosion. The molten chloride medium especially in neutron and gamma fields, at high temperatures and velocities with chlorine being virtually freed in the fission process from plutonium chloride presents a very serious problem which must, and probably could be solved.
- the most likely structural material seems to be molybdenum alloy which among other thing gives rise to parasitic absorption of neutrons.
- the fuel is circulated by a pump which must be located in or close to the core which increases the corrosion problems.
- the high fuel and collant velocities result in high pumping costs and could cause severe erosion.

Fig. 5.1 Circuit schematic

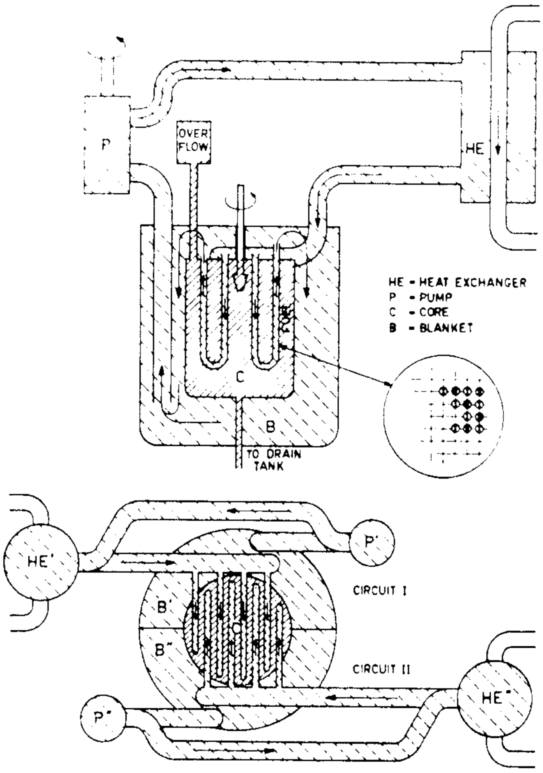


Table 5.2 Molten chloride fast breeder reactor (MCFBR) "CHLOROPHIL"

Electrical power, approx	MW(elect)	800
Thermal power, total/in core	MW(thermal)	2050/1940
Core volume	m^3	8.75
Specific power	MW m^{-3}	220
Core geometry	m	height2.02/2.36 dia.
Fuel: liquid PuCl$_3$ NaCl	mol%	16/84
Liquidus/boiling point	^{o}C	685/1500 (appr.)
Fuel mean temperature	^{o}C	984
Fuel volume fraction in the core		0.386
Power form-factors radial/axial		0.60/0.78
Fast flux, mean across core	n cm^{-2}s^{-1}	7 x 10^{15}
Fuel density at 984^{o}C	kg m^{-3}	2344
Heat capacity, 984^{o}C	kJ Kg^{-1} K^{-1}	0.95
Viscosity, 984^{o}C	g cm^{-1}s^{-1}	0.0217
Thermal conductivity, at 750^{o}C	W cm^{-1} K^{-1}	0.007
Fuel salt in core	kg	7900
Total plutonium in core/in system	kg	2900/3150
Plutonium in salt	weight %	36.4
Mean plutonium specific power	MW(th) kg^{-1}	0.67
Mean plutonium specific power in entire system	MW(th) kg^{-1}	0.62
Coolant liquid U-238 Cl$_3$/NaCl	mol %	65/35
Liquidus/boiling pint	^{o}C	710/1700
Coolant temperature inlet/outlet	^{o}C	750/793
Coolant volume fraction in the core		0.555
Coolant density	kg m^{-3}	4010
Coolant salt in core/in blanket	kg	19,500/165,000
Total volume (95 cm)	m^3	47.85
Fuel (shell side, pumped), velocity	m s^{-1}	2
Coolant velocity	m s^{-1}	9
Number of coolant tubes		23,000
Tubes inner/outer diameter	cm	1.20/1.26
Tubes pitch	cm	1.38
Breeding ratio, internal/total	-	0.716/1.386
Doubling time, load factor 1.0/0.8 yr.	years	8.5/10.5

Mean flux across core $7.0 \times 10^{15} n\ cm^{-2}s^{-1}$
in centre $1.2 \times 10^{16} n\ cm^{-2}s^{-1}$

Temperature coefficient of reactivity $\delta k(\%)/\delta T(^{o}C)$
- fuel $- 3.8 \times 10^{-2}$
- coolant $+ 1.29 \times 10^{-2}$

Reactivity wall fo vessel 0.72% (10 mm thickness).

5.3 Neutron Physics (according to J. Ligou, 1972)

The 22 group transport calculation gives 125 cm (8-18 m^3) for the critical radius of the core with a blanket thickness of 95 cm (36.42 m^3). The detailed neutron balance is given in table 5.3.

The relative fluxes for each group are to be found in table 1.3 for core centre and core boundary. The corresponding one group cross sections are given in table 5.4. In Fig. 5.2 the neutron spectra (flux per lethargy unit) are compared to that of the fast critical facility ZPR - 3 - 48. This last spectrum is slightly harder but the spectrum of the molten chlorides fast breeder compares facourably with that of a power LMFBR. From table 5.3 one can deduce the following parameters

$k_\infty = 1.385$

Breeding ratio $Br_{core} = 0.716$
$BR_{blanket} = 0.670$
$BR_{total} = 1.386$

For the given core (125 cm radius) the blanket thickness was varied between 75 cm and 115 cm. Fig. 5.4 shows the variation of breeding ratios obtained from the new transport calculations. The reactivity and the core breeding ratio remain practically constant in this rankge making the adjustment of the core volume unnecessary.

Above 100 cm improvement of the breeding ratio by increasing the blanket thickness gives a poor return. For example to increase the breeding ratio from 1.40 to 1.45 requires a thickness increase of 20 cm or a blanket volume increase of 32%.

Table 5.3

Neutronics of internal cooled fast breeder

Core atomic densities (atoms $\times 10^{24}$)

Pu-239	6.6797×10^{-4} atoms cm^{-3}
Pu-240	1.6699×10^{-4}
U-238	3.5629×10^{-3}
Cl	1.9495×10^{-2}
Na	6.3017×10^{-3}
Mo	7.386×10^{-4}
Fe	5.078×10^{-3}

Blanket (coolant) densities (atoms $\times 10^{24}$)

U-238	6.4023×10^{-3} atoms cm^{-3}
Cl	2.2718×10^{-2}
Na	3.457×10^{-3}

Neutron Balance

Region	Nuclide	Atoms $(cm^3 \times 10^{21})$	Absorption		Leakage	Production
Core	^{238}U	3.5629	25.50	(n,γ) 22.51 (n,f) 2.99		8.23
	^{239}Pu	0.66796	34.56	(n,γ) 5.58 (n,f) 28.98		85.55
	^{240}Pu	0.16699	3.78	(n,γ) 2.24 (n,f) 1.54		4.72
	Na	6.3017	0.26			-
	Cl	19.495	3.16	(in fuel 1.10) (in cool. 2.06)		-
	Fe	5.078	1.30			-
	Mo	0.7386	2.04			-
	F.P.	0.0697	0.50			-
Total core			71.10		27.40	98.50
Blanket	^{238}U	6.42	23.70	(n,γ) 23.15 (n,f) 0.55		1.50
	Na	3.457	0.08			-
	Cl	22.72	2.22			-
Total blanket			26.00		2.9	1.50

Fig. 5.2 Chlorine absorpt. cross-sections ENDF B/III (Jan. 1972)
Absorption cross-section of chlorine

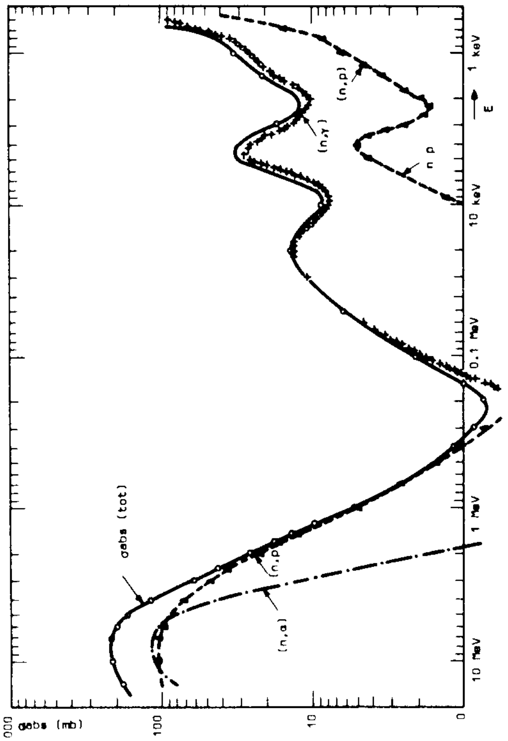


Fig. 5.3 Neutron Spectra and Chlorine Cross Sections

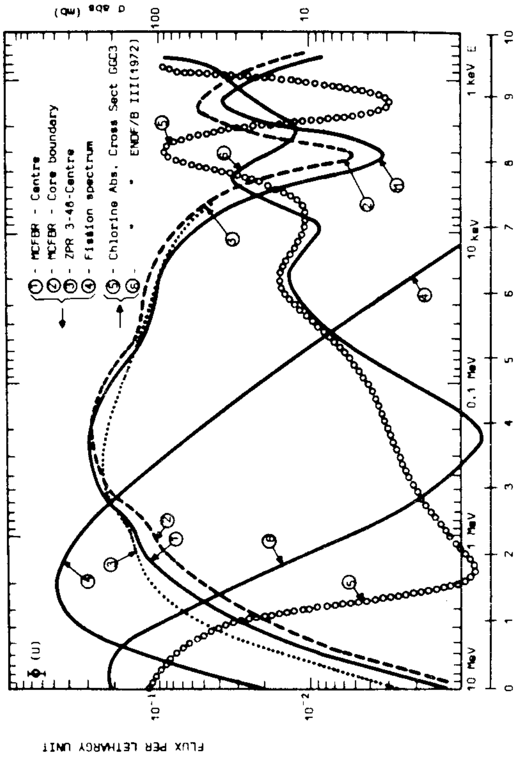


Fig. 5.4

Breeding Ratio C and blanket thickness t
(Core volume = 8.18 m³)

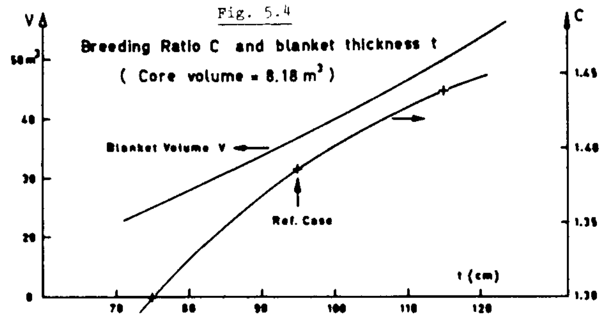


Fig. 5.5

Reactivity change with Core volume
(Blanket Thickness = 95 cm)

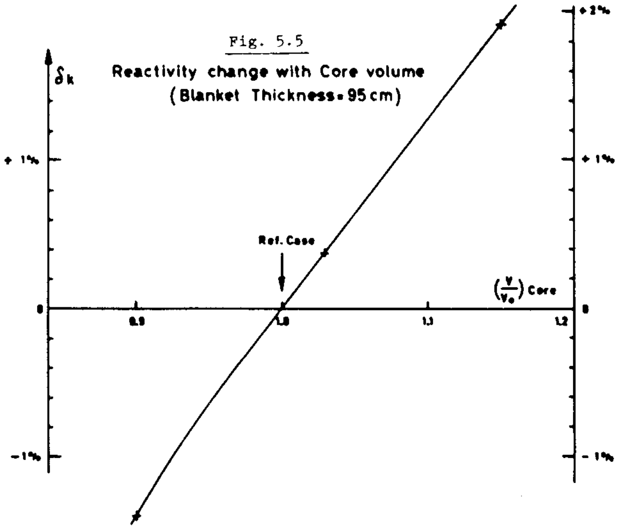


Fig. 5.6

Radial distributions of the 10th and 12th flux (Sph. Geom.)
—— transport calculations
---- Asymptotic mode in the core

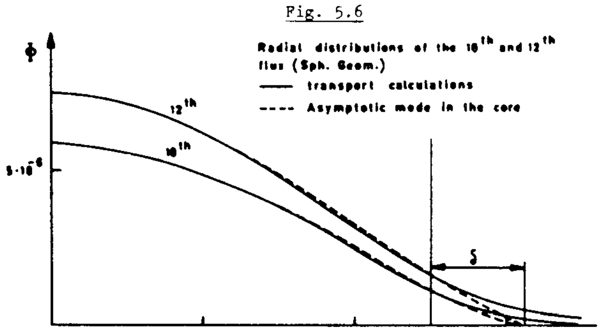
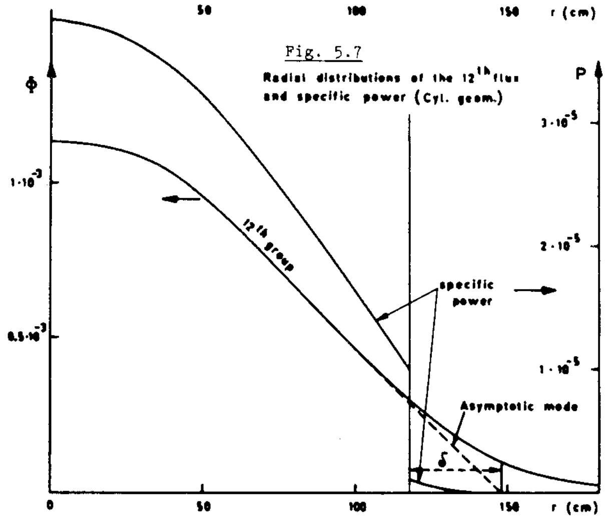


Fig. 5.7

Radial distributions of the 12th flux and specific power (Cyl. geom.)



On the other hand the reactivity depends on the core radius. Fig. 5.5 shows the variation of reactivity with increasing core size. Such a curve is very useful when it is required to translate the cost in reactivity of a supplementary parasitic absorbtion into an increase in core volume (or plutonium inventory). The transport calculations used in Fig. 5.5 could not be used to obtain the corresponding variations in breeding ratio, rather they have been calculated from the information in table 5.3. (assuming small core volume variation < 10% and no important changes in spectra).

This gives

1) $BR_{total} \simeq 1.386 - 2.45\ \delta k$ (breeding in core unchanged)

The effects of modifying the core composition can be evaluated by the same method. This is arranged to avoid altering the properties of the coolant and fuel by varying the coolant tube diameter. The reactivity is very sensitive to this parameter. If ε and η are respectively the relative increases in fuel

Table 5.4 One group cross sections based on the reference 22 group transport calculation

(barn = $10^{-24} cm^2$)

Type of Nuclide	Core Spectrum			Blanket Spectrum		
	σ_c	σ_f	$\bar{\nu}$	σ_c	σ_f	$\bar{\nu}$
^{238}U	0.249	$3.30\ 10^{-2}$	2.746	0.326	$7.71\ 10^{-3}$	2.715
^{239}Pu	0.329	1.709	2.951	0.536	1.841	2.917
^{240}Pu	0.527	0.364	3.052	0.790	0.174	2.998
Na	$1.62\ 10^{-3}$	-	-	$2.044\ 10^{-3}$	-	-
Cl	$6.38\ 10^{-3}$ $(8.64\ 10^{-3})$*	-	-	$8.83\ 10^{-3}$ $(9.11\ 10^{-3})$*	-	-
Fe	$1.01\ 10^{-2}$	-	-	$1.43\ 10^{-2}$	-	-
Mo	0.109	-	-	0.166	-	-
FP	0.225	-	-	0.362	-	-

* These values were computed on the bases of ENDF/B. III data.

and coolant volume for a constant pitch are as

$$\eta = -1.44\ \varepsilon$$

$$\text{and } k \simeq \frac{1 + 8.23 \times 10^{-2}\varepsilon + 0.9027\ \eta}{1 + 0.2756\varepsilon + 0.3944\ \eta}$$

which gives

$$\delta k \simeq -0.924\ \varepsilon$$

$$\eta = 1.56\ \delta k$$

The corresponding relationships for breeding ratios (BR) are

$$BR_{core} \simeq 0.716 - 1.72\ \delta K$$

$$BR_{blanket} \simeq 0.670 - 1.05\ \delta K$$

$$BR_{total} \simeq 1.386 - 2.77\ \delta K$$

It can be seen that the penalty on the total breeding ratio for the same δK is only slightly greater while the penalty on the increase of plutonium inventory is five times less. Therefore a reduction in the diameter of the coolant tubes is preferable to an increase in coolant diameter, provided of course that an increase in coolant velocity is admissible. This last assumption is implicit in these calculations since the coolant density was kept constant.

In the whole system the chlorine absorption represents 5.38%. We have seen that the GGC-3 values are different from the more up to date ones (ENDF/B-III). On the basis of these new cross sections given in Fig. 5.3 (curve 6) and assuming that the reference spectrum is unchanged, a computation of the one group corss sections gives 8.46 mb instead of 6.38 mb in the core, and 9.11 mb instead of 8.83 mb in the blanket. In this last region the spectrum is softer and the increase of cross sections in the energy range (E ∿ 0.6 MeV) is almost compensated for by the decrease at the lower energies (10 keV - 0.6 MeV).

The total absorbtion by chlorine for the whole system is 6.57% instead of 5.38% giving a loss of reactivity of 1.2%. This loss could be replaced by a 1.9% increase in Pu inventory if a very small decrease (o.65%) of the coolant tube diameter is accepted. Otherwise by changing only the core radius a greater increase of Pu inventory (10%) is required (Fig. 5.5)

Even with this latest data the problem of parasitic capture in the chlorine is not dramatic and there is no reason to believe that there is no need to enrich the chlorine ^{37}Cl. This is consistent with the conclusions of Nelson. Fig. 5.3 clearly shows the importance of the energy distribution (spectrum corresponds to the chlorine cross section minimum (65% of neutrons are in an energy range where $\sigma_{Cl} \leq 5$ mb). This fact was not perhaps recognised 15 years ago when fine spectrum calculations were not possible. This could explain the pessimistic conclusions of several eminent physicists.

For the molbydenum alloy chosen (20% Mo) the reactivity penalty (2%) is quite acceptable. However the cost could rapidly become prohibitive if the volume of the structural material and/or the molybdenum content should increase for design reasons. In the context of more detailed design studies this point may become more important than the definition of the proper chlorine cross-sections. It does however seem likely that the molybdenum cross sections used in GGC-3 were overestimated.

In the simplified calculations, no core vessel was allowed for at the core/blanket boundary but all the required information is available - fluxes, one group cross sections etc. (Table 5.4). Using the same alloy for the vessel (20% Mo) the one group macroscopic absorbtion corss section is $2.56 \times 10^{-3} cm^{-1}$ giving a loss of reactivity:

$$\delta k(\%) = 0.721\ e$$ where e is vessel thickness in cm.

For 19 mm thickness a value of 1.37% is obtained for loss of reactivity which would have to be compensated for by an increase of core volume of about 10% i.e. about 9 m^3 instead of 8.18 m^3. A better solution would be an increase in the plutonium inventory of about 2.1% in the reference core (loss in breeding ratio 1.386 → 1.350).

Transport calculations (GGC-3 + SHADOK) have been made for different coolant fuel densitites, different temperatures (in this case with the density constant to determine only the Doppler effect). The reactivity changes with respect ot the reference core are given in table 5.5.

Table 5.5 Reactivity changes

Type of modification	Parameter	Reactivity changes
Fuel density	- 5%	-3.0% { -1.9 (k_∞) -1.1 (leakage)
Coolant density	- 5%	+0.77% { +2.0 (k_∞) -1.23 (leakage)
Fuel temperature	+300°C	0.01%
Coolant temperature	+300°C	-0.14%
Full loss of coolant	-	+12.0% { very high on k_∞ (78%)

The partial changes in k_∞ or leakage are only approximate but the total reactivity changes are evaluated directly and are therefore more precise.

The Doppler effect in the fuel is quite negligible due to compensation between the capture and fission processes.

The effect of full loss of coolant is large and positive but considerably lower than might be expected from crude calculations (k_∞ changes in the reference spectrum).

From table 5.5 one can deduce the feed back effect which is very important for kinetic studies

$$\frac{\delta k}{\delta k}(\%) \simeq 60\left(\frac{\delta\rho}{\rho}\right)_{fuel} - 15\frac{(\delta\rho)}{\rho}_{fuel} - 15\frac{(\delta\rho)}{\rho}_{coolant} - 4.8\times10^{-4}\ \delta T_{(coolant)}$$

The void coefficient of the fuel (1st therm) is strongly negative. 1% voild

$$\left(\frac{\delta\rho}{\rho} = -\ 10^{-2}\right)$$

gives a 0.6% loss in reactivity. If boiling occurs in the fuel it will be rapidly arrested by a decrease in reactor power.

If one considers that all density modifications come from thermal expansions (liquid phase only) one can define general temperature coefficients.

The thermal expansion coefficients are

$$\left(\frac{\delta\rho}{\rho\delta T}\right)_{fuel} = -0.63 \times 10^{-3} \text{ K}^{-1}$$

$$\left(\frac{\delta\rho}{\rho\delta T}\right)_{coolant} = -0.89 \times 10^{-3} \text{ K}^{-1}$$

Replacing these values in above given equation leads to the following expression

$$\frac{\delta k}{k}(\%) \simeq -3.8 \times 10^{-2} \ (\delta T)_{fuel} + 1.29 \times 10^{-2} \ (\delta T)_{coolant}$$

In the second term the part played by the Doppler coefficient (4.8×10^{-4}) is quite negligible. For + 100°C in the fuel the loss of reactivity is - 3.8% which is very important from the safety point of view. Compared to any kind of power reactor (even the BWR) the advantage of this kind of reactor is quite evident.

For the Nelson (1967) value of the thermal expansion -3.10^{-4} instead of -6.3×10^{-4} one gets - 1.8% which is very close to the Nelson result -1.5%.

If we postulate an accident condition and assume that the same increase of coolant temperature immediately follows the fuel temperature rise, the overall change in reactivity is defined by:

$$\frac{\delta k}{k}(\%) \simeq -2.51 \times 10^{-2} \ \delta T$$

This important isothermal and pessimistic coefficient is still negative. Nevertheless during a detailed study of this reactor concept it would be necessary to check the values of the thermal expansion coefficient for fuel and coolant more carefully. The relative value of the coolant term which is positive might prove to be too high if the differences between fuel and coolant became too marked. This problem did not arise with the present data.

In a spherical assembly the fluxes in the core are given to a good approximation

$$\Phi(r,E) \simeq \frac{\sin Br}{Br} f(E)$$

where the space function is called the "fundamental mode" (solution of

$$\nabla^2\Psi + B^2\Psi = 0$$

in spherical geometry). The critical buckling B^2 is obtained from homogeneous calculations based only on the cross section data of the core, $f(E)$ is the asymptotic spectrum which is space independent far from the core boundary. For the same 22 energy groups one gets: $B^2 = 4.08 \times 10^{-4}\ \mathrm{cm}^{-2}$. Using this value a good fit of the "exact fluxes" have been obtained from the complete transport calculations (Fig. 5.6). The asympototic fluxes cancel for

$$r = R_e = \frac{\pi}{B} = 155.5\ \mathrm{cm}$$

where R_e is the extrapolated radius. By definition the blanket saving is given by

$$\delta = R_e - R_c = 30.5\ \mathrm{cm}$$

where R_c is the core critical radius. The blanket saving depends mainly on the nuclear properties of core and blanket and on blanket thickness. However for thicknesses greater than 60 cm this last effect is very weak. Finally the shape and size of the core have almost no influence on this saving. This parameter, for this reason so important in reactor physics, will be used in the next section for the one dimensional cylindrical calculations.

The axial blanket thickness is taken to be equal to the radial thickness (95 cm), and the core height as $H_c = 200$ cm. The critical radius of this cylindrical core has to be determined. The two dimensional transport calculations are too expansive (and unsafe) and only one dimensional calculations have been made, which is sufficiently accurate. Axial transport calculations are not required since the balnket saving is known from the spherical geometry calculations. One can therefore assume the following flux shape.

$\Phi(r,Z,E) \simeq (\cos \beta Z \Psi(r,E)$ for any

r value (including the radial blanket) and

$$-\frac{H_c}{2} \leq Z \leq \frac{H_c}{2}$$

$-\beta^2$ is the axial buckling computed from the extrapolated height: $H_e = H_c + 2\delta = 261$ cm which gives

$$\beta = \frac{\pi}{H_e} = 1.204 \times 10^{-2}\text{cm}^{-1}$$

$$(\beta^2 = 1.450 \times 10^{-4}\text{cm}^{-2})$$

The computation of core radius and spatial distribution have been made with the SHADOK code (cylindrical version) by introducing axial leakage defined by β^2. Before that a first approximation is obtained by introducing the radial buckling α^2, that is to say assuming for the core only, the shype $\Psi(r,E) \simeq J_0(\alpha r)f(E)$ where J_0 is the usual Bessel function. One obtained $\alpha^2 = B^2 - \beta^2$ where the critical total buckling is 4.08×10^{-4} cm^{-2} which gives $\alpha^2 = 2.63 \times 10^{-4}$ cm^{-2} and $\alpha = 1.625 \times 10^{-2}$ cm^{-2}. Then the extrapolated radius of the cylindrical reactor is

$$R_e = \frac{2.405}{\alpha} = 148.0 \text{ cm.}$$

Finally with the previous blanket saving we get a core radius proper of $R_c = 117.5$ cm. The direct transport calculations with SHADOK-code gives $R_c = 118$ cm! This clearly indicates the value of the blanket saving concept. Nevertheless these transport calculations are still necessary because they give the radial distribution of fluxes and specific power over the whole system and more detailed informations. Fig. 5.6 shows some of the radial distributions of flux and specific power. The energy production in the blanket is quite small (1.7% of the core power) because no fissile materials are present (and only fast fissions occur in ^{238}U). In practice it would be higher (say 5%) since the reprocessing process would not be able to remove all the fissile nuclides produced even with continuous fuel (coolant) reprocessing.

The radial form factor for specific power distribution is, for this core

$$\alpha_r = \frac{\text{mean power}}{\text{maximum power}} = 0.60$$

Note; it would be possible to improve this coefficient by choice of different lattices particularly the most reactive at the peripheral region.

The axial distribution of specific power is given with a good approximation. If the axial mean value is unity then this distribution is:

$$P(Z) = \frac{\beta H_c}{2\sin\frac{\beta H_c}{2}} \cos \beta Z$$

$$\text{with } \beta = \frac{\pi}{H_e} = \frac{\pi}{H_c + 2\delta} = 1.2 \times 10^{-2}\ \text{cm}^{-1}$$

and H_c = 200 cm giving

$$P(Z) = 1.284 \cos\pi \frac{Z(\text{cm})}{261}; \ (-100 \leq Z \leq 100)$$

and α_Z = 0.78 for the axial form factor.

This axial distribution is very close to that used by Nelson.

The critical volume is higher for cylindrical geometry 8.75 m^3 compared to 8.18 m^3: this increase was expected.

Usually the number of energy groups required for a good definition of neutron spectrum is at least 12 for fast critical assembly studies and 22 can be considered desirable. Therefore a 22 group cross section set has been prepared, the code GGC-3 which allows 99 group calculations for a rather simple geometry has been used for this condensation. The cross sections were produced separately for core and blanket and the scattering anisotropy was limited to P_1 which is sufficient for this reactor type. Most of the GGC-3 library data were evaluated by G G A before 1967 but some are more recent.

- Iron - evaluated from ENDF/BI data (Feb. 1968)

- Molybdenum - evaluated from isotopes of ENDF/BI data (July 1968)

- Plutonium 239 - evaluated from KFK -750 Resonance Nuclide (Feb. 1969)

New data concerning chlorine absorbtion cross sections are available at EIR (Fig. 5.2) they are obtained from ENDF/B-III (Jan. 1972). Unfortunately this information came too late to be used for the transport calculations. Fig. 5.3 (curves 5 and 6) shows that the GGC-3 values were underestimated above 0.6 MeV and overestimated between 10 keV and 0.6 MeV. The effect of this on the reactivity is not great. Taking also the molybdenum cross section from ENDF/B-III one can see that the GGC-3 values are too high. (experiments made with molybdenum control rods in fast critical assemblies could not be reproduced with ENDF/B-I which makes a new evaluation of data necessary).

Fission product data are from Bodarenko (1964). The absorbtion cross sections for resonent nuclides are obviously shielded. The Nordheim (1961) theory is included in the GGC-3 code, it requires some special data as shown in table 5.6.

Table 5.6

Resonant Nuclide R.	Atomic density N_R $(\times 10^{24})$	T (K)	a_R	C_R	σm^{R}_{1}	σm^{R}_{2}	σm^{R}
^{238}U (Coolant)	6.42×10^{-3}	1042	0.58	0.925	2.15	7.10	20.3
^{239}Pu (fuel)	1.725×10^{-3}	1257	0.40	0.83	26.3	23.0	178.3
^{240}Pu (fuel)	4.32×10^{-4}	1257	0.40	0.83	105.0	111.7	731.7

5.4 Safety Problems, Comments

The molten chlorides reactor seems to be a relatively safe system due to the following rasons

- an extremely high negative temperatur coefficient of reactivity, since during a temperature rise part of the liquid fuel is pushed out of the core into a non-critical geometry buffer tank. The dumping of fuel in case of an incident is also possible in an extremely short time.

- in a more serious incident when the fuel temperature increases to 1500-1700°C (depending on external pressure) the fuel begins to boil. The vapour bubbles give rise to a new and unique, very high negative "fuel void effect"

- the leakage of fuel to the coolant is probably not a serious problem because the coolant is continuously reprocessed.

- the leak of coolant to the fuel for the same reason cannot cause large problems (provided the leak remains small).

A rahter adverse property of such a molten fuel reactor is the need to initially heat the solidified fuel in a non critical geometry with external power. (e.g. from the electrical grid). This problem has been fully overcome in the case of the molten fluoride thermal reactor (Oak Ridge National Laboratory).

6. CHEMICAL AND RELATED PROBLEMS

6.1 Physical and chemical criteria for salt components

The limiting criteria in the search for fuel, fertile material and coolants for internally cooled systems are as follows

1. small elastic scattering for fast neutrons (Fig. 6.1)
2. small inelastic scattering.
3. low neutron capture cross-sections for fast neutrons
4. thermodynamic and kinetic stability of plutonium and uranium compounds (Fig. 6.2, 6.3, 6.4).
5. melting point below 700°C in the pure state or in the dissolved state (Fig. 6.5, 6.6, 6.7, 6.8, 6.9, 6.10).
6. boiling point above 1500-1600°C for both pure and dissolved states (low vapour pressure) (Fig. 6.11).
7. stability against atmospheric constituents, oxygen, water carbon dioxide. (Fig. 6.12).
8. good heat transfer properties and specific heat capacity (low viscosity, high conductivity etc.) (Fig. 6.13)
9. good corrosion properties if possible (Fig. 6.14)
10. adequate technological or laboratory experience.
11. relatively cheap.

These wide ranging criteria are fulfilled best by the following compounds

$PuCl_3$, UCl_3, NaCl (Table 6.1, 6.2)

Fig. 6.2

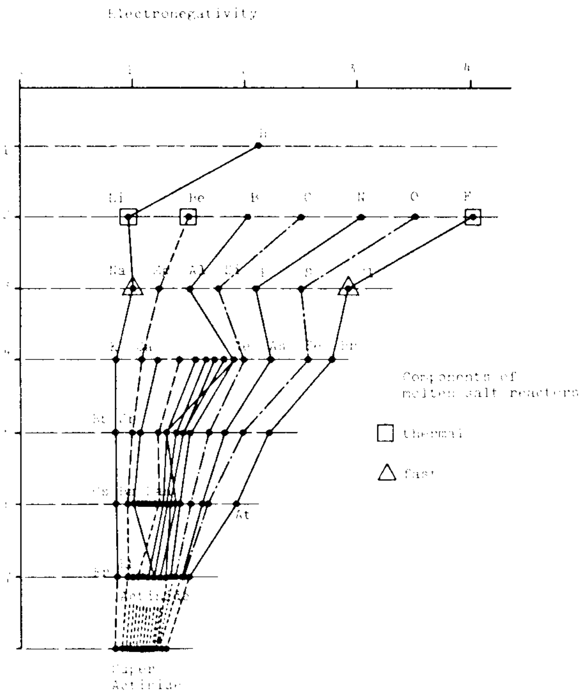
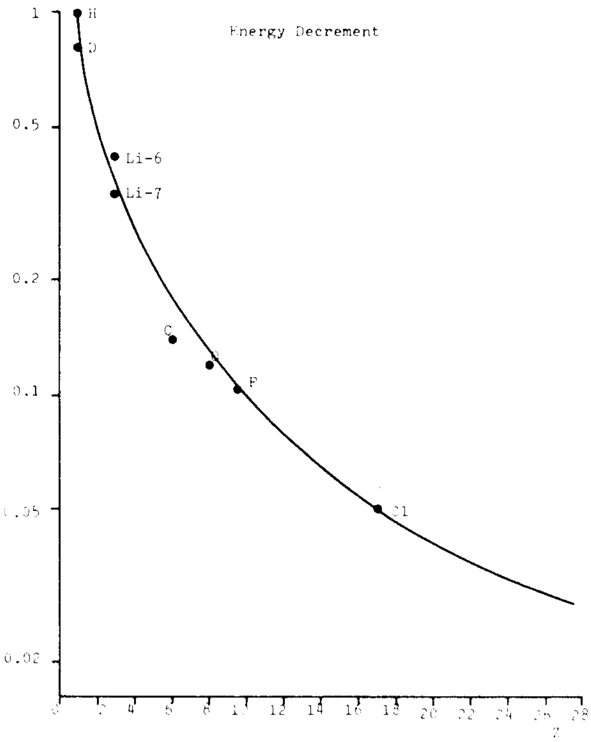


Fig. 6.1



Numerical Values given for ΔF^{1000K} in 2LiF-BeF$_2$ solution

Fig. 6.3

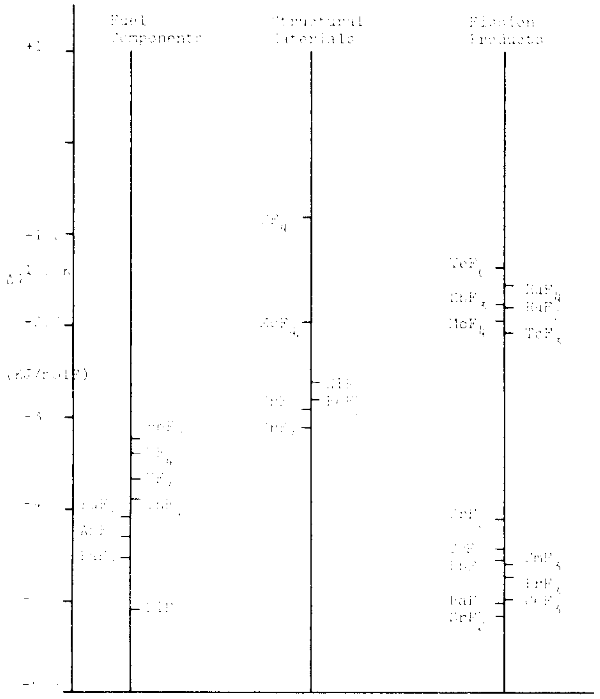


Fig. 6.4

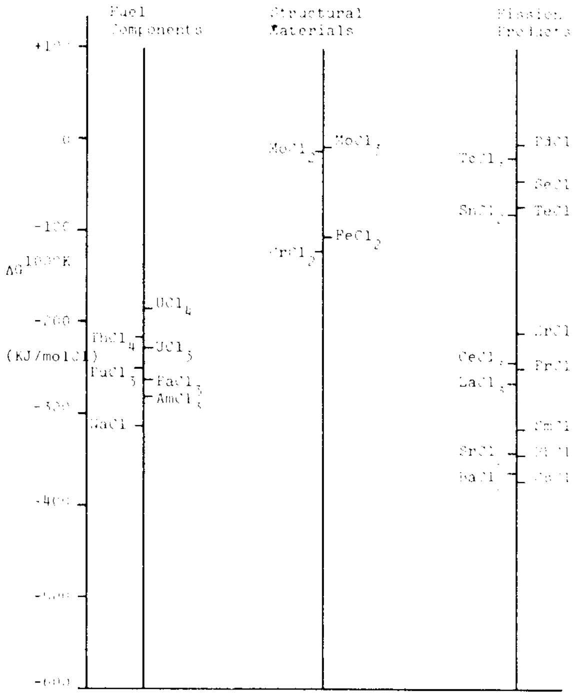
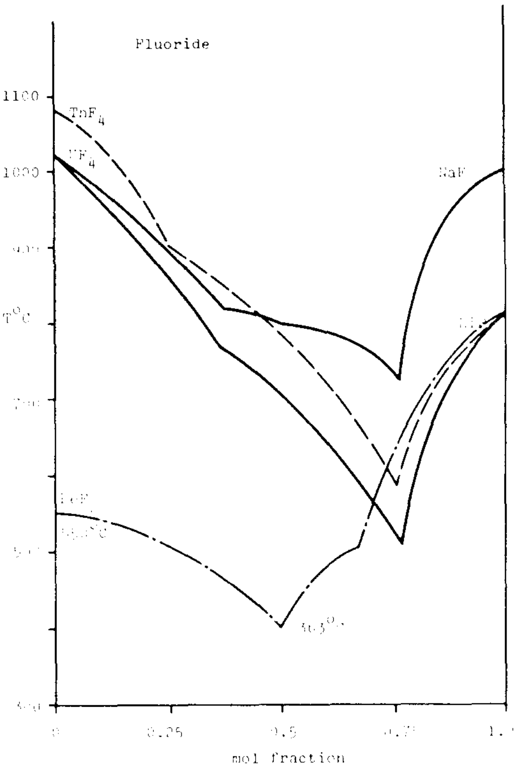


Fig. 6.6 Melting Point of - - Selected Salts Fig. 6.5



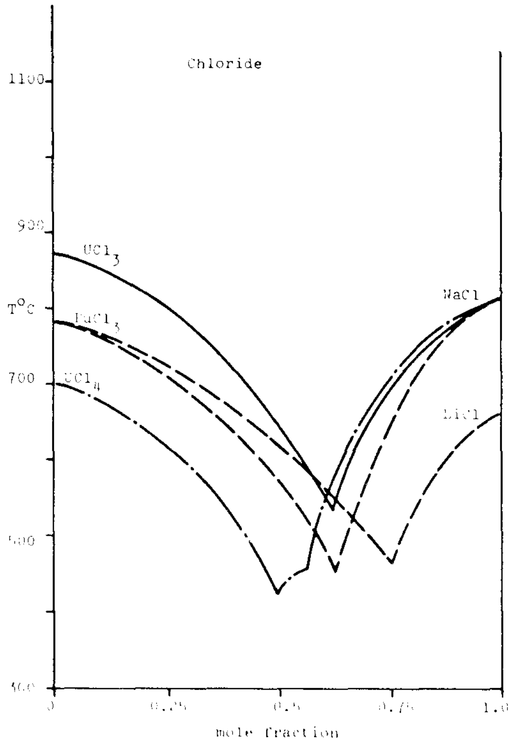


Fig. 6.7 PuCl$_3$-Alkali Metal Chlorides

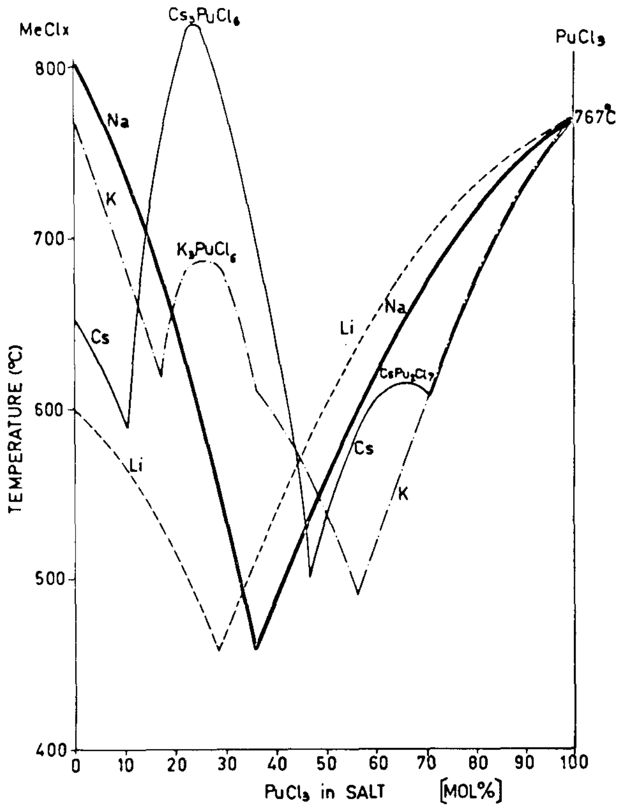


Fig. 6.8 Phase Diagram for PuCl$_3$/NaCl and UCl$_3$/NaCl

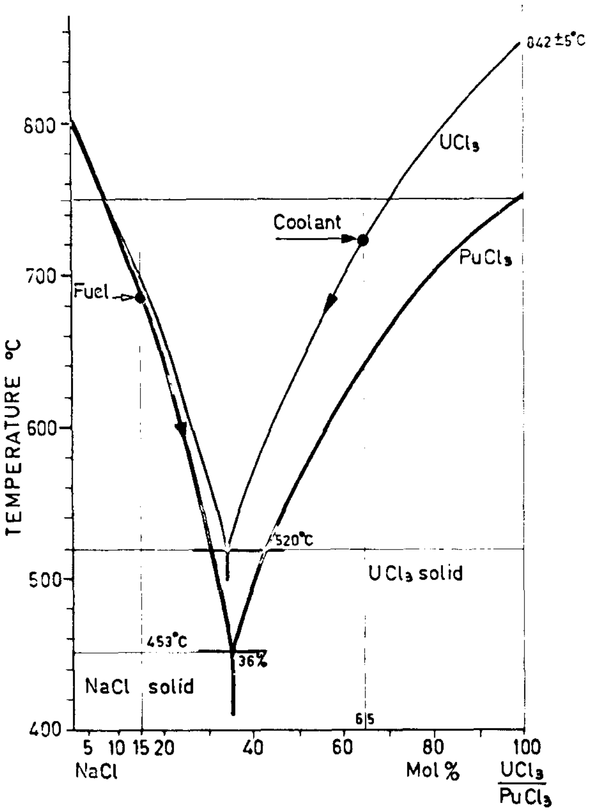


Fig. 6.9 The System $PuCl_3$-$NaCl$-UCl_3

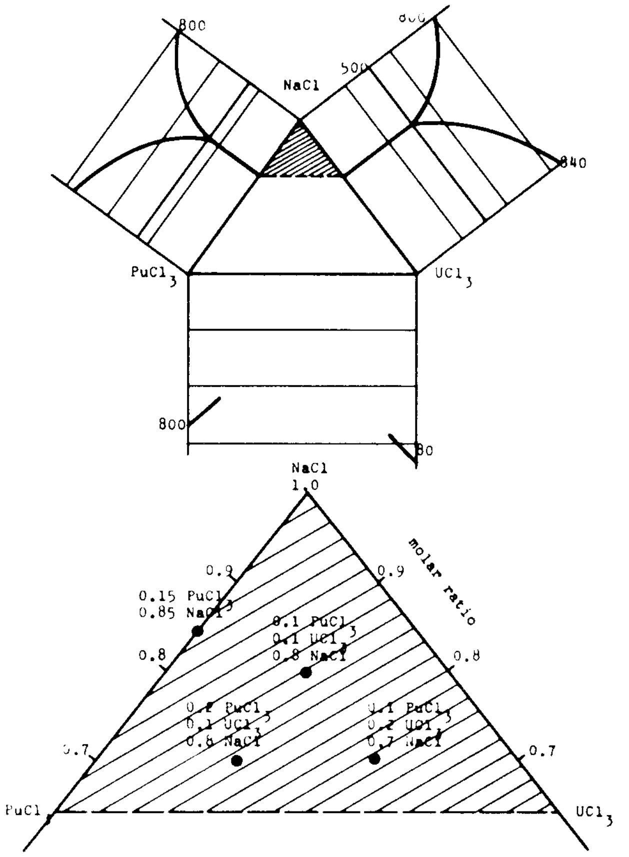


Fig. 6.10 **Phase diagramme** with thorium

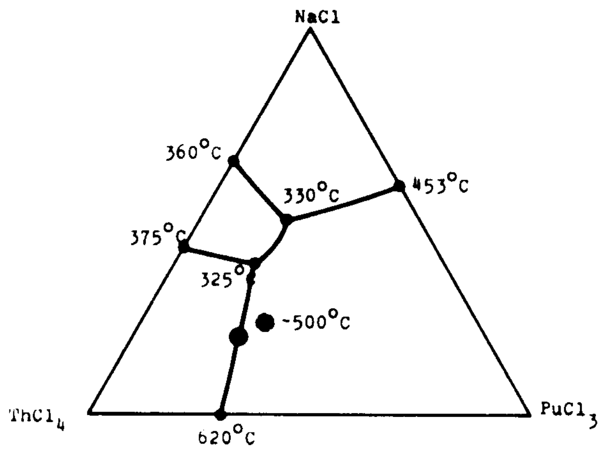


Fig. 6.11 Vapour pressure - metal chlorides

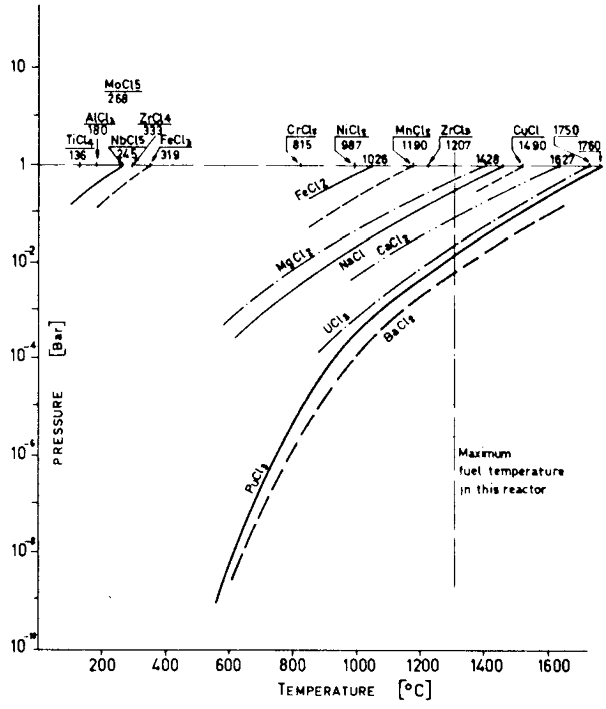


Fig. 6.12 CHLORIDES-OXIDES EQUILIBRIUM DIAGRAM AT 1000K

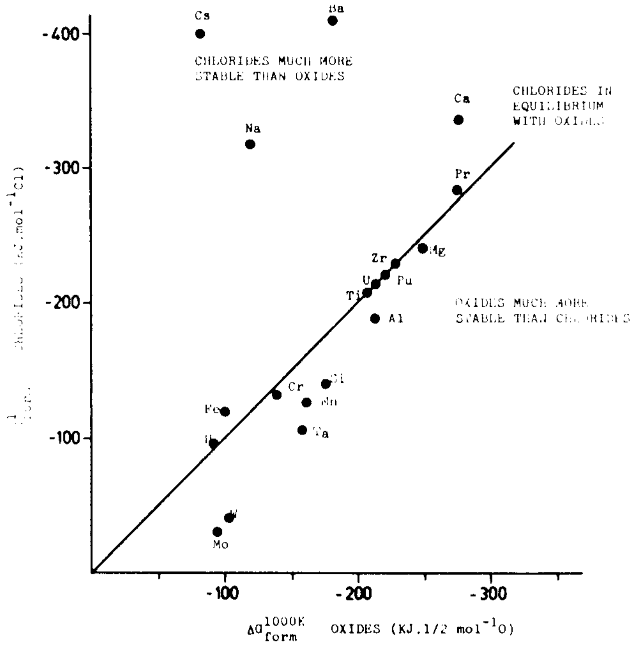


Fig. 6.13 Salts properties at 650°C v. Chemical composition Data derived from Nelson for $UCl_3/PuCl_3/MgCl_2/NaCl$

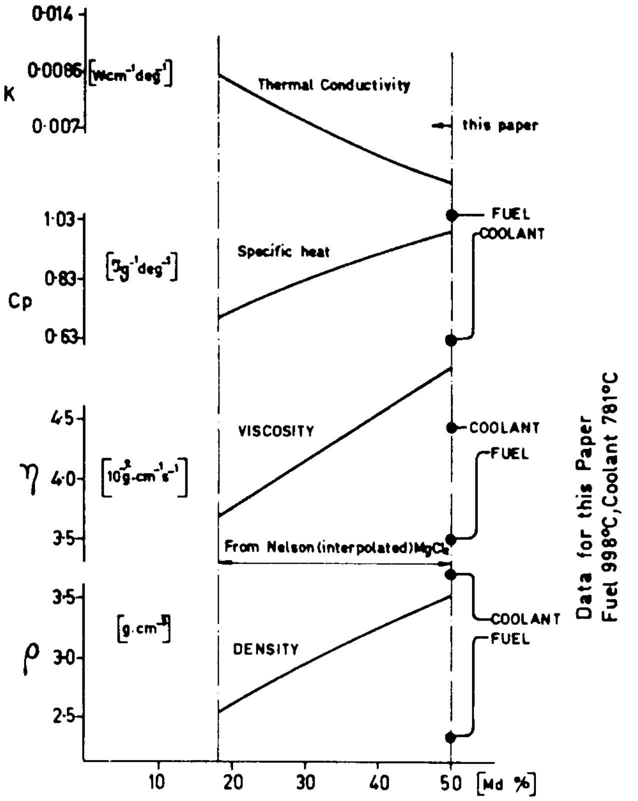


Fig. 6.14 Free enthalpy of formation chlorides

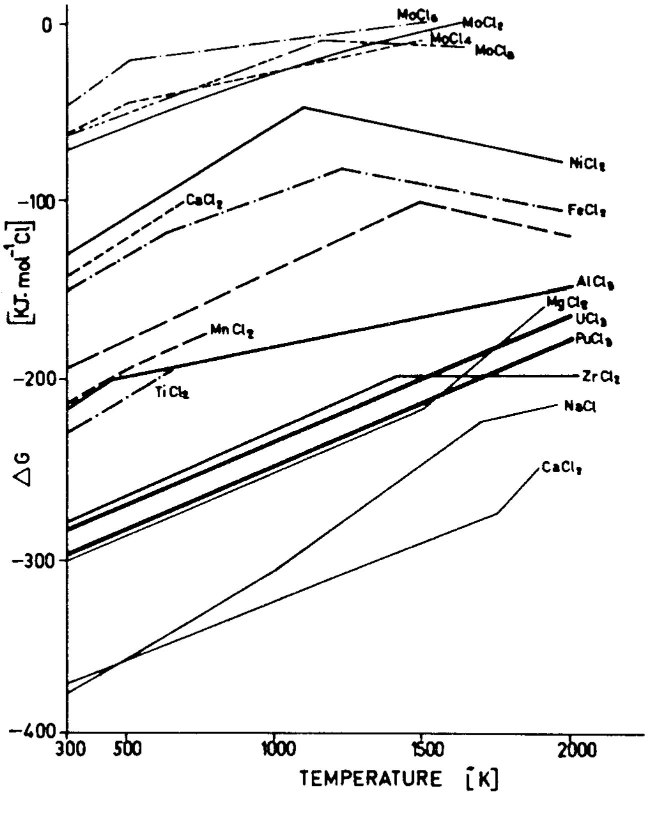


Table 6.1 Properties of fuel components

	$PuCl_3$	UCl_3	NaCl
Molecular weight	348.3	347	58.4
Postulated molar ratio-fuel	0.15	-	0.20
- blanket material	-	0.65	0.20
Density solid state ($kg \cdot m^{-3}$)	5.7	5.57	2.14
Melting point ($^{\circ}C$)	767	835	800
Boiling point at atmospheric pressure ($^{\circ}C$)	1730	1720	1465
Melting enthalpy ($kJ \cdot mol^{-1}$)	64.0	64	28
Enthalpy of vapourisation ($kJ \cdot mol^{-1}$)	240	300	188
Temp coeff. of density (K^{-1})	0.0010	0.0010	0.0005
Specific heat ($J \cdot mol^{-1}\ K^{-1}$)	140	140	77
Thermal conductivity ($W \cdot cm^{-1}\ deg^{-1}$)			
Viscosity ($g \cdot cm^{-1}\ s^{-1}$)	Fuel: 0.025	Coolant: 0.045	0.0143
Temp coeff. of viscosity (K^{-1})	0.0005	0.0005	
Free enthalpy of formation at 1000 K ($kJ \cdot mol^{-1}$)	-750	-675	-320

Table 6.2 Other chlorides of plutonium and uranium

	Plutonium	Uranium		
	$PuCl_4$	UCl_4	UCl_5	UCl_6
Melting point (^oC)	All efforts to produce pure solid $PuCl_4$ have been unsuccessful; only in gaseous state with free chlorine, or in molten salt solution or in aqueous solution as complexes	590	(287)	178
Boiling point (^oC)		792	(417)	(372)
Free enthalpy of formation at 1000 (kJ/mol)	4 x -180 = -760	4x-182 = -768	5x-165 = -825	6x-130 = -780

6.2 Corrosion of structural material

6.2.1 General criteria

It is clear that one of the most problematic areas in molten salt reactor technology is the area of corrosion. Some criteria can be formulated as follows

- the free energy of chlorides formation for structural materials must be relatively low, significantly lower than those of plutonium and uranium chlorides but still lower than those of the main fission products

- the partial pressure of the chlorides formed from the structural materials must be rather low which corresponds to a relatively high boiling point for these chlorides (Fig. 6.16)
- the neutron capture cross section for (n,γ), (n,p) and $n,2n)$ must be low (see later)

The structural materials are in principle different for the two types of core discussed.

- internally cooled, using tubes plus the effect of the cooling agent
- externally cooled by pumping the liquid fuel out of the core

These two variants call for different structural materials and different requirements

Cooling method	heat conductivity of the structural material	mechanical behaviour
internally by tubes	very good	very good (thin wall tubes)
externally	not important	not so important

Fig. 6.16 Chlorides - Boiling point V Free enthalpy of Formation

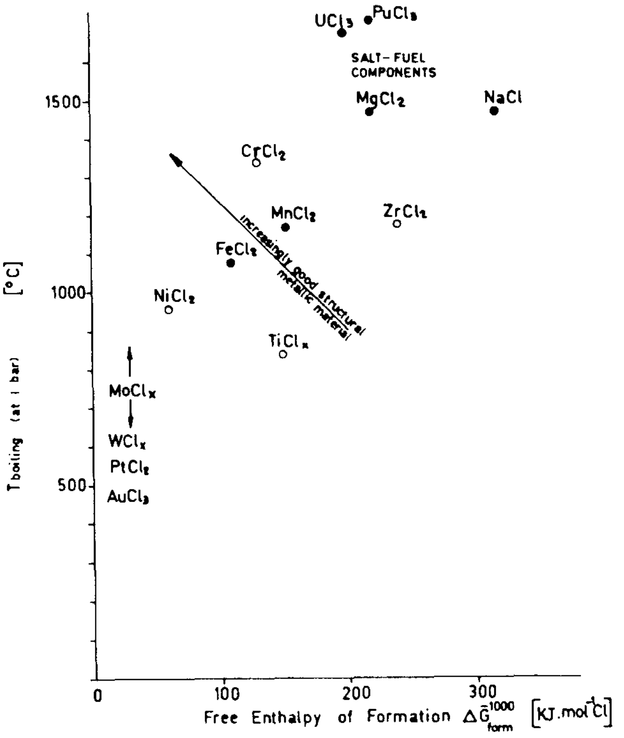


Fig. 6.17 Chlorides of Molybdenum

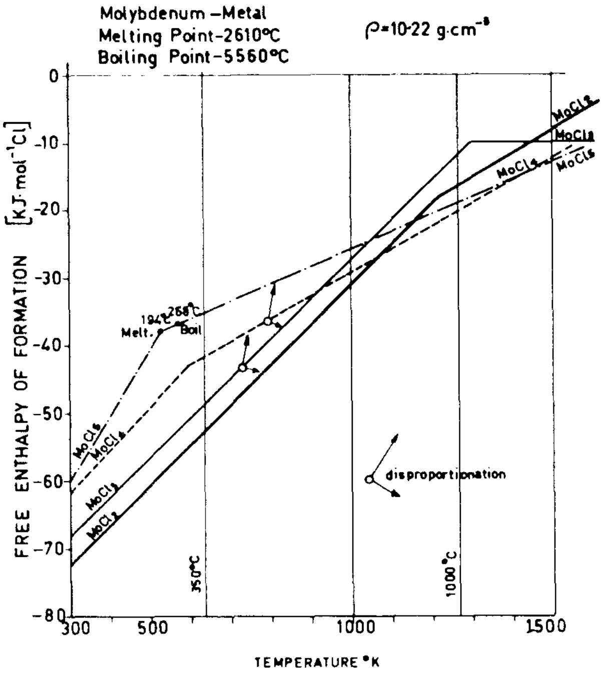


Fig. 6.18

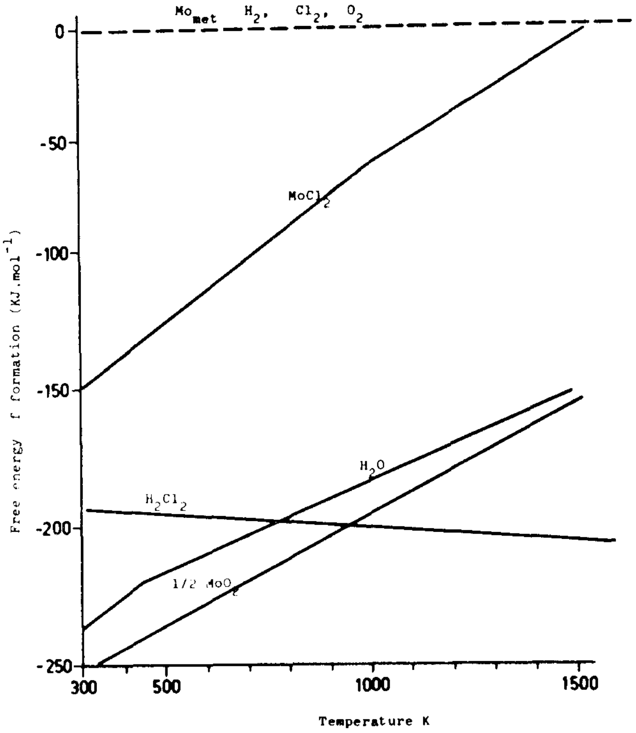
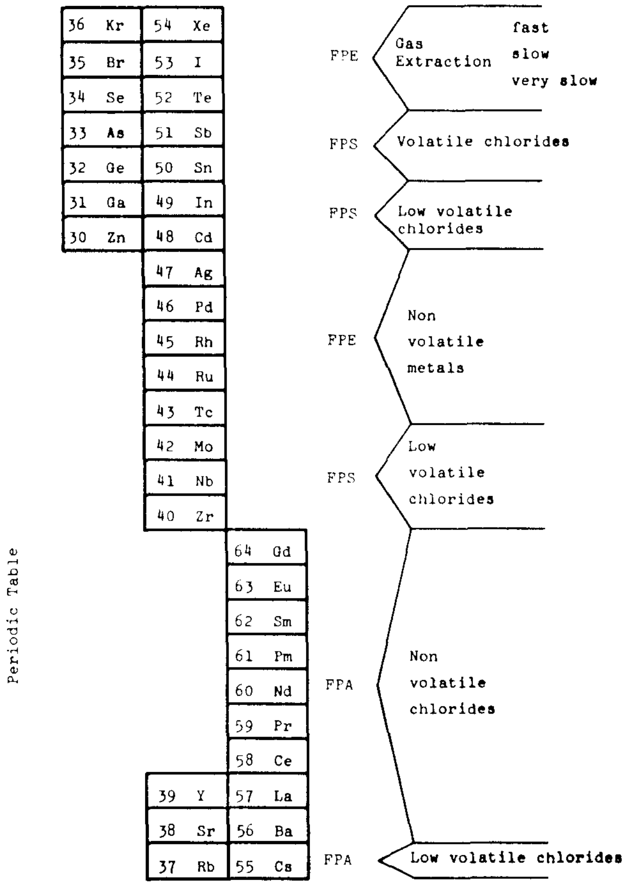


Fig. 6.20 Fission Products in Molten Chlorides Media



From these and other criteria the following choices may be made

- for the internally cooled core tubes: molybdenum
- for the wall of the spherical core, in the case of the externally cooled reactor: graphite and berrylia.

6.2.2 Molybdenum as structural material

The main corrosion processes result from the following mechanisms (m = metallic phase, s = salt phase, Me = metallic component of irradiated fuel or coolant)

$$Mo_{(m)} + \frac{2}{x} Me\,Cl_{x(s)} \longrightarrow Mo\,Cl_{2(s)} + \frac{2}{x} Me_{(m)}$$

For the behaviour of fresh fuel $PuCl_3$ in NaCl the most likely reaction is (T = 1250 K)

$$Mo_{(m)} + {}^{2}/_{3}\, PuCl_{3\,(s)} \longrightarrow Mo\,Cl_{2\,(s)} + {}^{2}/_{3}\, Pu_{(m)}$$

$$\Delta G^{1000K} = +\,450 \text{ kJ/mol Cl.}$$

The equilibrium constant of this reaction is small and equals 10^{-17} so that this reaction is completely unimportant.

In the blanket zone the most dangerous reaction is connected with uranium trichlorides (chlorine from the fission of $PuCl_3$).

$$UCl_3 + 1/2\; Cl_{2(s)} \longrightarrow UCl_{4(s)}$$

$$2UCl_{4(s)} + Mo_{(m)} \longrightarrow 2UCl_{3(s)}$$

the control of the UCl_3/UCl_4 ratio in the fertile coolant might be feasible due to the continuous reprocessing of this material together with the control of zirconium from the fission products oxidation state.

An additional problem comes from the fact that molybdenum has different oxidation states +2, +3, +4, +5 and all of them have the corresponding chlorides. (see Fig. 6.17)

Futher difficulties arise from the problem of the reactions between metal chlorides and oxygen and water.

These reactions (for oxidation state +2) could be written in simplified form

$$Me\,Cl_2 + H_2O \longrightarrow MeO + 2HCl$$

$$Me\,Cl_2 + \frac{1}{2}O_2 \longrightarrow MeO + Cl_2$$

The metal oxides are mostly insoluble in molten chlorides which results in a serious disturbance of the fuel system. From this point of view the metallic elements could be divided into three classes (see Fig. 6.12).

- those which are stable with H_2O and O_2, that is the chlorides are more stable than the oxides (e.g. Na, Cs, Ba) and partially Ca.
- those which are not stable with H_2O and O_2 and the resulting product is a mixture of chloride, oxychloride and oxide (e.g. Pu, U but also Zr, Ti, Al, Fe, Cr, Mn, Mg - this is the most numerous group of metals).
- those in which chlorides are converted to the most stable oxide in the presence of H_2O or O_2 (e.g. Mo, W) metals of this class seem to be less numerous than those in the other two classes.

This property causes the rapid elimination of traces of water or oxygen in the molten chloride salts of Pu and U. It is also well known that traces of H_2O and O_2 have a very big influence of the corrosion rate. Molybdenum belongs to the last class, the oxide is much more stable than the chloride (Fig. 6.18)

This means that the traces of exygen or even water will result in the production of molybdenum oxide. This effect requires considerable further study.

6.2.3 The irradiation of molybdenum and iron in a fast high flux reactor

The high neutron flux irradiation causes physical and chemical changes in structural materials.

Molybdenum is a mixture of stable isotopes. The most important by-product of neutron irradiation is the Tc-99 beta-emitter with $t_{1/2} = 2.1 \times 10^5$ years and belongs to the decay chain shown here.

$$\begin{tikzcd}
\frac{\text{Mo -98}}{(23\%)} \; (n,\gamma) \; \text{Mo -99} \arrow[r, "\beta^-", "t_{1/2} \approx 66h"'] & \text{Tc -99} \arrow[r, "\beta^-", "t_{1/2} = 2.1 \times 10^5 y"'] \arrow[d, "(n,\gamma)"] & {} \\
 & \text{Tc -100} \arrow[r, "\beta^-", "t_{1/2} = 17s"'] & \text{Ru -100}
\end{tikzcd}$$

For approx 1000 kg molybdenum in the core in the form of cooling tubes or about 10,000 moles. the Mo-98 gives 2300 mol. The irradiation rate N (mols/s) equals

$$N^{\text{Mo-99}}_{s^{-1}} = (2.3 \times 10^3) \times (6 \times 10^{23}) \times (10 \times 10^{-27}) \times 10^{16} = 1.2 \times 10^{17} \text{atom/s}$$

After an irradiation of 700 hrs a steady state concentration of Mo-99 is reached

$$C^{\text{Mo-99}}_{\substack{\text{steady}\\\text{state}}} = \frac{1.2 \times 10^{17}}{3 \times 10^{-5}} = 3 \times 10^{21} \text{atoms} = 0.005 \text{ mol}$$

The radioactivity of the Tc-99

$$(t_{1/2} = 2.1 \times 10^5 y = 6.2 \times 10^{12} s : \lambda = 10^{-13} s^{-1})$$

after three years irradiation of 1000 kg of molybdenum in the fast reactor core:

$$\text{Activity}\ \begin{matrix}\text{Tc-99}\\ \text{(3 year)}\end{matrix} = 1.2\times10^{17}\text{atoms/s} \times (3\times3.1\times10^{7}\text{s/year})$$

$$\times \frac{10^{-13}}{3.7\times10^{10}} = 3\ \text{Curie/tonne of Mo}$$

The diffusion rate of hydrogen from the molten fuel to the coolant and blanket (here also UCl_3 - NaCl) must also be mentioned.

One can assume that this melt containing hydrogen is saturated so that the porosity of the wall (molybdenum) will play a minor role. The most important factor is the variation in the mechanical properties of the molybdenum caused by the uptake of hydrogen.

The problem of molybdenum corrosion in chlorine containing media is particularly complicated by the numerous molybdenum chlorides: $MoCl_2$, $MoCl_3$, $MoCl_4$, $MoCl_5$ (Fig. 6.17)

6.2 Fission product behaviour in the fuel

The fission of $PuCl_3$ causes the formation of two fission products E' and E" and three atoms of chlorine

$$PuCl_3 \xrightarrow{(n,f)} E' + E'' + 3Cl$$

For the fissioning of 100 atoms of Pu the following balance has been suggested

$$\begin{aligned}100\ PuCl_3 \xrightarrow{(n,f)}\ & 0.008\ Se_{gas} + 0.003\ Br_{gas} + 0.942\ Kr_{gas}\\
& + 1.05\ RbCl + 5.49\ SrCl_2 + 3.03\ YCl_3 + 21.5\ ZrCl_3\\
& + 0.29\ NbCl_5(?) + 18.16\ MoCl_2 + 0.28\ MoCl_3 + 4.01\ Tc_{met}\\
& + 31.45\ Ru_{met} + 1.73\ Rh_{met} + 12.66\ Pd_{met} + 1.88\ AgCl\\
& \qquad + 0.66\ CdCl_2 + 0.06\ InCl + 0.324\ SnCl_2 + 0.67\ SbCl_3\\
& + 7.65\ TeCl_2 + 6.18\ I_{gas} + 21.23\ Xe_{gas} + 13.34\ CsCl\\
& + 9.50\ BaCl_2 + 5.78\ LaCl_3 + 13.98\ CeCl_3 + 4.28\ PrCl_3\\
& + 11.87\ NdCl_3 + 1.44\ PmCl_3 + 3.74\ SmCl_3 + 0.60\ EuCl_2\\
& + 0.03\ CdCl_3\end{aligned}$$

the average balance of fission can be represented in the following manner

$$100\ PuCl_3 \xrightarrow{(n,f)} 200\ ECl_{1.5}$$

Table 6.3 The behaviour of fission products in the molten chlorides fuel. (Yields given represent products for 100 Pu atoms fissioned).

Free enthalpy of formation, $- G^{1250\ K}$ ($kJ \cdot mol^{-1}$ Cl)	Principal fuel and structural material	Fission products	Remarks
0	-Mo metal (struct.)	Pd(12.66), Tc(4.01), Rh(1.73), Ru(3.144), I(6.17), Te(7.65) Xe(21.2), Kr(0.94), Br	no chlorine for the synthesis of possible metal chlorides
100	-$MoCl_2$ (corrosion product)	-$MoCl_x$ (18.16) -$MoCl_3$ (0.28)	
		-AgCl (1.88) -$SbCl_3$ (0.67) -$CdCl_2$ (0.66) -$SnCl_2$ (0.32) -InCl (0.06)	
200	-UCl_4 (?)		
	-UCl_3	-$ZrCl_3$ (2.15) -$ZrCl_2$	
	-$PuCl_3$	-YCl_3 (3.02) -$PrCl_3$ (4.28) -$EuCl_3$ (1.06) -$CeCl_3$ (13.98) -$LaCl_3$ (5.78)	
300	-NaCl	-RbCl (1.05) -CsCl (13.75) -$SmCl_2$ (3.73) -$SrCl_2$ (5.48) -$BaCl_2$ (9.50)	

F.P. in elemental form

300 atoms of chlorine released from fission of 100 atoms of Pu

From the earlier published data (Chasanov, 1965; Harder et al., 1969; Taube, 1961) it appears that the problem of the chemical state (oxidation state) in this chloride medium for the fission product element constituent requires further clarification.

From a simple consideration it seems that the freeing of chlorine from the fissioned plutonium is controlled by the fission product elements with standard free enthalpy of formation up to ∿ 20 KJ/mol of chlorine, that is up to molybdenum chloride. The more 'noble' metals such as palladium, technetium, ruthenium, rhodium and probably tellurium and of course noble gases: xenon, krypton plus probably iodine and bromine, remain in their elementary state because of lack of chlorine. Molybdenum as a fission product with a yield of 18% from 200% all fission products may remain in part in metallic form. Since molybdenum also plays the role of structural material the corrosion problems of the metallic molybdenum or its alloys are strongly linked with the fission product behaviour in this medium.

The possible reaction of UCl_3 and $PuCl_3$ with $MoCl_2$ resulting in further chlorination of the actinides-trichlorides to tetrachlorides seems, for $PuCl_3$ very unlikely (ΔG^{1000K} = - 450 kJ/mol Cl) but this is not so for UCl_3.

A rather serious problem arises out of the possible reaction of oxygen and oxygen containing compounds (e.g. water) with $PuCl_3$ and UCl_3 which results in a precipitation of oxides or oxychlorides. The continuous reprocessing may permit some control over the permissible level of oxygen in the entire system as well as the continuous gas bubbling system with appropriate chemical reducing agent.

Corrosion of the structural material, being molybdenum is also strongly influenced by the oxygen containing substances, A protective layer of molybdenum however, may be used on some steel materials using electrodeposition or plasma spraying techniques.

Note that all these considerations have been based on standard free enthalpy: but even a change in the thermodynamic activity from = 1 to = 0.001 which means a change in free enthalpy of $14 kJ\ mol^{-1}$ thus appears insignificant as far as these rough calculations go.

In the fertile material reactions also occur and the most important are

fission process: $UCl_3 \longrightarrow$ Fiss. products $+ 3Cl$
oxidation process: $UCl_3 + 1/2Cl_2 \longrightarrow UCl_4$; $\Delta G^{1250K} = 25 kJ\cdot mol^{-1}$
disproportionation: $UCl_3 + 3UCl_3 \rightarrow 3UCl_4 + U_{met}$

6.4 Some comments on reprocessing

Breeder reactors as is known form part of a breeder system which includes not only the power reactor but also the reprocessing plant.

The advantages of molten salt breeder reactors become particularly apparent when the reprocessing plant is under the same roof as the power reactor and when chemical separation processes take place in the high temperature molten salt media in a continuous cycle. From Fig. 6.2 it can be seen that all fission products might be classified into 3 classes.

FPA = fission products of alkali and alkali earth but also rare earth elements which have free enthalpy of chloride formation greater than those of $PuCl_3$.

FPS = fission products of seminoble metals with free enthalpy of formation smaller than those of $PuCl_3$.

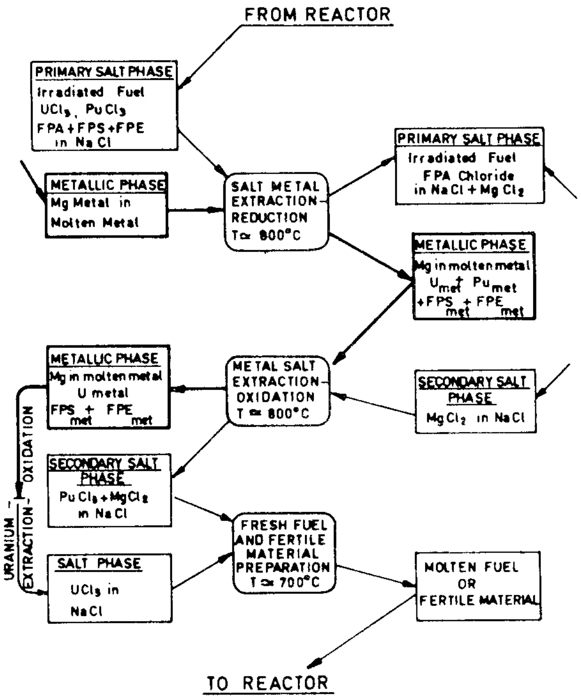
FPE = fission products existing in elementary form because of the low free energy of chloride formation or negative balance of chlorine.

The separation of plutonium or Uranium form the irradiated fuel by means of pyrochemical techniques could be carried out for example in the following way.

Molten salt, primary phase Pu, FP (part of FP remains)
Metallic phase (part of FP remains)
Molten salt, secondary phase containing only Pu.

This is the so called metal transport process. The proposed schematic of separation processes utilizing metal transport is given in Fig. 6.21 and 6.22.

Fig. 6.21 Fuel reprocessing flow Scheme

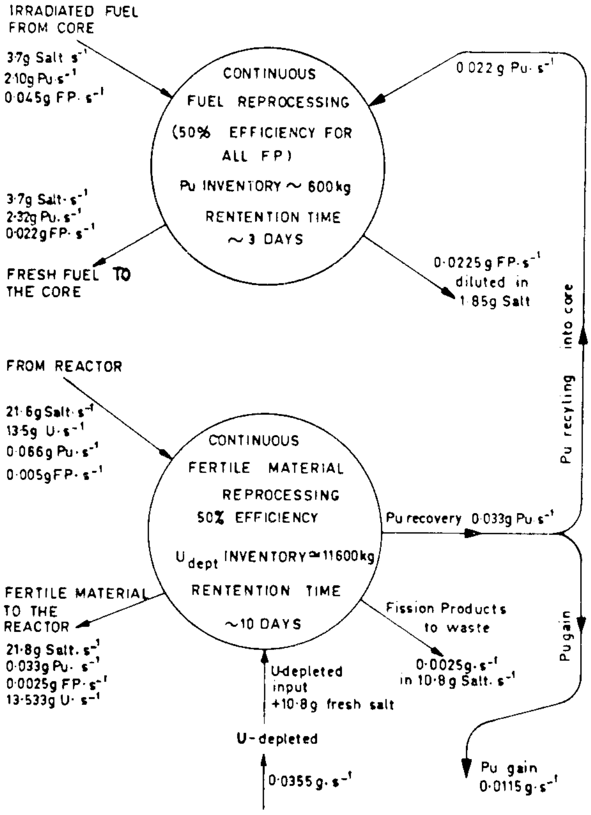


FPA — Alkali and Alkali earth fission products e.g. Cs, Ba, Sr.

FPS — Semi and noble metals and metal chlorides.

FPE — Noble metals in metallic states and noble gases.

Fig. 6.22 Fuel Reprocessing Material balance



6.5 In-core continuous gas purging

6.5.1 The proposal

In this type of reactor an in-core continuous gas purging of the molten fuel which can significantly improve the safety in an in-core accident, is possible.

A mixture of hydrogen-helium gas is continuously bubbled through the liquid fuel in the core. The mean dwell time of the gas-bubbles needs to be controlled and the mean transport time of the molten components to these bubbles must also be controlled (e.g. if speed-up is desired-intensive mixing, if delay-local addition of a further gas stream).

The aim of the gas stripping is as follows:

1) to remove the volatile fission products which in the case of an accident control the environmental hazard. (I-131, Xe-133, Kr-85 and precursors of Cs-137) and at the same time for the thermal reactor, removal of the I-135, precursor of Xe-135, improves the neutron balance.

2) to control the production of delayed neutrons since most of the precursors and nuclides of this group are very volatile, e.g. + Br-I-isotopes.

3) removal of oxygen and sulphur, continuously (see Chapter 7)

4) *in situ* control of corrosion problems on structural materials

For the sake of a first approximation a gas flux of 30 cm^3 per sec. (normal state) of H_2/He is arbitrarily assumed. At 20 bar pressure and with a dwelling time in core of 20 seconds, the gas bubbles will only occupy a fraction of the core equal to 10^{-5} of its volume and have little influence of the criticality, (but the collapsing of bubbles results in a positive criticality coefficient).

The system proposed for continuous removal of the volatile fission product from the core itself has a retention time of some hundreds of seconds only. Each reprocessing mechanism which operates out of core is limited by the amount of molten fuel being pumped from the core to the reprocessing plant. This amount, due to the high capital cost of the fuel and high operation costs cannot be greater than that which gives a fuel in-core dwell time of about one week. Even with a 1 day dwell time, that is, if after one day the fuel goes through the reprocessing plant, no acceptable solution to the I-131 problem is obtained since the activity of this nuclide is only diminished by one order of magnitude. Only a direct in-core removal gives the dwell time in core as low as some hundreds of seconds.

6.5.2 Delayed neutron emitters

The principal question arise out of the fact that some of the short lived iodine and bromine (perhaps also arsenic, tellurium) isotopes are the precursors of the delayed neutrons.

Table 6.4 Precursors of delayed neutrons for Pu-239 fast fission

Group	Half life	Fraction	Probable Nuclide
	t 1/2 (seconds)	%	
1	52.75	3.8	Br-87
2	22.79	28.0	I-137, Br-86
3	5.19	21.6	I-138, Br-89
4	2.09	32.8	I-139, Br-90
5	0.549	10.3	
6	0.216	3.5	

As other possible nuclides the following can be considered + As-85; Kr-92, -93; Rb-92, -94; Sr-97, -98; Te-136, -137; Cs-142, -143. The removal of these delayed-neutron precursors from the core reduces the value of β, which is lower for Pu-239 than U-235. Thus we have a problem of reaching a compromise between an as rapid as possible removal of the hazardous I-131, and as long a dwell time in the core for the delayed neutron precursors: I-140, I-139, I-138, I-137 and the appropriate bromine isotopes.

In this case the mean dwell time of iodine in the steady state reactor is about 100 seconds. It can be seen that the activity of iodine for a 2.5 GW(t) reactor is of the order of only 10 kilo curies (for seconds) activity of approx. 10^4 (or 10^3 for 1000 second extraction rate).

The gas-extraction also influences the other volatile nuclides. From a very rough estimation for these molten salts (with a small excess of free hydrogen) the following fission products and their associated precursors of iodine and bromine can be volatile at 1000°C.

In elementary form:	Xe, Kr, Te (?)
In simple volatile hydrides:	BrH, IH
In simple volatile chlorides:	$SnCl_2$, $SbCl_3$, $NbCl_3$, $CdCl_2$.

This amount of finally volatile components including I, Br, Xe, Kr amounts to approximately half the total fission products (i.e. 100 micromoles per second). In addition there is the corresponding amount of tritium (from ternary fission). This amount of all fission products corresponds to a gas volume ratio of about 2 cm^3/s or 10 times smaller than the postulated amount of hydrogen flow at 30 cm^3/s.

The extraction removes all short lived fission products which are volatile under these conditions. Thus not only is the removal of the iodine isotopes and the consequent reduction in production of xenon (e.g. for the atom number: A = 135, 136, 137, 138, 139) achieved but it slows down the in-core production of Cs-137, Cs-138, Cs-139, and then also barium-139

The higher components of the liquid fuel: $PuCl_3$ and NaCl. The Fuel consists of:

- 15 mol% $PuCl_3$; boiling point 2040 K
- 85 mol% NaCl ; boiling point 1728 K

One can as a first approximation say that it would have the following composition in the vapour phase: 5 mol% $PuCl_3$ - 95 mol% NaCl.

The order of magnitude of vapour for pure components at a temperature of about 1250 K is

$$NaCl \sim 5 \times 10^{-2} \text{ bar}; \quad PuCl_3 \sim 10^{-4} \text{ bar}$$

For the $PuCl_3$-NaCl system one assumes here a lowering of the vapour pressure (thermodynamic activity coefficient approx. 0.1). At the postulated volumetric flow rate of 30 cm^3 H_2 normal per second, the vapourized amount of plutonium is given by:

$$\frac{30 \text{ cm}^3}{22000 \text{ cm}^3/\text{mol}} \times 10^{-4} \text{ bar} \times 10^{-1} = 10^{-8} \text{ mol Pu/s}$$

This amount of plutonium is of the order of 10^{-4} relative to the amount of plutonium fissioned in the same time (approx 10^{-4} mol Pu/s). However, it still has to be recovered, which unfortunately makes the reprocessing more complicated.

Last but not least is the in-core gas extrection of two other elements

- oxygen in the form of H_2O : oxygen from impurities (i.e. PuOCl)
- sulphur in the form of H_2S: sulphur from the nuclear reaction: ^{35}Cl (n,p) ^{35}S

(see Chapter 7).

7. EXPERIMENTAL WORK

7.1 Chemical behaviour of radiosulphur obtained by $^{35}Cl(n,p)^{35}S$ during in-pile irradiation

(according to Janovici, 1975)

The rather large concentration of sulphur formed by $^{35}Cl(n,p)^{35}S$ reaction in the molten chlorides system proposed for the fast reactor makes it necessary to obtain the full information on the chemical behaviour of the radiosulphur. The most recent studies on the chemical states of radiosulphur obtained by n-irradiation of alkali chlorides have shown the complexity of this problem.

To obtain new data on the behaviour of radiosulphur we have investigated the influence of the time and temperature of irradiation and of post-irradiation heating on the chemical distribution of the sulphur.

EXPERIMENTAL

Sodium chloride ("Merck" reagent) was heated for 60hr at 200°C in an oven in vacuo. The dried samples of 100 mg sealed in evacuated (10^{-4}torr) quartz tubes were irradiated near the core of the "Saphir" swimming pool reactor for different periods at a neutron flux of 5×10^{12} and 4.3×10^{12} n cm^{-2} s^{-1}. Reactor irradiations were carried out at an estimated temperature of 150°C and -190°C. After irradiation the samples were kept for 8 days to allow the decay of ^{24}Na.

The method of ^{35}S-species separation. The crushing of the irradiated ampoule was made in a special device from which the air was removed by purging with a nitrogen stream containing 10ppM of oxygen. After crushing a gentle stream of nitrogen was allowed to flow for about 10 min. The gases evolved were collected in cooled traps. The irradiated slat was dissolved in 2 M KCN solutioncontaining carriers of S^{2-}, CNS^-, SO_3^{2-}, SO_4^{2-}. During dissolution oxygen was not completely excluded although nitrogen gas was passed continuously through the system. The radiosulphur found in gaseous form was determined as barium sulphate. For the ^{35}S-species separation the chemical method described recently by Kasrai and Maddock was used. The radioactive samples were counted under a thin window Geiger counter. All measurements were made in duplicate with and without Al-absorber.

Post-irradiation heating. The sealed irradiated ampules were heated in an electric oven at 770°C for 2 hr or at 830°C for about 5 min. and then cooled and crushed in a closed system under a stream of nitrogen.

Results and discussions

Effect of length of irradiation time. As can be seen from Fig. 7.1 S^{2-} remains the preponderent fraction independent of the irradiation time. Formation of S^{2-} is indicated by charge conservation during the $^{35}Cl(n,p)^{35}S$ reaction. Alternatively it can be supposed that reduction of sulphur takes place by capture of electrons due to the discharge of F-centers. The presence of S^0 in this oxidation state in the lattice is no longer contested. The precursors of higher forms may be S^+ as a result of an electron loss from S^0. However, the interaction of chlorine entities formed by irradiation with radiosulphur to form species as SCl, SCl_2^-, SCl_2 may be an important mechanism in forming the percursors of sulphate and sulphite.

During longer irradiations some of the sulphide is converted into higher oxidised forms. This may be a consequence of radiation-produced defects with oxidising character (e.g. V-centres or derivatives). It is possible that the concentration of defects responsible for reduction of the sulphur decreases by annihilation when new traps are formed. The oxidation of radiosulphur with increase of radiation damage concentration may also be due to the reaction of recoil sulphur with chlorine atoms. The presence of OH^- in the crystal must not be neglected. It has been suggested that radiolysis of OH^- can be responsible for accelerating the oxidising process.

Effect of post-irradiation heating. The effect of post-irradiation heating (including melting) can be seen in Table 7.1.

Comparisons between heated and unheated samples are made for irradiations of 2, 12 and 24 hr. For 2 hr irradiation, results on samples heated at a temperature below the melting point of NaCl are also presented. As is seen, on heating, a part of the radiosulphur is found in a volatile form. The volatile radiosulphur appears at the expense of S^0 and higher oxidation forms. The results show that with temperatures above the boiling point of sulphur and above melting point of NaCl the S^0 and S^+ and/or S_xCl_y receive sufficient kinetic energy to migrate to the surface or even to escape from the crystal and be collected as volatile radiosulphur. However, there are some differences in the ^{35}S-chemical distribution on heating below and above the melting point of NaCl (experiments 2-3). It seems that for relatively short periods of irradiation (2 hr) only the sulphate and sulphite precursors account for the volatile radiosulphur fraction. For a longer time of irradiation, on melting the S^0 value decreases to about 2% and this corresponds to an increase in the volatile radiosulphur (experiments 5, 7). However, a small and practically constant yield of S^0 is found in the melt after longer irradiation

Table 7.1

Expt.	Irrad. time	Post-irrad. treatment	S^{2-} %	S^0 %	SO_4^{2-} + SO_3^{2-}* %	S-volatile %
1	2 hrs.	no	73.1 ± 0.4	9.8 ± 0.8	16.9 ± 0.8	0.01
2	"	770°C 2 hrs.	75.4 ± 2.8	5.3 ± 0.3	3.6 ± 2.3	15.4 ± 1.1
3	"	830°C 5 min.	77.2 ± 2.0	11.0 ± 0.8	6.6 ± 2.5	5.0 ± 1.4
4	12 hrs.	no	67.5 ± 0.7	12.1 ± 0.1	20.4 ± 0.6	0.01
5	"	830°C 5 min.	7.15	1.9	18.9	7.6
6	24 hrs.	no	64.4 ± 2.5	11.9 ± 0.5	23.7 ± 2.0	0.01
7	"	830°C 5 min.	68.2 ± 3.4	2.3 ± 0.5	21.4 ± 2.3	7.9 ± 0.7

Expt. 1-5 = 4.3 10^{12} n cm^{-2} s^{-1}

Expt. 6-7 = 5.0 10^{12} n cm^{-2} s^{-1}

* Sulphite fraction is less than 5% in our experiments and always lower than sulphate fraction

times. No significant changes are observed for the sulphide and higher oxidation forms. Comparison of these results and those presented in Fig. 1 shows that in the post-irradiation melted samples the radiation damage does not have the same effect as in the unmelted samples. Supplementary information can be obtained by studying the effect of high temperature irradiation on the distribution of the radiosulphur. It is possible that in the molten state the active oxidising agents have a different identity from those present below melting. The presence of oxygen and probably sodium oxides during melting may have a determinant role in deciding the state of the radiosulphur.

Effect of irradiation temperature.

A comparison of results obtained by irradiation at 423 K and 77 K (Table 7.2) shows that the higher oxidation fraction is lower (3%) at 77 K. As is seen the increased S^0 after low temperature irradiation occurs at the expense of the sulphate + sulphite and sulphide fractions. The defects with oxidising and reducing character formed by low temperature irradiation such as F and V-centres (or derivatives) become important factors in determining the radiosulphur behaviour.

7.2 Temperature dependence of sulphur species (according to Furrer, 1977)

Significant amounts of the order of magnitude of thousands of ppm ^{35}S would be present as steady-state concentration in a proposed fast breeder reactor fuelled with molten Pu/U-chlorides diluted in NaCl. To obtain information about the chemical behaviour, mainly the distribution of oxidation-states, the influence of irradiation temperatures (-190 and 15°C) and the ffects of a post-irradiation heat treatment, solid NaCl was investigated and the results published. The subject of the present note are investigations at higher irradiation temperatures, especially with samples molten during irradiation.

Fig. 7.1 Effect of length of irradiation time on the ^{33}S -species distribution

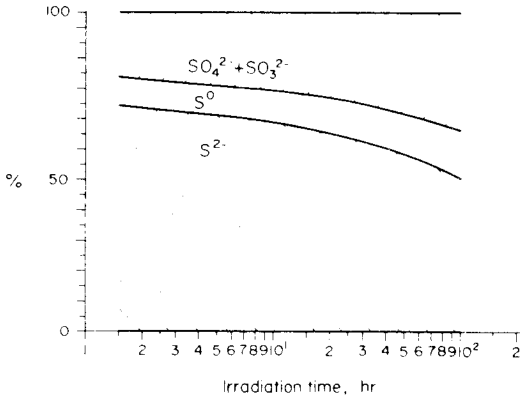


Fig. 7.2 Irradiation-temperature dependence of the oxidation-state distribution of ^{35}S-species

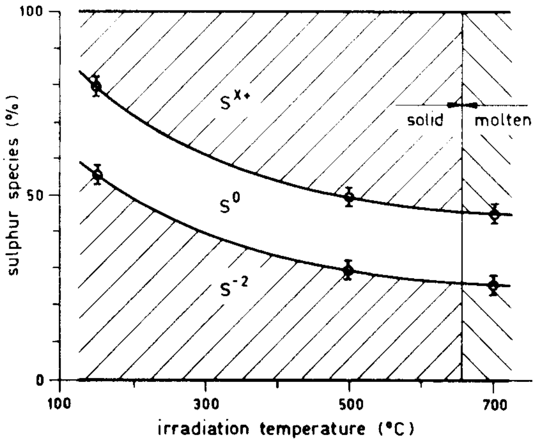


Table 7.2

Irrad. conditions flux, time	Irrad. temp. (K)	S^{2-} %	S^{0} %	$SO_4^{2-} + SO_3^{2-}$ %
$5.10^{-12} n\ cm^{-2} s^{-1}$ 2 hrs.	423	73.1 ± 0.4	9.8 ± 0.8	16.9 ± 0.8
"	77	65.4 ± 0.9	31.5± 1.1	3.0 ± 0.1

Experimental

Equimolar mixtures of NaCl and KCl ("Suprapur", Merck) were studied instead of pure NaCl (m.p.658 vs 800°C) because the irradiation device permitted temperatures of up to 750°C only. The finely crushed slat-mixture was dried in vacuo at 250°C for 24 hr and subsequently treated with dried HCl-gas at 300°C for 24 hr in order to remove traces of water and hydroxides. 100 mg samples were weighed into quartz ampoules in a glove-box with a purified nitrogen atmosphere (O_2, H_2O 10ppm).

In order to study the influence of oxygen from the surface of the SiO_2-ampoules, parts of the samples were weighed into small crucibles made of gold-foil and closed by folding the foil. The ampoules were evacuated to a pressure of less than 10^{-5}mm Hg (24 hr) and during evacuation heated to 250°C for about 8 hr to remove adhering traces of HCl, sealed and irradiated for 2 hr at 500, 600 or 700°C in the swimming-pool reactor SAPHIR near the core at a total flux of about 4×10^{12} n cm^{-2} s^{-1}. The neutron-spectrum is not well characterised but known to be rather hard.

The post-irradiation treatment followed closely the work of (6) as described in detail elsewhere (4.5). The ampoules were crushed at room remperature in a nitrogen atmosphere, volatile reaction products carried by a nitrogen gas-stream into cold-traps cooled with liquid nitrogen and the samples subsequently dissolved in an oxygen-free carrier-solution containing cyanide and sulphide, thiocyanate, sulphite and sulphate as carriers. The fractions were separated. independently oxidized to sulphate, precipitated as $BaSO_4$ and the activities measured with a thin-window GM-counter. Some experiments were made at 150°C irradiation-temperature in order to be able to compare the CaCl/KCl-system with pure NaCl. New irradiations of pure NaCl were carried out in order to study the influence of the HCl pre-irradiation treatment.

Results and discussions

The data for at least 2 independent experiments at 150 and 500°C or 4-5 experiments at 700°C for each set of parameters (pre-irradiation treatment, gold-foil packing, irradiation-temperature) are shown in the following table. Sulphite, sulphate and volatile fractions are tabulated together as S^{x+}. While the sulphite-fraction was always less than 2% of the sulphate-content, the volatile part showed large fluctuations, especially with samples packed into gold-foil, (0.20% of the sulphate content) caused by the variations in sample surface and tightness of the package, even if mechanically destroyed before dissolving. The volatile species still remaining in the slat at dissolution, presumably as S_xCl_y, are immediately hydrolized to sulphate and only to a small extent to sulphite in the basic cyanide-carrier-solution. The following figure shows the temperature-dependence of the oxidation state distribution.

The experiments at 150° on NaCl treated with HCl-gas confirmed the existing published results (4.5), which were obtained with a slat dried in vacuo only.

The results of the investigations at higher temperatures and with molten samples show a monotone decrease of the S^{-2}-species and a corresponding increase of the S^{x+}-species with increasing irradiation-temperature. The content of S^0 is influenced neither by the pre-irradiation treatment not by the irradiation-temperature and is always about 20%. Melting of the samples during irradiation does not influence the distribution of oxidation states. The studies at 150°C show for NaCl/KCl-mixtures a shift of about 20% in S^{-2} towards higher oxidation-states, mainly S^0, compared to pure NaCl. No influence of the pre-irradiation sample treatment could be shown at 150°C, but it is of importance for work with molten samples. Untreated samples without gold-foil protection showed S^{-2}-levels of less than 2%. HCl-treatment increased this value to about 13%, an additional gold-foil protection to 26%. Oxygen of the quartz surface in contact with the melt is clearly significant at 700°C. The assumption that oxygen from the quartz surface should be of importance for reactions over the gas-phase is not plausible, without mentioning that oxygen-levels in the evacuated ampoules (10^{-5}, Hg) must be much greater but have no significant influence, as low temperature experiments show.

Salt-type	HCl-treatm.	Gold-packing	Irrad. temp. (^{o}C)	State	S^{-2} (%)	S^{0} (%)	S^{x+} (%)
NaCl	No	No	150	Solid	73	10	17(4)
NaCl	Yes	No	150		72 ± 2	13 ± 2	15 ± 2
NaCl/KCl	Yes	No	150		55 ± 2	25 ± 2	20 ± 2
NaCl/KCl	Yes	Yes	500		30 ± 2	19 ± 2	51 ± 3
NaCl/KCl	Yes	No	600		17 ± 3	22 ± 3	61 ± 4
NaCl/KCl	No	No	700	Molten	2 ± 1	22 ± 3	76 ± 5
NaCl/KCl	Yes	No	700		13 ± 3	22 ± 3	65 ± 4
NaCl/KCl	Yes	Yes	700		26 ± 4	10 ± 3	55 ± 4

Table 7.3 Oxidation state distribution of radiosulphr produced by ^{35}Cl(n,p)^{35}Cl in NaCl, NaCl/Kl solid mixtures and NaCl/KCl-melts

8. THERMOHYDRAULICS

8.1 Introduction

This chapter gives the results of studies on the thermal properties of the following units.

- High flux liquid fuel core with external heat exchanger. This core has the geometrical form of a spherical shell (Fig. 8.1A)
- Power-breeder-core with molten fuel and external heat exchanger. The core is spherical. (Fig. 8.1B).
- Sodium/molten salt heat exchanger.

All the calculations were made in simplified form and were intended only to roughly define the scope of the problems.

8.2 High Flux reactor with the core as a spherical shell

This reactor was designed to meet the following requirements

a) In the shell type core to achieve the highest possible flux in having a fast spectrum requiring a relatively high power density. Very good mixing in the fluid, i.e. a high turbulence is a vital requirement. For this the flow rate of the fuel has to be kept high.

b) In the central regions a thermal neutron flux is obtainded (Beryllium coated Deuterium hydroxide as moderator). The thermal neutron flux extends partly into the fast core which causes a very high and localised power density. This is the reason for requiring a high fuel turbulence.

c) In the termal zone having a neutron flux of approx 4×10^{16}n cm^2s^{-1} it is possible to transmute the most 'dangerous' fission products Strontium-90 and Caesium-137. Table 8.1 shows the most important parameters.

Fig. 8.1

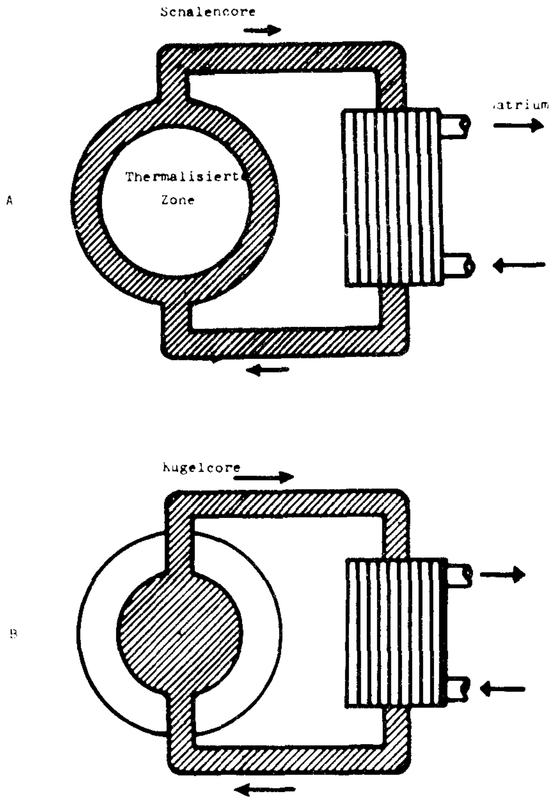


Fig. 8.2 Referenzfall Schalencore

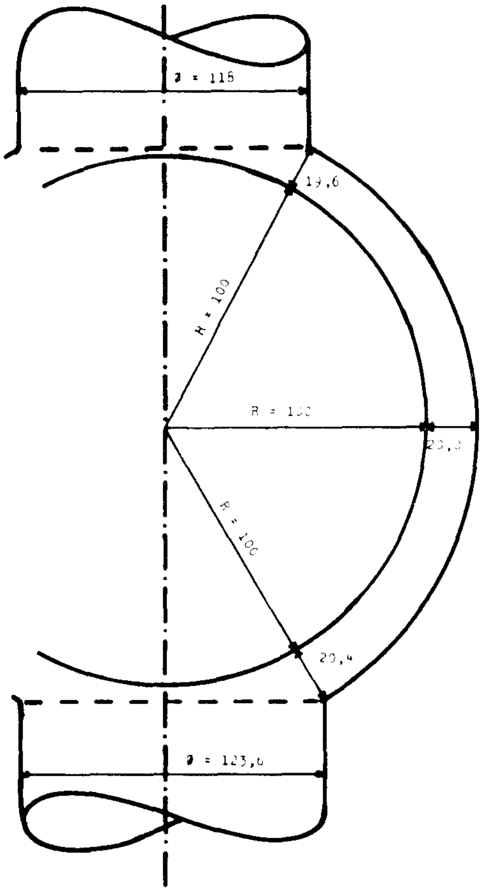


Table 8.1 Shell form High Flux reactor

Property	Unit	Value
Total power	GW_{tot}	6
Core Geometry	see Fig. 8.2	
Core volume	cm^3	3.1×10^6
Mean power density	$kW \cdot cm^{-3}$	2
Specific heat of the fuel (T in °C)	$J\ cm^{-3}\ K^{-1}$	$0.83\ (1-0.5 \times 10^3 (T-850))$
Density at 850°C	$8\ cm^{-3}$	2.12

The thermal properties of this core were calculated using the following parameters

- Flow rate at input 10-22 $m.s^{-1}$

- Diameter of the inlet pipe 0.5 - 0.6 m

  The inlet temperature was kept constant at 750°C
  The most important variable was the outlet temperature which was varied between 935°C and 965°C

For various reasons a reference concept was used as a basis for the calculations, see Fig. 8.2. A description of the reference design is also given in Fig. 8.3.

The relevant characteristics are given in the following figures:

Temperature curve	Fig. 8.4 A
Flow rate curve	Fig. 8.4 B
Mean circumference and shell thickness	Fig. 8.4 C
Number of Pu atoms in a layer	Fig. 8.5 A

The sudden changes at the start and end are dues to the uncertainties in the geometry at the pipe/shell interface.

Density Fig. 8.5 B

Fig. 3.3

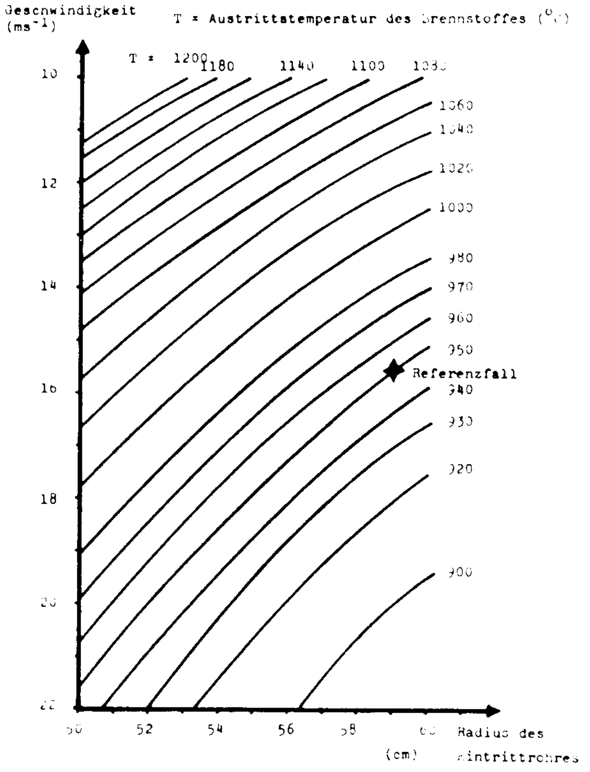


Fig. 3.4

Referenzfall Schalencore

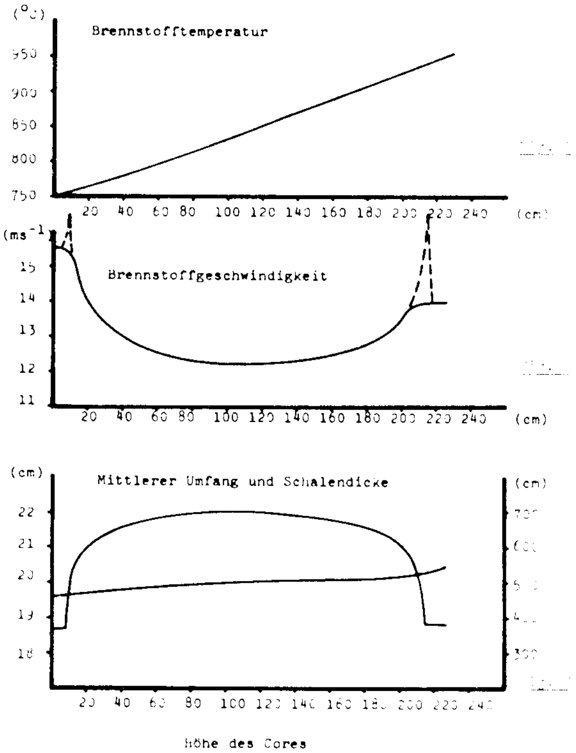


Fig. 3.5

Referenzfall Schalencore

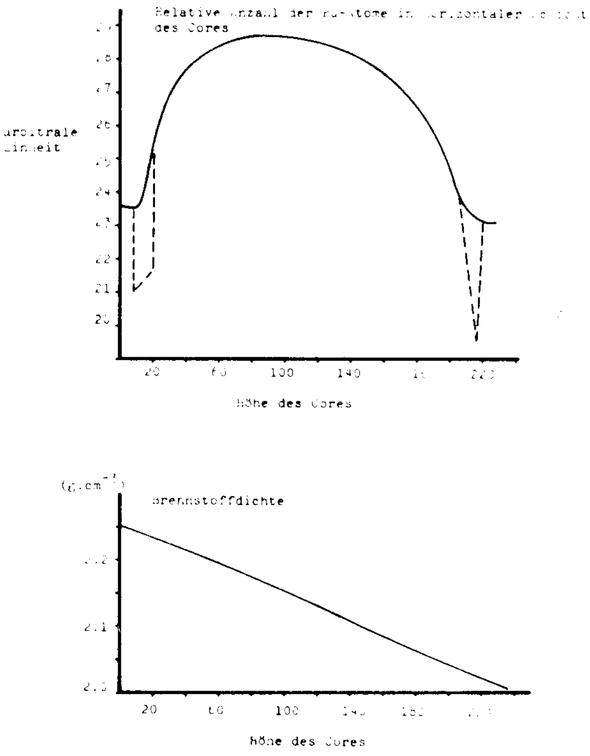
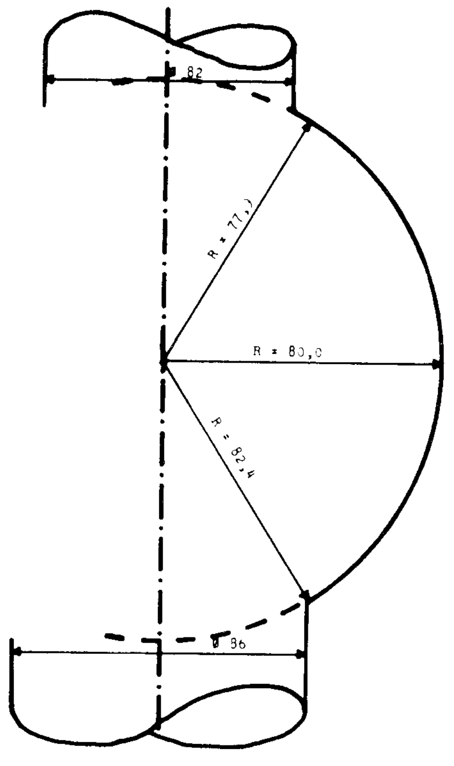


Fig. 3.6 Referenzfall Kugelcore



8.3 Power Reactor with spherical core

This reactor was designed to meet the following requirements

- a spherical core with the minimum of structural material in order to optimise the neutron balance and to achieve a hard spectrum.
- a maximum breeding ratio in the core and blanket
- the liquid fuel cooled in an external heat exchanger.

Table 8.2 shows the most important characteristics.

Table 8.2 Power breeding reactor

Characteristic	Unit	Value
Total power	GW(th)	3
Core geometry spherical radius		see Fig. 8.6
Core volume	cm^3	2.3×10^6
Mean power density	$kW \cdot cm^3$	1.3
Specific heat (T in ^0C)	$J \cdot cm^{-3}K^{-1}$	$0.83x(1 - 0.5x10^3(T-850)$
Density at 850^0C	$g \cdot cm^{-3}$	2.92

The thermal parameters of this core have been calculated varying the following parameters

- inlet velocity
- diameter of inlet tube

The inlet temperature was kept constant at 750^0C

The main parameter varied was the outlet temperature in the range 935^0C to 965^0C (see Fig. 8.7).

The given reference case (see Fig. 8.7) is shown in Fig. 8.6. The parameter changes under consideration are shown in figures 8.8 A, B, C and Fig. 8.9.

Fig. 8.7 **T = Austrittstemperatur des Brennstoffes (°C)**

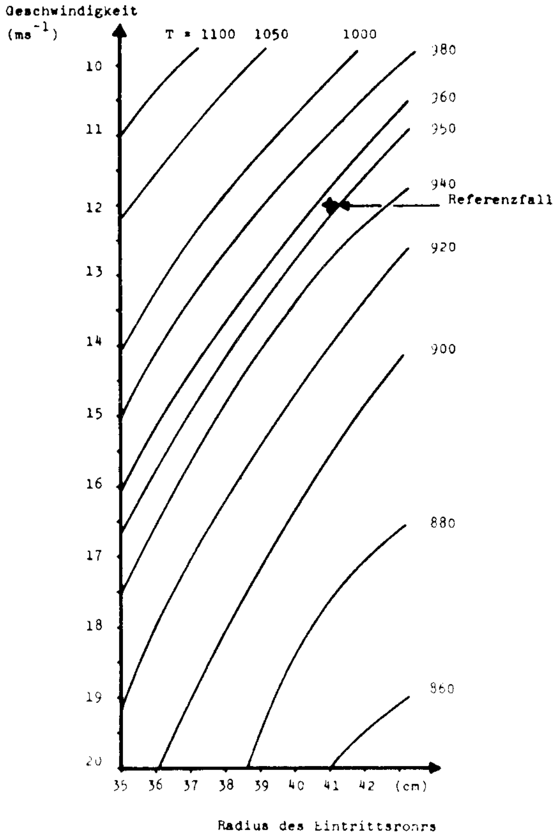


Fig. 8.8

Referenzfall Kugelcore

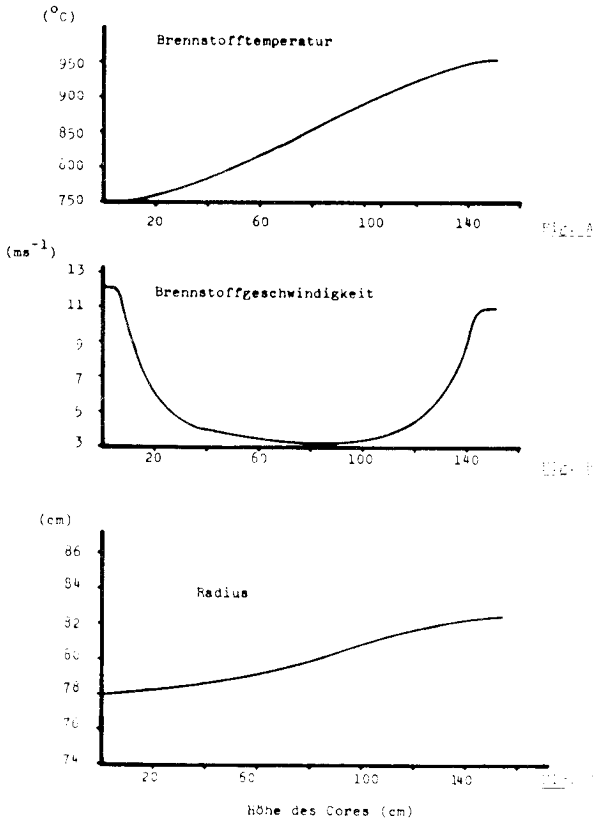


Fig. 8.9

Referenzfall Kugelcore

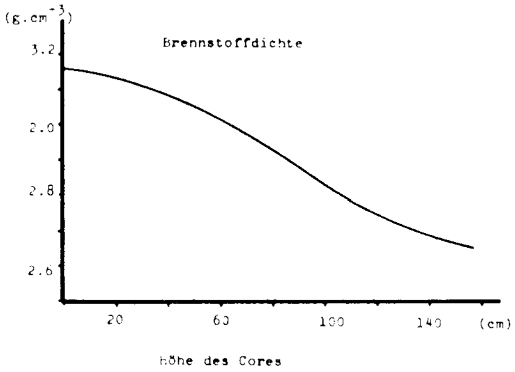
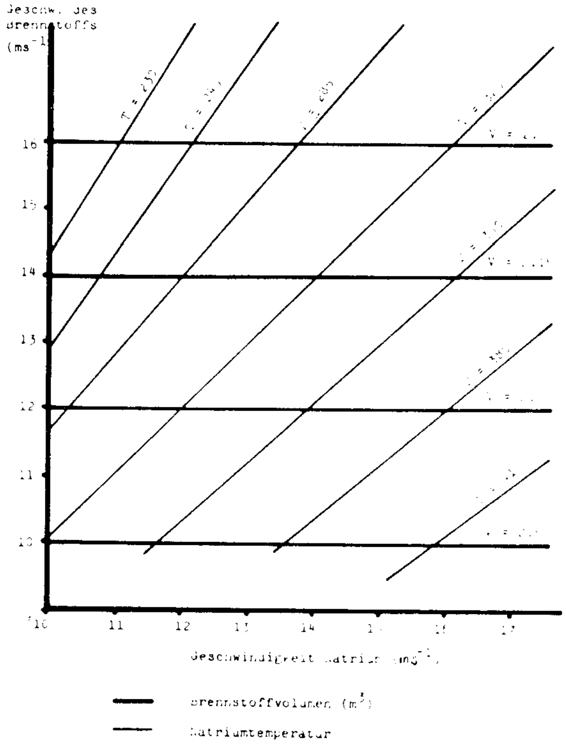


Fig. 8.10 V = totales Volumen des Brennstoffs in den [illegible]

T = Eintrittstemperatur des Natriums (°C)



8.4 The external heat exchanger

The most important parameter for the external heat exchanger was selected as the volume of fuel contained in it. The decisive factor is the minimisation of the fuel volume existing outside the core, which influences:

- the effective specific power (MW(th) per kg guel in the whole system)
- the effective doubling time
- safety considerations under accident conditions.
- minimisation of the loss of delayed neutrons.

For this reason a simple molten salt/sodium heat exchanger was chosen.

The most important properties of the two media are shown in table 8.3.

The following parameters were arbitrarily chosen

- Velocity of fuel 10-16 $m \cdot s^{-1}$
- Velocity of sodium 10-16 $m \cdot s^{-1}$
- Inlet temperature of the fuel 950°C
- Fuel outlet temperature 750°C
- Sodium outlet temperature 600°C
- Salt on the tube side dia 0.6 cm
- Sodium on the shell side 0.8/1.2 to 2.2 cm pitch

Important variables

- Inlet sodium temperature
- Length of tubes
- Volume of fuel in the tubes

All these data are given in Fig. 8.10 for the case of a pitch of 1.2 cm.

Table 8.3 Heat Exchanger: 3 GW thermal

Property	Unit	Fuel	Coolant
Composition	Mol	$1\ PuCl_3 \cdot 2\ UCl_3 \cdot 3.65\ NaCl$	Sodium metal
Density	gcm^{-3}	$3.526 \times (1-0.001\ (T-850))$	$0.784\ (1-0.001\ (T-350))$
Heat Capacity	$Jg^{-1}K^{-1}$	$1.0 \times (1.0.0005\ (T-850))$	$1.256\ (1-0.0005(T-350))$
Thermal conductivity	$Wcm^{-1}\ K^{-1}$	$0.015 \times (1.0.001\ (T-850))$	$0.663\ (1+0.001\ (T-350))$
Viscosity	$gcm^{-1}s^{-1}$	$0.05 \times (1-0.001\ (T-850))$	$0.0018\ (1-0.001\ (T-350))$
Heat transfer coefficient	$Wcm^{-2}K^{-1}$	$0.023 \times RE^{0.8} \times PR^{0.4}$	$5.3 + 0.019\ RE^{0.8} \cdot PR^{0.8}\ (R'/R'')^{0.3}$
Inlet temperature	°C	750	Variable (see Fig. 10)
Outlet temperature	°C	950	600
Inlet velocity	$m \cdot s^{-1}$	10 • 16	10 • 16

Fig. 8.11

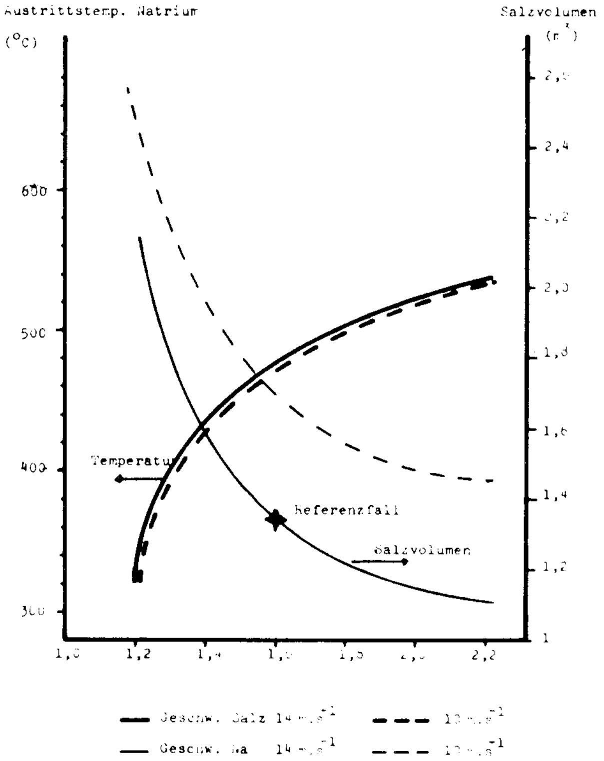


Fig. 8.12
Referenzfall

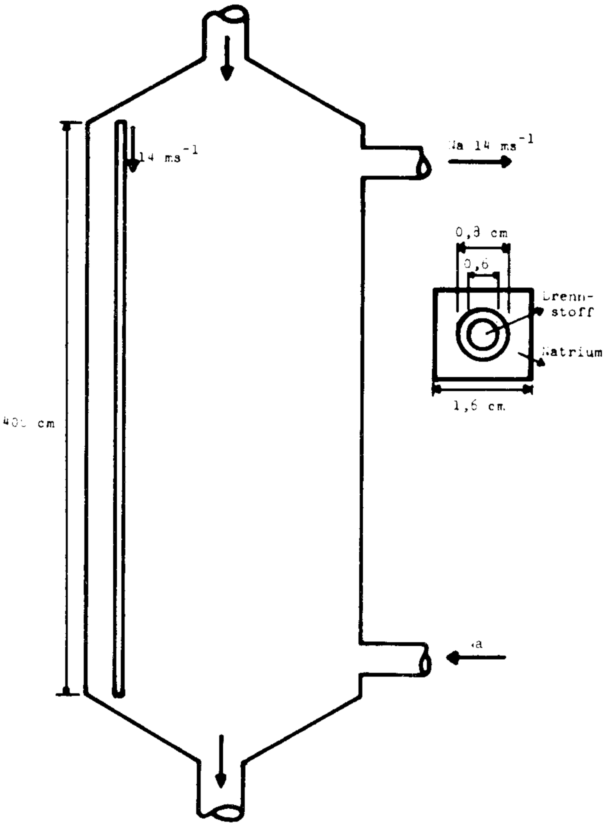
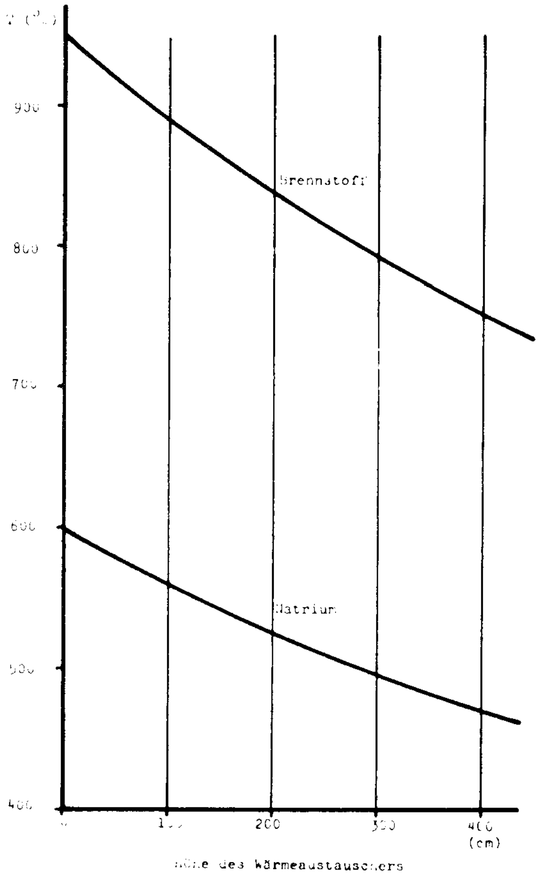


Fig. 8.13 Referenzfall



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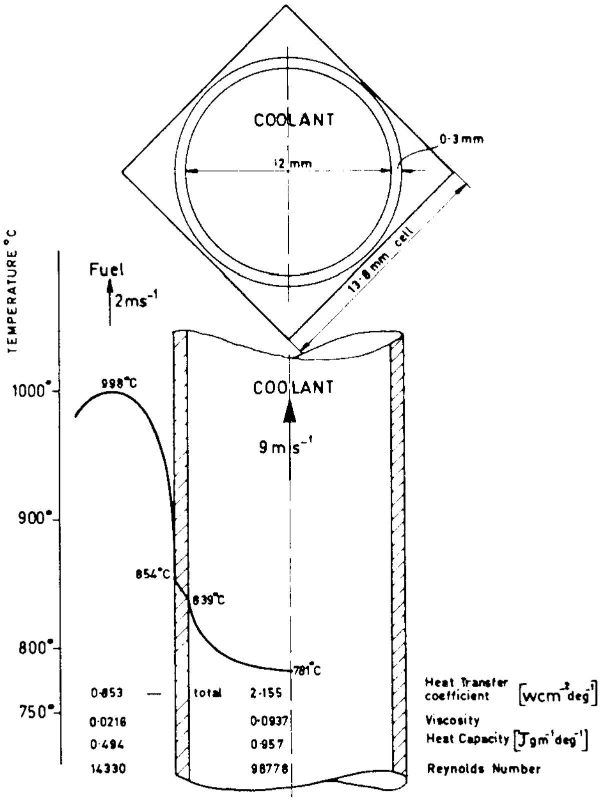


For other pitches between 1.2 and 2.2 and for two selected fuel and sodium velocities values can be seen in Fig. 8.11. The reference case was arbitrarily chosen (see Fig. 8.11). The geometry of the heat exchanger for this reference case is shown in Fig. 8.12. Further parameters are summarised in table 8.4.

Table 8.4 Heat Exchanger

Characteristic	Unit	Fuel	Metallic sodium
Inlet velocity	$m \cdot s^{-1}$	14	14
Viscosity	$g \cdot cm^{-1}\ s^{-1}$	0.0524	0.00183
Heat capacity	$J \cdot g^{-1} K^{-1}$	1.024	1.275
Number of tubes		5787	
Volume	cm^3	1.32×10^6	

The temperature curves for both media are given in Fig. 8.13.

8.5 The internally cooled reactor

This type of reactor, has fuel circulating in the core only, with a relatively low temperature gradient of approx. 35°C and a high heat capacity, and a high velocity coolant $9 m\ s^{-1}$ with again a low temperature gradient of approx. 43°C and high heat capacity. This, coupled with the very high negative temperature coefficient of reactivity results in an unusually high negative thermal and 'reactivity' stability.

Decrease of the fuel circulation and/or coolant velocity (in the U-tubes) results in a definite and "automatic" decrease of reactor power without recourse to engineered methods. This points to such a reactor being a surprisingly stable and self regulating device.

The achievement of the required fuel velocity in the core seems to require a forced circulation system since the rough estimate using natural convection gives a heat transfer coefficient which not acceptable.

Such a forced circulation system (core only) can be one of the following types - pump installed directly in the core, pump outside the core, an external pump with injector, a gas lift pump using inert gas. Consideration of the factors involved using criteria such as reduction of the out of core inventory, elimination of additional heat exchangers, minimisation of the fuel leakage, minimisation of the ausiliary power, optimisation of the fuel flow regulation - all point ot an in-core pump solution. of cource this gives rise to considerable technical problems (cooling of the rotor, corrosion and erosion, maintenance, neutron activation etc.).

The calculations for this type of reactor have been based on the following more or less arbitrarily selected parameters:

- fuel on the shell side, with tube pitch to diameter ratio equal to 1.10 to 1.18.

- fuel velocity: 0.5 to 5 $m \cdot s^{-1}$

- core dia: 2 and 2.2 m

- core height: 2 m

- coolant in tube with tube internal diameter equal to: 1.0 to 1.5 cm

- velocity of coolant: 1 to 17 $m\ s^{-1}$

- coolant inlet emperature 750° and 800°C

The neutronic calculation and thermo-hydraulics were made for 1 cm of the core height.

The detailed representation for the temperature distribution in a typical power reactor with a core output of 1.936 GW(th) are given in Fig. 8.14 (for a position 43 cm above the bottom of the core where the neutron flux is normalised to 1).

The bulk temperature of the fuel is here 988°C, the temperature of the tube walls 857 - 889°C and the bulk temperature of the coolant 781°C.

For the total output of the core 1.936 GW(th), the power distribution is as shown in Fig. 8.15.

Of course a flatter power ditribution could be obtained by adjusting tube deameters and pitch across the core. (Note that in this calculation the radial neutron flux distribution has been taken as unperturbed).

Fig. 8.15 Power distribution in the core

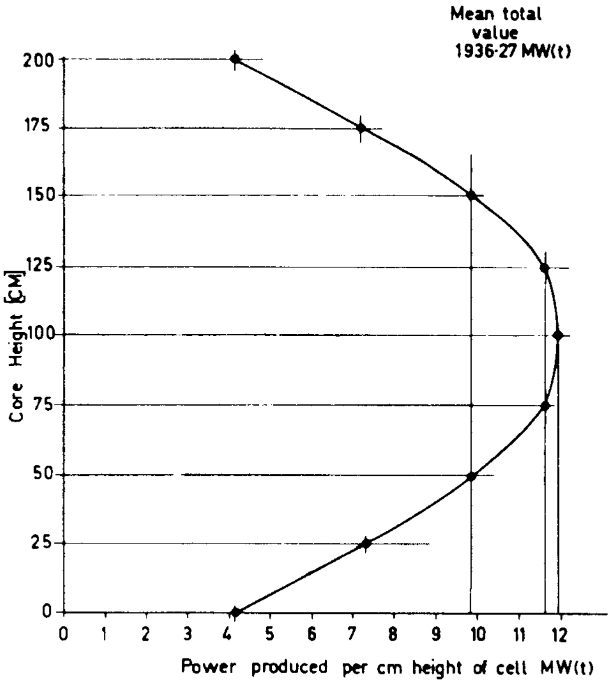


Fig. 8.16 Temperature of fuel and coolant reference core

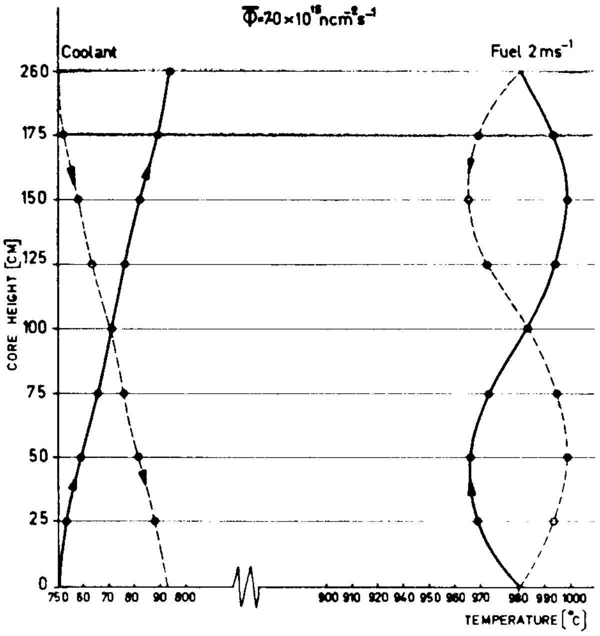


Fig. 8.17 Core power output versus coolant velocity

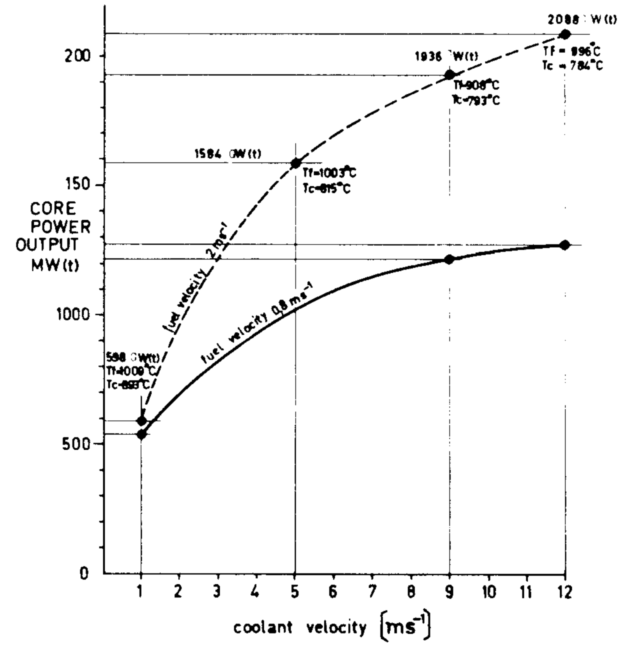
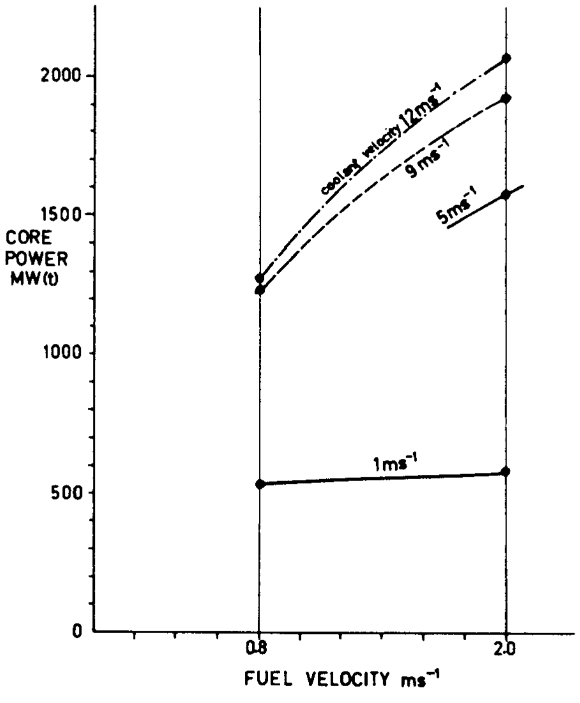


Fig. 8.18 Core power output versus fuel velocity



A very encouraging indication of the good temperature distribution with very small temperature gradients is shown in Fig. 8.16 which indicates the axial bulk temperature distribution in the fuel and in the coolant in the core.

The fuel bulk temperature changes from 980°C at the bottom to 965°C at 1/4 core height and is 998°C at 3/4 of core height. The coolant temperature lies between 750°C inlet and 793°C outlet. Both these small temperature gradients in the fuel and in the coolant (fertiel material) may prove beneficial in reducing corrosion processes due to the minimizing of mass transport phenomena.

The stable behaviour of this type of reactor results from many parameters. Two of them are the velocity of the coolant and its bulk temperature. The mean power output of the core is strongly dependant on the velocities of both fuel and coolant. Fig. 8.17.

For a fuel velocity of 2 $m \cdot s^{-1}$, when the coolant velocity falls from 12 $m \cdot s^{-1}$ to 1 $m \cdot s^{-1}$ the coolant outlet temperature increases from 784°C to 893°C for constant inlet temperature of 750°C. This change of coolant velocity and its bulk temperature results in the decrease of the mena core output from 2.088 GW(th) to 0.598 GW(th) - that is approximately a factor 3! It is clear since the lower coolant velocity results in a higher coolant outlet temperature and lower power output we have definite negative temperature coefficient (power output) varying with the given coolant velocity.

If the fuel velocity falls from 2 $m \cdot s^{-1}$ to 0.8 $m \cdot s^{-1}$ we again get an important decrease of power output (see Fig. 8.18). The decrease in both fuel and coolant velocity results in a sharp decrase of reactor power. This means that such a reactor can be considered as a surprisingly stable and self regulating device. In the case of a sudden fall in coolant and/or fuel velocities the power output decreases to a safe level without intervention.

The achievement of the required fuel velocity in the core seems to require a forced circulation system since the rough estimate using natural convection gives a heat transfer coefficient which is too low.

Such a force circulation system (core only) can be one of the following types

- pump installed directly in core

- pump outside the core

- an external pump with injector

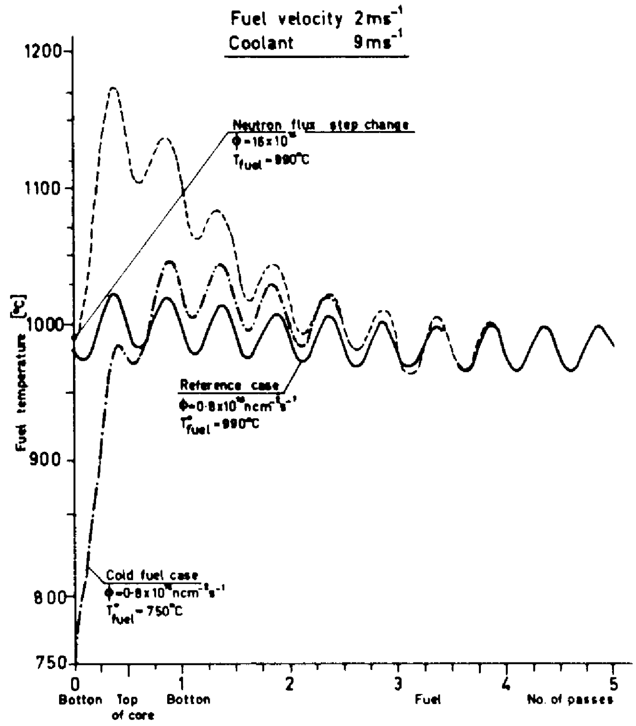
- a gas-lift pump using inert gas (argon)

Intensive consideration of the factors involved, using criteria such as - reduction of the out of core inventory, elimination of additional heat exchangers, minimization of the fuel leakage, minimization of the auxilliary power, optimisation of the fuel flow regulation, points to an in-core pump solution. Of course this gives rise to considerable technical problems (cooling of the rotor, corrosion and erosion, maintenance, neutron activation etc.).

The postulted fuel velocity makes it possible to make some calculation of the heat transfer problems and also gives a feel for the kinetics of the reactor under discussion.

It must be stressed that these kietics studies have no strong physical sense and use an itterative approach but it is clear that they give some useful information about the general reactor stability. (see Fig. 8.19).

Fig. 8.19 Fuel temperature during several passes through core under different inital conditions



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