Molten Chlorides Fast Breeder Reactor
Problems and Possibilities

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EIR-Bericht Nr. 215

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June 1972

Contents

Summary

A fast breeder reactor of 2000 MW(t) output using molten chlorides as fuel and coolant is discussed. Some of the most significant characteristics are

- the fuel contains only $PuCl_3/NaCl$,

- the coolant is $UCl_3/NaCl$ and also forms the fertile material along with the blanket, again $UCl_3/NaCl$

- the fuel circulates through the core by forced convection.

The thermal stability of the reactor is very good. Power excursions of fuel temperature transients are quickly damped by the phenomena of fuel expansion pushing part of the fissile material out of the critical zone.

The balance of fission products including free chlorine seems to be stabilized when some of the semi-noble metals (Ru, Rh, Pd and some Mo) are present in the elementary form.

Corrosion effects form the most difficult problem. Thermodynamic studies suggest the use of molybdenum alloys as structural materials. Separation of the fuel ~~component~~ component from the fertile component UCl_3 helps to overcome some of the corrosion problems.

A reprocessing system based on a salt-metal-transport process seems to be attractive from the point of view of the economics of the plant as a whole.

The possiblity of using a dissociating gas as a secondary working agent in the turbine for example Al_2Cl_6 and N_2O_4 is discussed.

Résumé

SURREGENERATEUR RAPIDE A SELS <u>FONDUS</u> (Chlorures)

Problemes et possibilités

Un surrégénérateur rapide de 2000 MW(t) utilisant des chlorures fondus comme combustible et comme réfrigérant est décrit. Quelques-unes de ses caractéristiques les plus intéressantes sont les suivantes:

- le combustible liquide, contient seulement un mélange $PuCl_3$/NaCl

- le réfrigérant est un mélange UCl_3/NaCl; il comporte donc le matériau fertile tout comme les couvertures de même composition

- le combustible demeure dans le coeur mais doit être entraîné suivant une boucle de convection forcée.

La stabilité thermique du réacteur est très bonne; les excursions de puissance et les transitoires concernant la température du combustible sont fortement limités par l'expansion du combustible qui se trouve déplacé vers les régions périphériques qui contribuent beaucoup moins à la réactivité.

L'équilibre des produits de fission incluant des atomes de chlore semble être réalisé quand certains métaux semi-noble (Ru, Rh, Pd et Mo) sont présents.

Les effets de corrosion constituent le problème le plus difficile. Des études de thermodynamique suggèrent l'utilisation d'alliages de molybdène comme matériau de structure.

Un système de régénération basé sur un procédé de "transport sel-métal" paraît intéressant du point de vue économique.

Enfin la possibilité d'utiliser les propriétés de dimérisation de certains gaz, par exemple Al_2Cl_6 et N_2O_4, dans le cycle thermo-

1. General Problems

The biggest attraction of breeder reactors is their ability to utilize directly or indirectly, the non-fissile nuclides U-238 - in fast breeder reactors, and Th-232 - in thermal reactors.

The relative advantages of the fast breeder over the thermal breeder are: higher breeding ratio, higher specific power which results in the shortening of the doubling time as well as in the more intensive use of the reactor volume.

In addition it must be emphasized, that each breeder reactor, which is built to last 25-30 years, must be considered not only as a power producing device but also as a source of fissile material. Therefore each breeder reactor should be considered as part of a long term complex ' breeding system ' which includes both the power reactor and the fuel reprocessing plant over this period.

From this point of view the reactors with molten fuel are better adapted to the long term ' breeding system ' than are the solid fuel reactors. (simpler reprocessing technology, minimal transportation problems, smaller environmental danger, better economics(?)). (Fig. 1)
Such a coupled 'breeder system' has additional advantages when the reprocessing technology is based on high temperature processes, such as pyrometallurgical or pyrochemical techniques instead of low temperature processes in aqueous solutions. These high temperature reactor reprocessing systems might be realised in the most favorable manner when the fuel in the breeders is in a molten liquid and not in a solid state.

Thus in the future, 'breeding systems'using molten fuel fast reactors seem to be of interest.

The molten fuel fast reactors can be classified in the following
manner

1) Molten metallic fuel

2) Molten salt fuel

Fast reactors with molten metallic plutonium fuels were constructed
in the sixties in Los Alamos under the name LAMPRE. The molten
alloy Pu-Co-Ce was the most promising fuel proposed. The results
were encouraging but further experiments have not been realised.
These reactor types were not breeders.

Fast reactors with molten salt fuel exist only as paper studies at
the present time. Som·· work has been done at Oak Ridge (1956),
Warsav (1960-68) Argonne (1965-68) and Harwell (1963-70). In addi-
tion some experimental work has also been carried out.

When nuclear, physical and chemical considerations are studied it
can be shown that the only possible fuel constituent of these fast
reactors are the molten chlorides. In the case of thermal reactors
the most suitable molten salt fuel proves to be a fluoride.
(Fluorides are moderators, and therefore could not be used in a
fast reactor, because of the dramatic softening of the neutron
spectrum).

The search for the best components of molten salt fuel for fast
reactors must take into account not only thermal and hydraulic
properties but also the following nuclear properties: elastic and
inelastic cross sections, cross sections for neutron absorbing
mechanisms, not only (n,γ) but also (n,p), (n,α) and $(n,2n)$. The
chlorides of uranium-238 and plutonium-239 diluted by sodium chlo-
ride are the selected components of the fused salt fuel. (Taube 1961)

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

a) <u>Externally cooled</u>, 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 outside the core does not of course contribute to the critical mass.
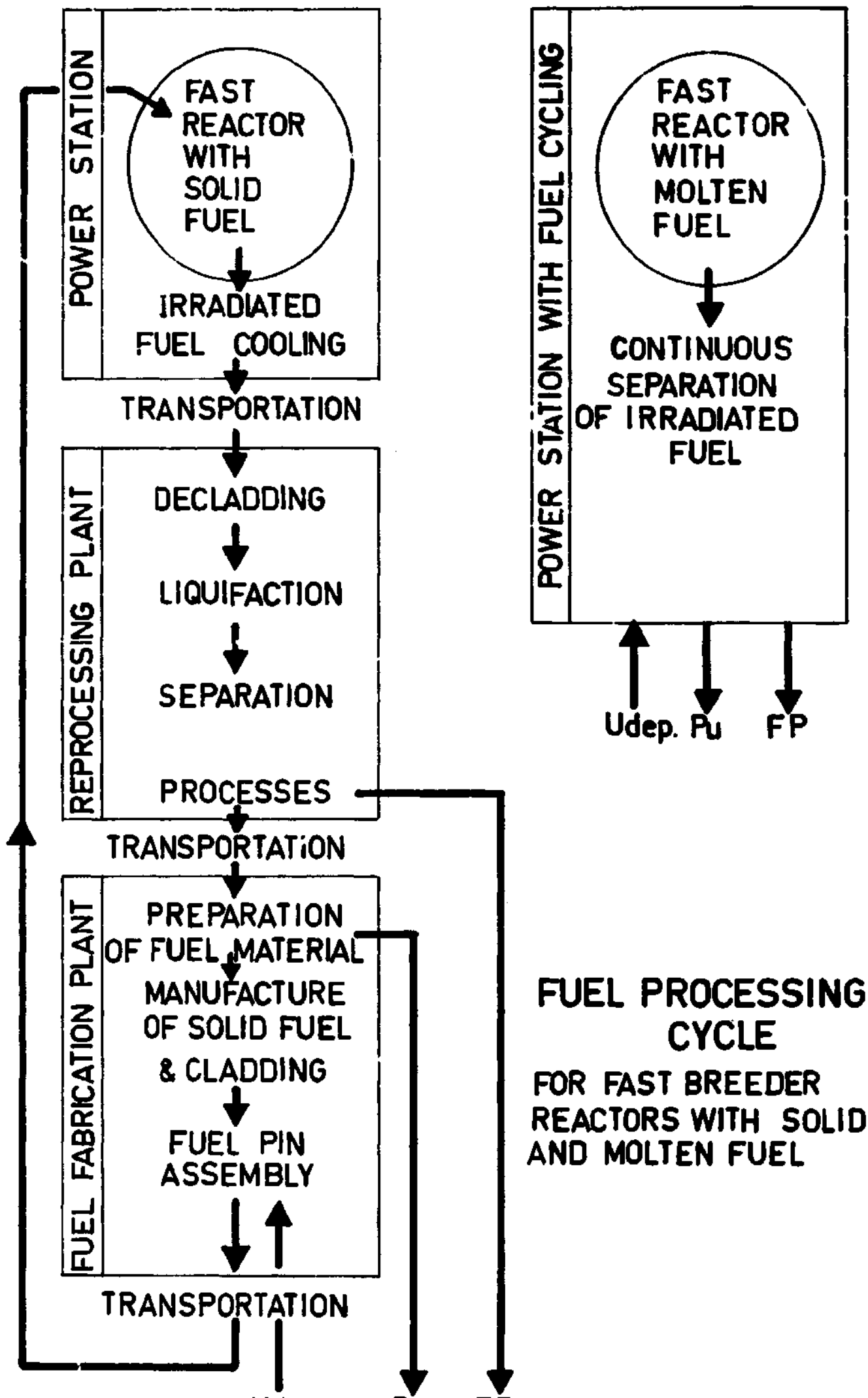
This type of reactor has been discussed for example by Nelson, (Argonne 1967) and Lane (USA 1970) especially as a high flux material testing fast reactor.

In externally cooled fast reactors the loss of a portion of the delayed neutrons could adversly affect reactor control. Also the biological shielding outside the core is very expensive.

b) <u>Internally, direct cooled reactors:</u> 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 studied eg. - cooled by molten lead (Long, Harwell and Killingback, Winfrith), cooled by boiling mercury (Taube, Warsaw) and cooled by boiling aluminium chloride (Taube, Warsaw). This type of reactor must be considered as an 'extreme exotic type'.

POWER STATION

FAST REACTOR WITH SOLID FUEL

IRRADIATED FUEL COOLING

TRANSPORTATION

REPROCESSING PLANT

DECLADDING

LIQUIFACTION

SEPARATION

PROCESSES

TRANSPORTATION

FUEL FABRICATION PLANT

PREPARATION OF FUEL MATERIAL

MANUFACTURE OF SOLID FUEL & CLADDING

FUEL PIN ASSEMBLY

TRANSPORTATION

POWER STATION WITH FUEL CYCLING

FAST REACTOR WITH MOLTEN FUEL

CONTINUOUS SEPARATION OF IRRADIATED FUEL

Udep. Pu FP

FUEL PROCESSING CYCLE

FOR FAST BREEDER REACTORS WITH SOLID AND MOLTEN FUEL

c) <u>Internally indirectly cooled reactor:</u> 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).

In this paper an internally indirectly cooled molten salt fast
reactor is discussed. The unusual difference is in the use of
a molten chloride coolant, including uranium chloride in place
of sodium or gaseous coolant. The uranium chloride component
is in fact the fertile constituent which doubles as coolant.

Fig. 1 shows the flow diagrams of two types of reactors: with
solid fuel and separate reprocessing plant and the molten fuel
reactor with integral reprocessing system.

2. General description of the reactor

In this paper a molten chlorides fast breeder reactor is discussed.
The most important features of this reactor are: (Fig. 2 and Fig. 3)

- thermal power 2050 MW(th) - 1936 MW(t) in core + 114 MW(t) in
  blanket giving

- electrical power 1000 MW(e) (in the most optimistic case)

- 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 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: the same as fertile material
(no other coolant in core)

- blanket material: the same as fertile material

- the core is internally cooled, there is no circulating fuel
outside the core.

- the fuel and the coolant are flowing in the same direction
(see fig. 3)

- the reprocessing plant is in intimate proximity with the
reactor (under the same roof)

- for the sake of obtaining relatively high plant thermal effi-
ciency a rather exotic working agent is proposed: aluminium
trichloride. This agent removes the heat from the primary
coolant (molen UCl_3/NaCl) in an external heat exchanger.

- for full use of the thermal energy a secondary working agent
is also proposed: nitrogen dioxide. The theoretical thermal
efficiency of such a power station may be significantly higher
than those of a 'classical' power station. Of course a more
conservative steam turbine system need not be excluded, but it
is possible to argue that one attraction of such system would
be to use for 'district' heating.

- the fuel in the core and the coolant are pumped with the velo-
city of 2 and 9 mS^{-1} respectively.
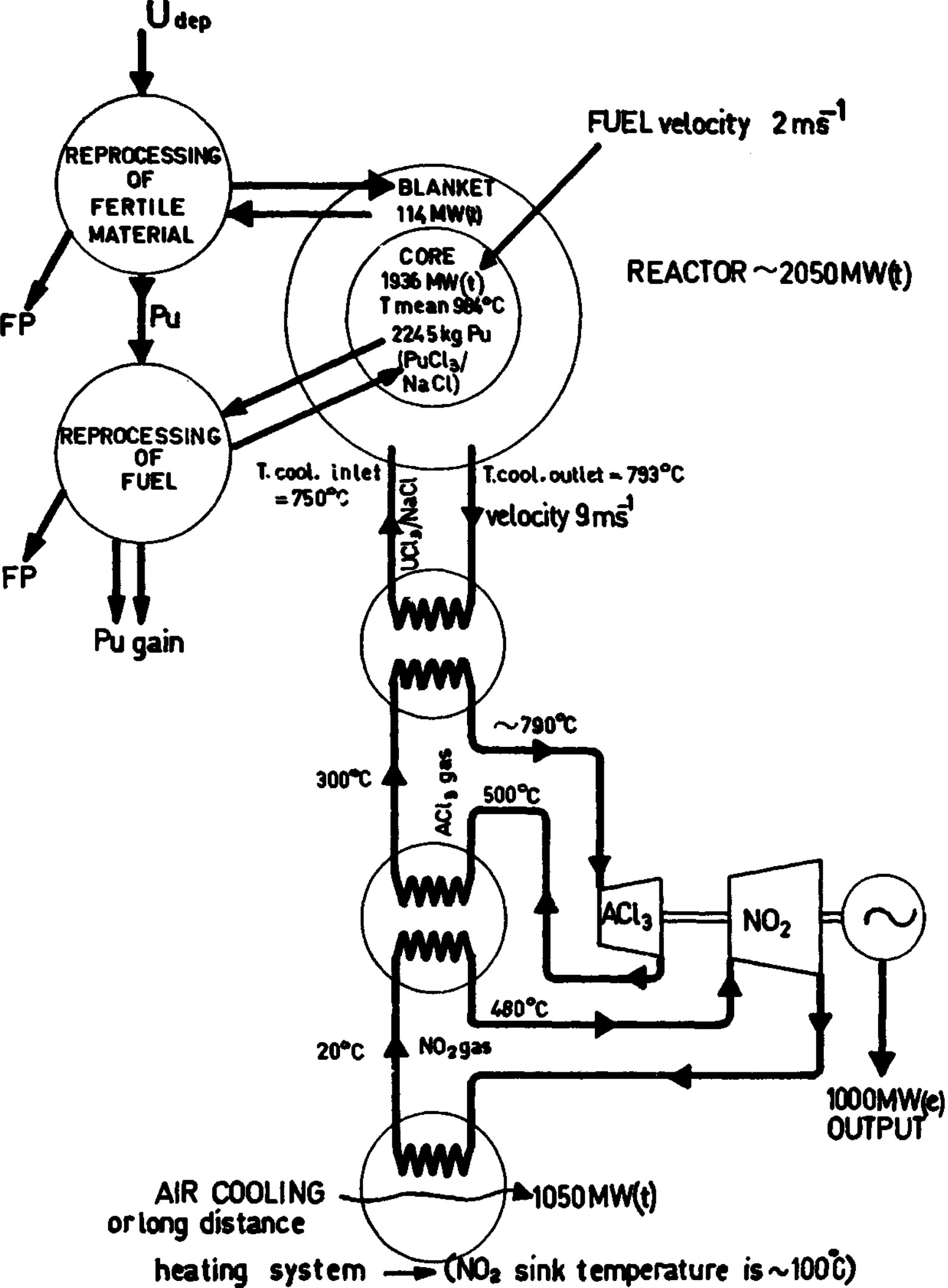
Fig. 2. MOLTEN CHLORIDE REACTOR PLANT

FUEL REPROCESSING PLANT

PUMP

OVERFLOW for FUEL EXPANSION

COOLANT UCl₃/NaCl

0.8 m

2 m

1 m

HEAT EXCHANGER

CORE 7·62 m³

Fuel PuCl₃/NaCl

BLANKET WITH MOLTEN FERTILE MATERIAL UCl₃/Na Cl

HEAT EXCHANGER

1 m

COOLING TUBES 12mm in Dia

2·2 m

No. of Tubes 19940

COOLANT UCl₃/NaCl

HEAT EXCHANGER FOR BLANKET
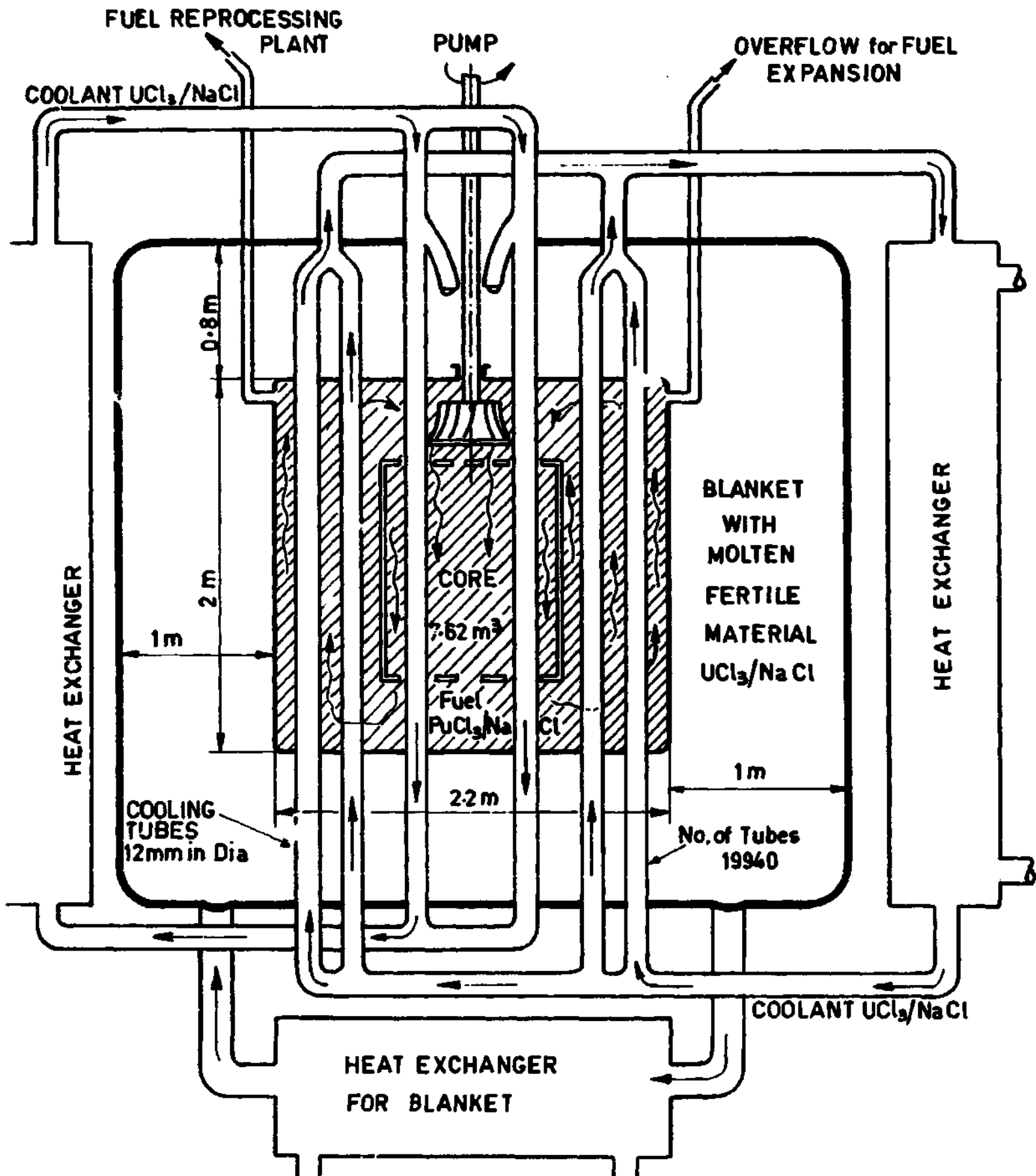
Fig 3   CORE AND BLANKET
SCHEMATIC

- the possible structural material: molybdenum alloy with small
  amounts of other metals e.g. Ni, Fe .....

The advantages of the proposed reactor are the following:

- 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 because of the small out-of-core inventory
  on the basis of the direct coupled continous reprocessing plant.

- the fuel contains only plutonium and no uranium which simpli-
  fies the processing technology and removes the danger of uranium
  trichloride oxidation which also improves the corrosion proper-
  ties of this medium.

- the corrosion problems are easier to solve when the aggressive
  turbine working agent, aluminium chloride is situated outside
  the core.

- the high velocities of both fuel and coolant significantly
  reduces the temperature gradients at the equilibrium state and
  reduces the mass transport mechanism. The mass transport mechanism
  is very sensitive to temperature gradients and plays a large
  part in corrosion mechanisms.

However the disadvantages are numerous:

- the first and most important disadvantage is of course corro-
  sion. The molten chloride medium, especially in neutron and
  gamma fields, at high temperatures and velocities with chlorine

being virtually free in the fission process of 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 things gives rise to parasytic 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 coolant velocities result in high pumping costs and could cause severe erosion.

Table I

Molten chloride fast breeder reactor 'CHLOROPHIL'

Electrical power (approximate estimate)	MW(e)	1000
Thermal power, total	MW(t)	2050
- in core	MW(t)	1936
- in blanket (approx)	MW(t)	114
Core volume	m^3	7.62
Specific power (core)	MWm^{-3}	255
Core structure	see fig. 3	
Plant efficiency (estimate)	%	49*
Liquid fuel properties		
$PuCl_3$	mol %	15
NaCl	mol %	85
Liquidus point	^oC	685
Boiling point (at 1 bar)	^oC	1500
Fuel temperature (mean)	^oC	984
Fuel volume in core	m^3	2.66

Density at $984^{o}C$	kgm^{-3}	2340
Heat capacity (for $984^{o}C$)	$KJ.kg^{-1}deg^{-1}$	0.95
Viscosity ($984^{o}C$)	$g.cm^{-1}s^{-1}$	0.0217
Thermal conductivity (for $750^{o}C$)	$W.cm^{-1}deg^{-1}$	0.007
Fuel in core	kg	6210
Total plutonium in core	kg	2252
Total plutonium in salt	weight %	36.4
Total plutonium in entire system	kg	2500
Mean plutonium specific power (core)	$MW(t).kg^{-1}$	0.86
Plutonium specific power (entire system)	$MW(t).kg^{-1}$	0.775

Coolant Properties

$(U_{238})Cl_3$ (depleted uranium)	mol %	65
	weight %	92
NaCl	mol %	35
U_{238} in salt	w %	63.5
Liquidus point	^{o}C	710
Boiling point	^{o}C	1700
Coolant temp. inlet	^{o}C	750
Coolant temp outlet	^{o}C	793
Coolant fertile salt in core	m^3	4.96
density	$kg.m^{-3}$	4000
in core	kg	20000
Coolent fertile salt total (blanket + core)	m^3	46.14
	kg	185000
Uranium inventory (blanket + core)	kg	171000
Uranium inventory reactor + reprocessing	kg	180000
Molybdenum alloy (80% Mo) in core	kg	3000

Core geometry - cylindrical

height	m	2.00
diameter	m	2.20
volume	m^3	7.62

Axial blanket height	m	0.80
Radial blanket thicknesses	m	1.00
Blanket + in core coolant (tubes) volume	m^3	38.25

Thermo-hydraulics

Fuel velocity	$m.s^{-1}$	2.00
Fuel pump		in core
Fuel		shell side
Coolant velocity	$m.s^{-1}$	9.00
Coolant pump		in external heat exchanger
Number of coolant tubes		19941
Tubes — inner dia	cm	1.20
— outer dia	cm	1.26
— tube material		Mo. alloy
— tube pitch	cm	1.38
pitch/inner dia ratio		1.15
Secondary working agent		$AlCl_3$ gaseous

Reprocessing

		continuous
Efficiency for Pu + F.P. separation (assumed)	%	50
Fuel stream to plant	$kg.s^{-1}$	0.003
Mean cycle time for fuel	days	21
Fertile stream to plant	$kg.s^{-1}$	0.216
Mean cycle time for fertile medium	days	56

Breeding ratio

Internal (in core only)**		0.709
Outer (in blanket)		0.680
total		1.389
Doubling time	years	9.2

* Total station efficiency only roughly estimated

** Neutron calculations neglecting parasitic absorbtion by vessel structive.

3. Reactor Physics

The neutronics calculations have been made in 3 steps:

- rough calculation with one group cross sections taken from sodium cooled oxide fuel fast reactor data.

- calculation with 15 group cross sections for given chloride composition.

- recalculation with newly calculated one group cross sections, normalized from spectrum obtained in the previous 15 group calculation. (see fig. 4)

The one group cross sections used in these calculations are given in table 2.

The neutron balance is given in table 3.

There are references in the literature to the adverse effect of neutron absorption by the chlorine isotopes.

For fast neutrons the two isotopes of chlorine have the following cross-sections.

Nuclide	% isotope in natural Cl	fast neutron cross sections	value (barns)
Cl-35	75.53	$\sigma(n,p)$ $\sigma(n,\alpha)$	0.072
Cl-37	24.47	$\sigma(n,p)$ $\sigma(n,\alpha)$	0.0015

From the data obtained the adverse influence of chlorine-35 is rather small and an isotopically enriched chlorine is not required. Such a suggestion was made by Weinberg and Wigner (1955) but was

Table 2

One group cross-sections for the molten chloride fast reactor on the basis of 15 group data of Bondarenko.

Nuclide	σ_f	$\sigma_{capture}$	γ
Pu-239 (and Pu-241)	1,826	0,256	2,962
Pu-240	0,546	0,437	2,660 (estimated)
U-238	0,0743	0,3145	2,660
Na	-	0,00133	-
Cl	-	0,015	-
Mo	-	0,0743	-
Fission products	-	0,273	-

Table 3

Neutron balance for 1 cm height of cell (1.91 cm^3)

Nuclide	x10^{20}		Neutrons produced	Neutrons absorbed
Pu-239	11.36	(n,f)	61.4	20.7
		(n,γ)	-	2.9
Pu-240	2.85	(n,f)	4.14	1.55
		(n,γ)	-	1.24
U-238	72.39	(n,f)	14.3	5.40
		(n,γ)	-	15.53
Na	104.00		-	0.13
Cl	375.00		-	5.62
Mo	56.00		-	4.2
F.P.	1.13		-	0.3
			79.8	57.57

$$K_\infty = \frac{79.8}{57.57} = 1.387$$

To a similar extent the problem of the exact choice of constructional materials for the coolant tubes will also affect the neutronics calculations.

At least two criteria must be considered in the selection of tube material:

- the absorption cross-section for fast neutrons ($\sigma_a \approx 0.1$ MeV) because of its effect on the breeding ratio. (fig. 5)

- chemical stability against the attack of chlorine ions which can be partly represented by the free enthalpy of chloride formation ($G_{form}^{1000 \, ^{\circ}K}$).

Fig. 5 shows both properties of some selected materials. From this it appears that one of the most suitable metals is molybdenum. Unfortunately this answer cannot be regarded as completely satisfactory because of the lack of real experimental data and because of some thermodynamic questions which are discussed in sections 8 and 9.
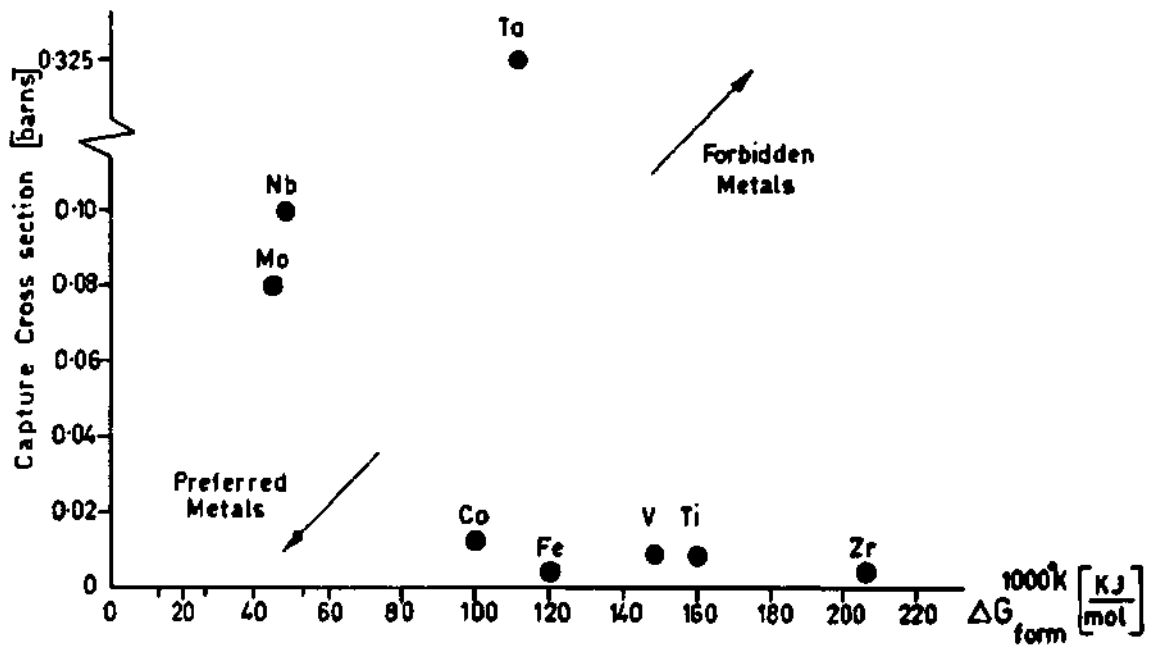
4. Thermohydraulics

The calculations for this type of reactor have been based on the following more or less arbitrary selected parameters:

- fuel in shell side, with tube pitch to diameter ratio equal to 1.10 to 1.18.

- fuel velocity: 0.5 to 5 m.s^{-1}

- core dia: 2 and 2.2 m

- core height: 2 m
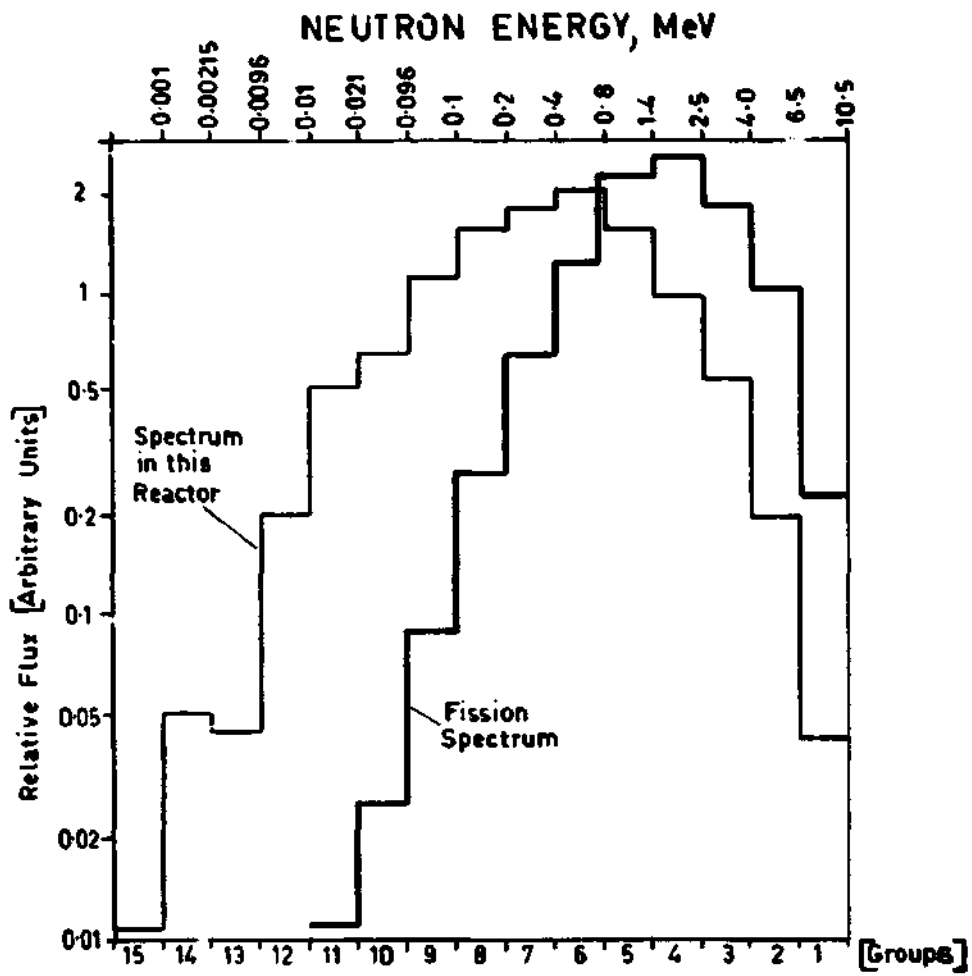
Fig 4 NEUTRON SPECTRA

Fig 5 CHOICE OF STRUCTURAL MATERIALS

- coolant in tube with tube internal diameter equal to: 1.0 to 1.5 cm

- velocity of coolant: 1 to 17 ms^{-1}

- coolant inlet temperature 750° and $800^{o}C$

The calculation of neutronics and thermo-hydraulics were made for 1 cm of the core height (see fig. 6 - flowsheet of program)

The data given in table 1 obtained from these calculations are for a steady state reactor.

The detailed representation for the temperature distribution in a typical power reactor with a core output of 1936 MW(t) are given in fig. 7 (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 $998^{o}C$, the temperature of the tube walls $857 - 839^{o}C$ and the bulk temperature of the coolant $781^{o}C$.

For the total output of the core 1936 MW(t), the power distribution is as shown in fig. 8.

Of course a flatter power distribution could be obtained by adjusting tube diameters and pitch across the core.

(Note that in this calculation the radial neutron flux distribution has been taken as unperturbed).

A very encouraging indication of the good temperature distribution with very small temperature gradients is shown in fig. 9 which indicates the axial bulk temperature distribution in the fuel and in the coolant in the core.

X = height of core x no. of passes [cm]

D = density of component

C = heat capacity

V = viscosity

K = heat conductivity

G = geometry of channels

ϕ = neutron flux

E = energy produktion

H = heat transfer

Tf = temperature of fuel

Tc = inlet temperature of coolant

U = velocities of fuel and coolant

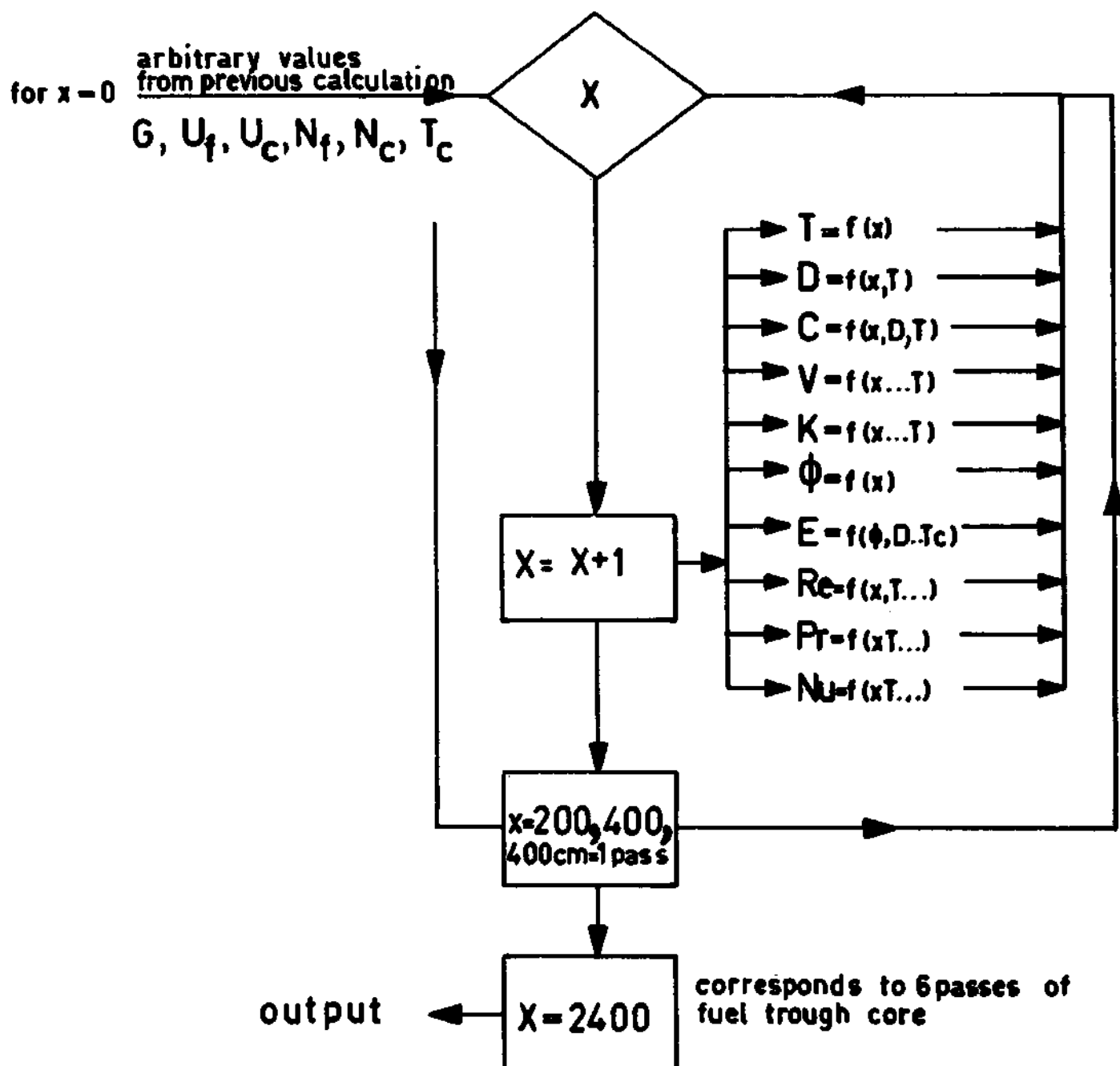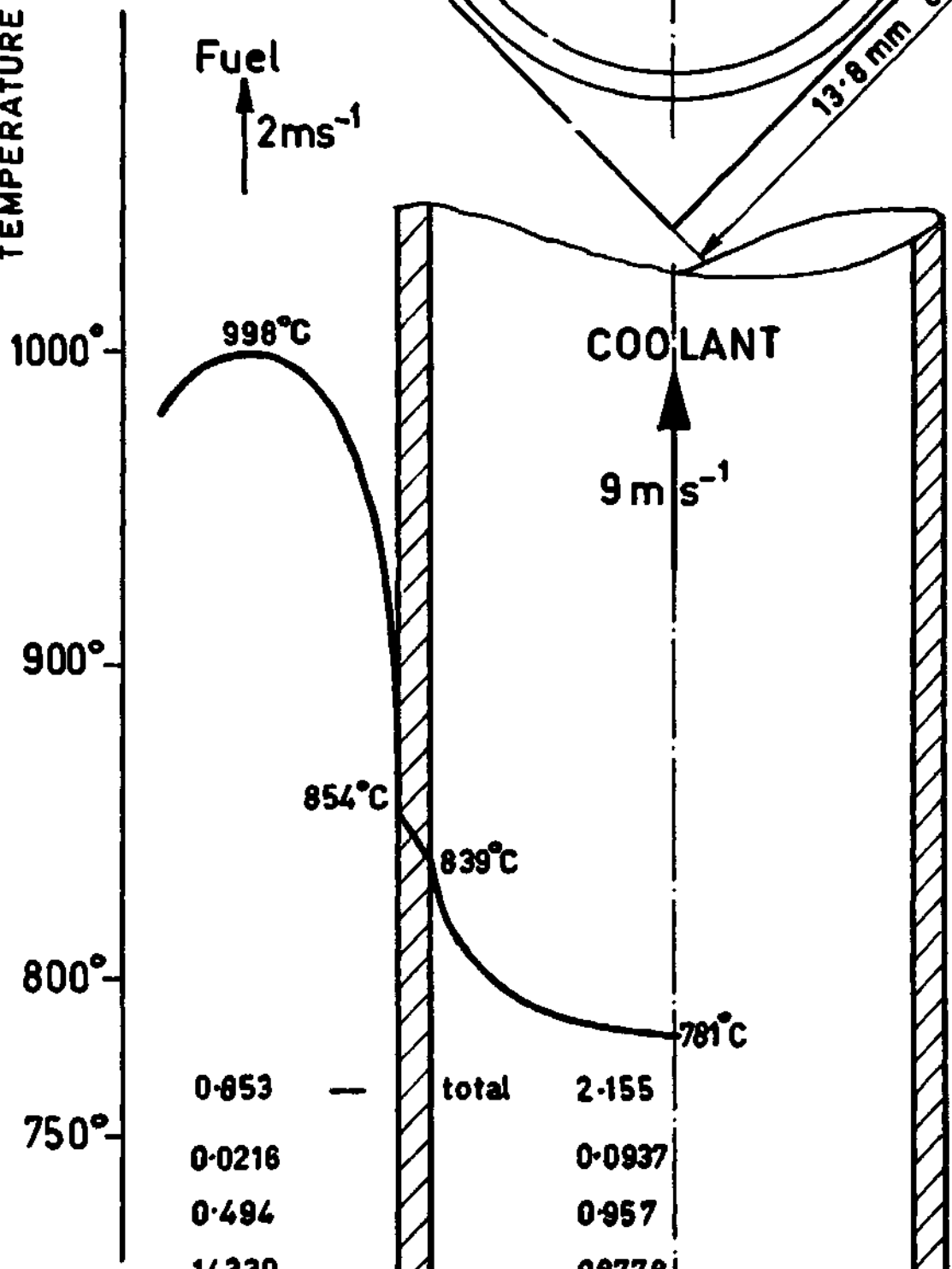Nf = fuel composition

Nc = coolant composition

Re = Reynolds number

Pr = Prandtl number

Nu = Nusselt number

for x = 0   arbitrary values
from previous calculation

$G, U_f, U_c, N_f, N_c, T_c$

X

$T = f(x)$

$D = f(x,T)$

$C = f(x,D,T)$

$V = f(x...T)$

$K = f(x...T)$

$\phi = f(x)$

$E = f(\phi, D. Tc)$

$Re = f(x,T...)$

$Pr = f(xT...)$

$Nu = f(xT...)$

X = X+1

x=200, 400,
400cm=1 pass

X = 2400   corresponds to 6 passes of
fuel trough core

output

COOLANT

0·3mm

12 mm

13·8 mm cell

TEMPERATURE °C

Fuel

2ms⁻¹

1000°

998°C

COOLANT

9 m s⁻¹

900°

854°C

839°C

800°

781°C

750°

0·653	—	total	2·155		Heat Transfer coefficient [wcm⁻² d
0·0216			0·0937		Viscosity
0·494			0·957		Heat Capacity [J gm⁻¹ deg
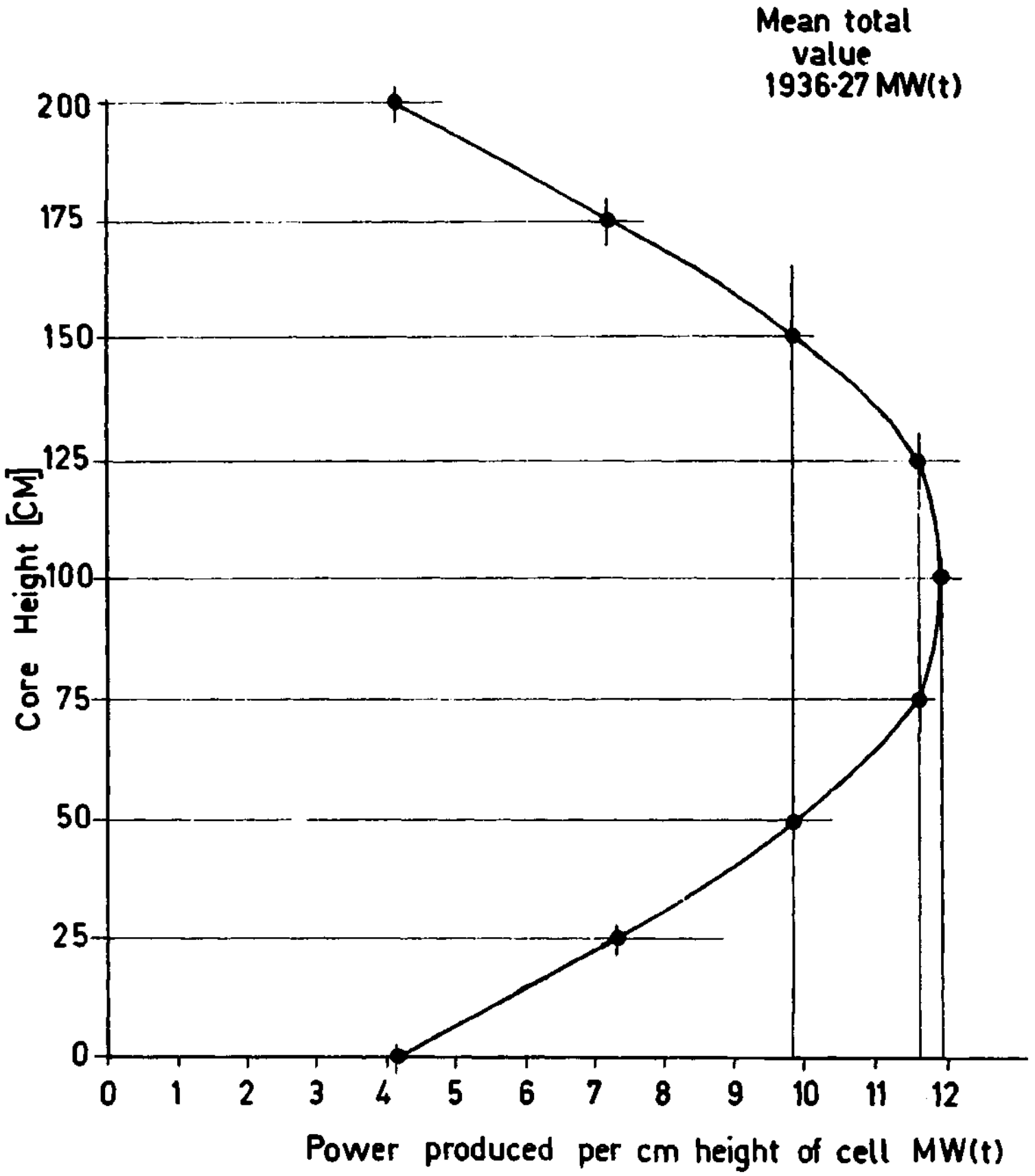
Fig 8

Power distribution in the core

P=1936MW(t)

$\overline{\Phi}$=70×10^{15}ncm^{-2}s^{-1}

Coolant 9ms^{-1}

Fuel 2ms^{-1}

CORE HEIGHT [CM]

TEMPERATURE [°C]

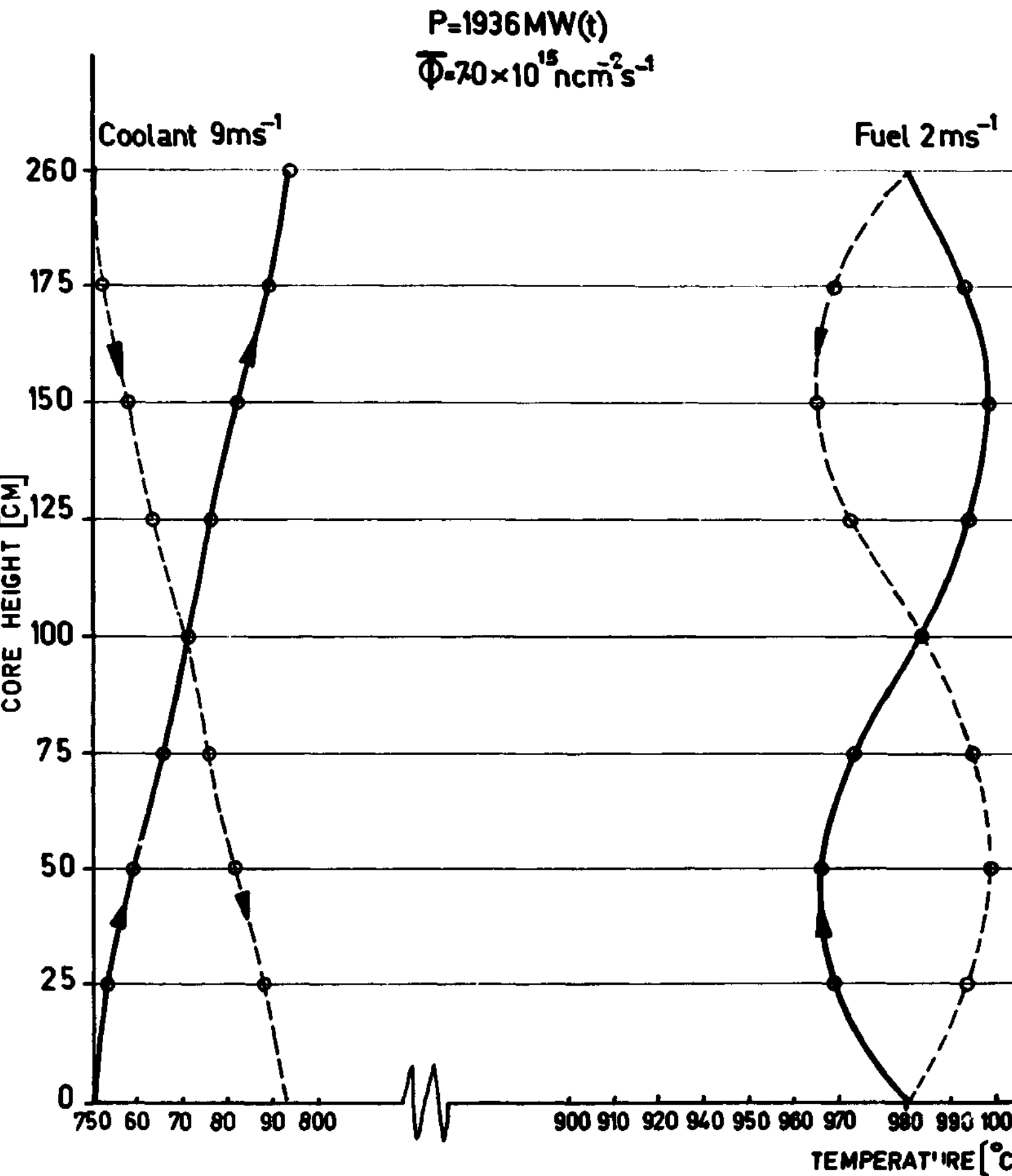Fig 9

TEMPERATURE of FUEL and COOLANT

The fuel bulk temperature changes form $980^{\circ}C$ at the bottom to $965^{\circ}C$ at 1/4 core height and is $998^{\circ}C$ at 3/4 of core height.

The coolant temperature lies between $750^{\circ}C$ inlet and $793^{\circ}C$ outlet. Both these small temperature gradients in the fuel and in the coolant (fertile and blanket 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. 10)

For a fuel velocity of 2 $m.s^{-1}$, when the coolant velocity falls from 12 $m.s^{-1}$ to 1 $m.s^{-1}$ the coolant outlet temperature increases from $784^{\circ}C$ to $893^{\circ}C$ for constant inlet temperature of $750^{\circ}C$. This change of coolant velocity and its bulk temperature results in the decrease of the mean core output from 2088 MW(t) to 598 MW(t) - that is approcimatly a factor 3! It is clear since the lower coolant velocity results in a higher coolant outlet temperature and lower power outp... we have definite negative temperature coefficient (power output) varying with the given coolant velocity.

If the fuel velocity falls from 2 $m.s^{-1}$ to 0.8 $m.s^{-1}$ we again get an important decre se of power output (see fig. 11)

The decrease in both fuel and coolant velocity results in a sharp decrease of reactor power (see fig. 12). 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.
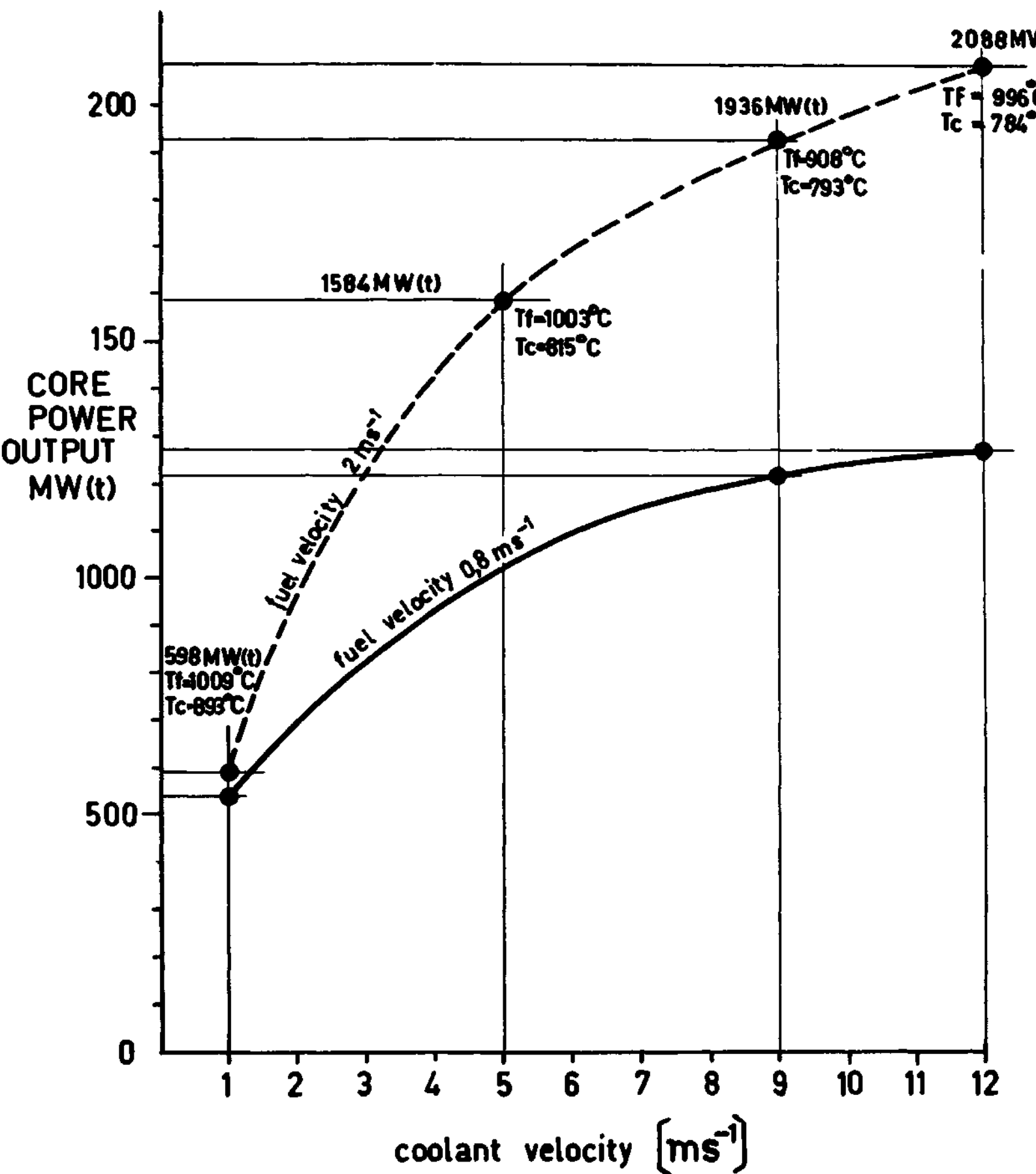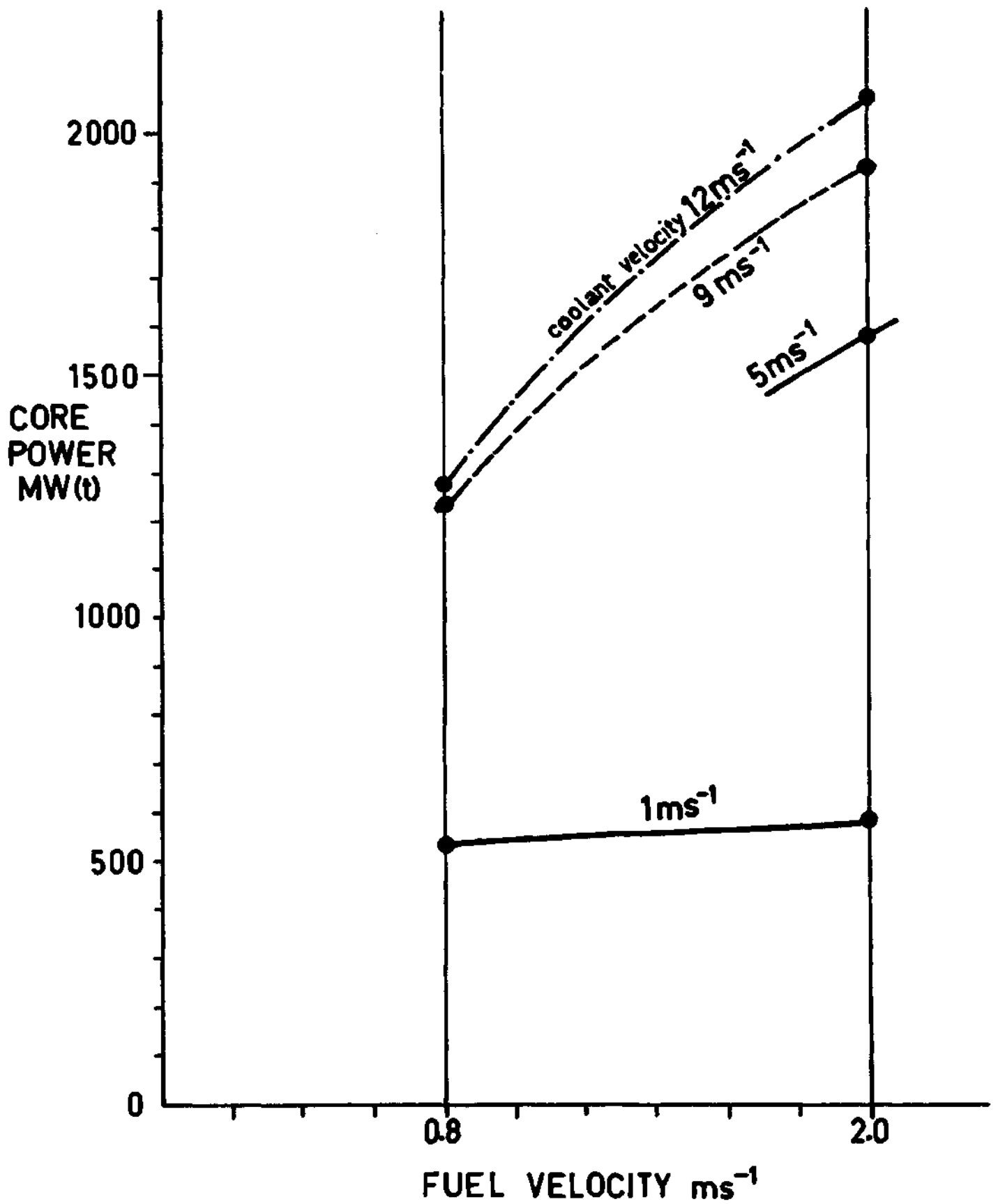
Fig 10

CORE POWER OUTPUT VERSUS COOLANT

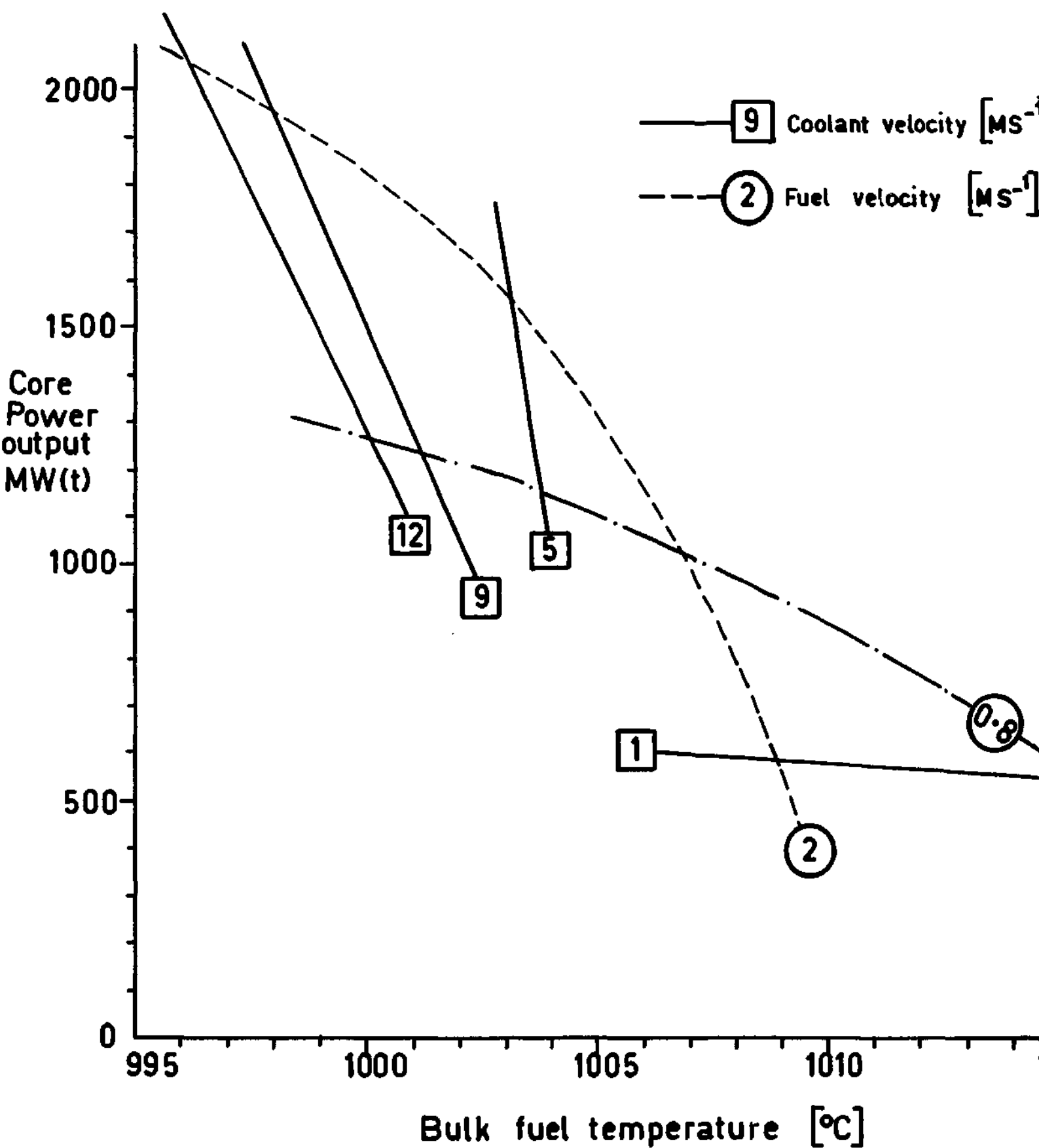**Fig 11**

CORE POWER OUTPUT VERSUS FUEL VELOCITY

Fig 12

Power output versus bulk fuel temperature

The reference core with 1936 MW(t) coolant velocity 9 m.s^{-1}, fuel velocity 2 m.s^{-1}, tube pitch/dia ratio 1.15, has a plutonium inventory of 2252 kg of Pu (Pu-239 + Pu-240) which gives a specific power of 0.86 MW(t)/kg Pu.

The breeding ratio in the core for the reference case is calculated as

$$B_{core} = \frac{N(U_{238}) \times \sigma_\gamma + N(Pu_{240}) \times \sigma_\gamma}{N(Pu_{239}) \times (\sigma_f + \sigma_\gamma)} = 0.709$$

The blanket breeding ratio for a 1 m blanket is calculated as approx 0.680 which gives a total breeding ratio 1.389.

The doubling time for this reactor is given the following definition (at 80 % load factor)

$$\text{Doubling time} = \frac{\text{Pu inventory in reactor}}{\text{Pu gain}}$$

$$= \frac{2252 \text{ kg Pu}}{1936 \text{ MW(t)} \times 1.1 \text{ kg Pu}/_{1000 \text{ MWd}} \times 300 \text{ days/year} \times (1.389-1)}$$

$$= 9.2 \text{ years}$$

It must be stressed that this doubling time is a linear one.

5. Quasi-kinetic and pumping problems

The achievment 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 forced 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 inertgas (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, mini-
mization of the auxilliary opwer, 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 postulated fuel velocity makes it possible to make some calcu-
lation on the heat transfer problems and also gives a feel for the
kinetics of the reactor under discussion.

It must be stressed that these kinetics studies have no strong
physical sense and use an iterative approach but it is clear that
they give some useful information about the general reactor stabi-
lity. (see fig. 13 and fig. 14)

There is little information on the density of $PuCl_3/NaCl$ and
$UCl_3/NaCl$ in the temperature range of interest. For these calcula-

Fuel velocity 2ms⁻¹
Coolant 9ms⁻¹

Neutron flux step change
$\phi = 16 \times 10^{16}$
$T_{fuel} = 990°C$

Reference case
$\phi = 0.8 \times 10^{16} \, n \, cm^{-2} s^{-1}$
$T_{fuel}^{o} = 990°C$

Cold fuel case
$\phi = 0.8 \times 10^{16} \, n \, cm^{-2} s^{-1}$
$T_{fuel}^{o} = 750°C$

Botton   Top   Botton   Fuel   No. of passes
          of core
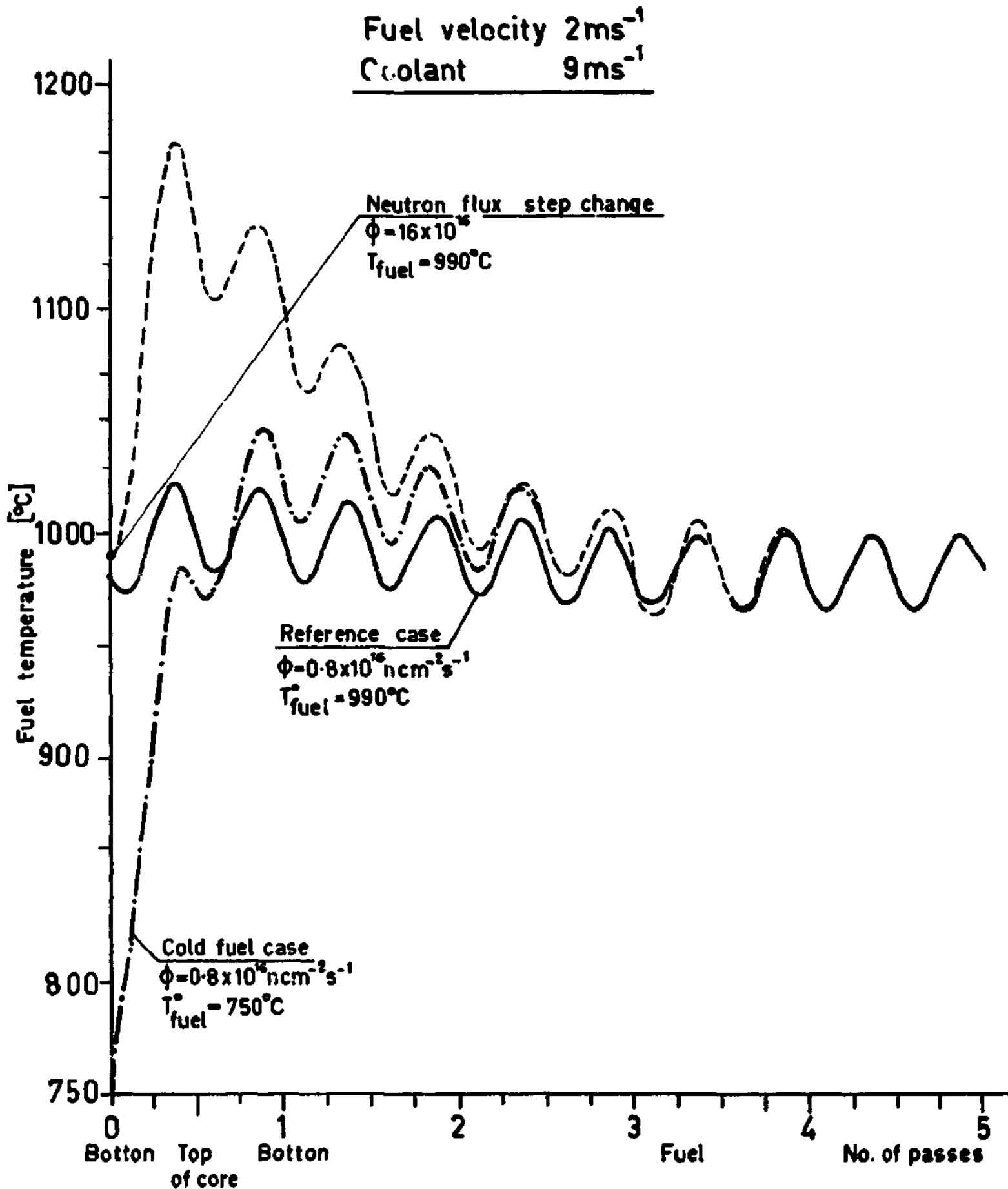
Fig 13

Fuel temperature during several passes
through core under different inital conditions

Fuel velocity $2ms^{-1}$
$T_{fuel} = 990°C$
$\phi = 0.8 \times 10^{16} n cm^{-2} s^{-1}$

Coolant velocity $1ms^{-1}$

Reference case
Coolant velocity $9ms^{-1}$

Coolant velocity $12 ms^{-1}$

No. of Passes
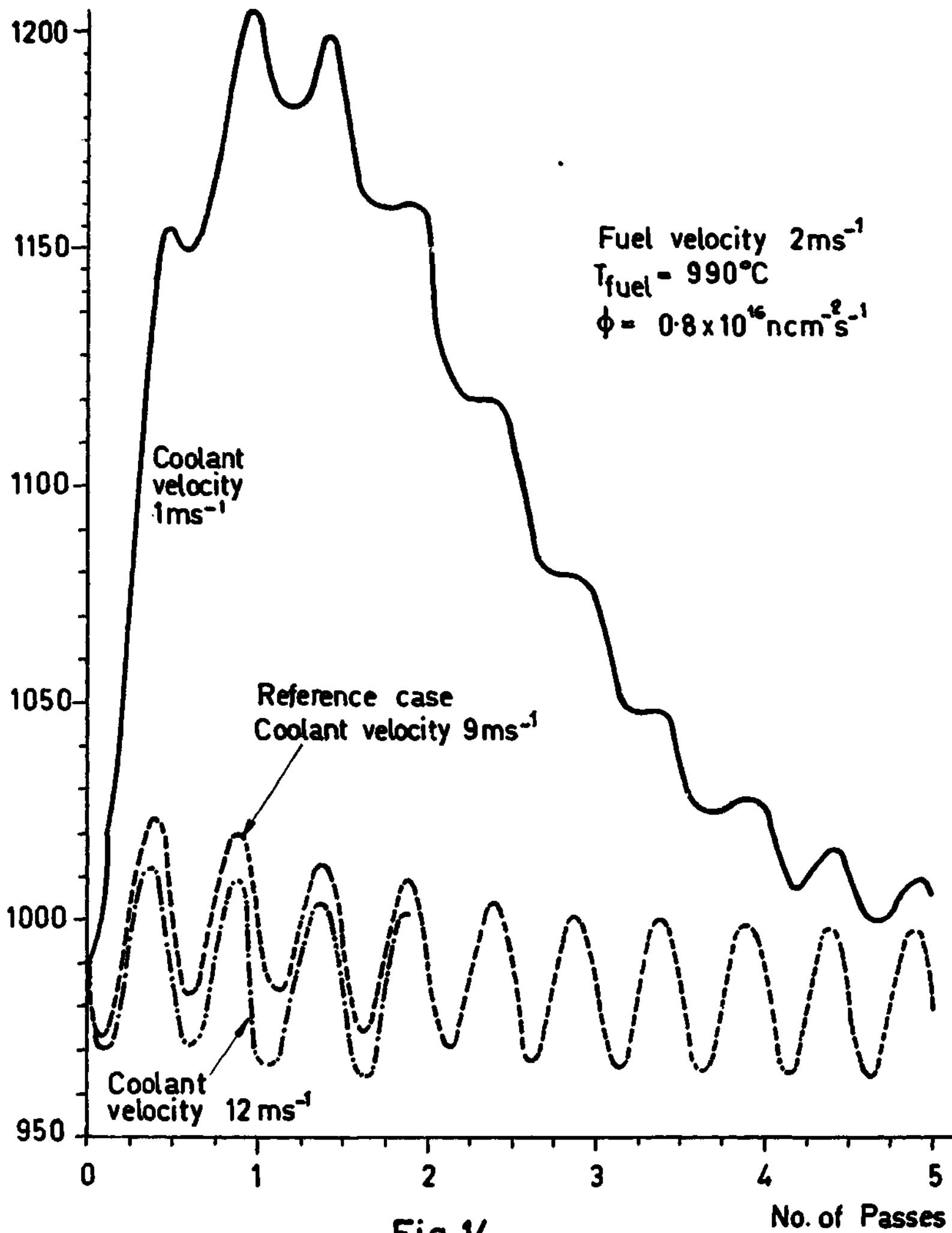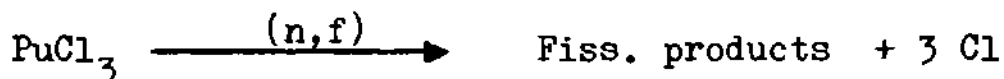
Fig 14

given in figure 15.

The influence of the temperature coefficient of density
$(\Delta\rho/\rho$ per deg) is calculated here for 3 cases, 0.5×10^{-3}
$1x \ 10^{-3}$ and 1.5×10^{-3}. Fig. 16 gives the results which show
only a small influence on reactor power.

6. Fission Products

The fission process of plutonium in a molten chloride medium may
be expressed as

$$PuCl_3 \xrightarrow{\quad (n,f) \quad} \text{Fiss. products } + 3 \ Cl$$

From the earlier published data of Taube 1965 and Chasanov 1965
and the more recent data of Lang 1970 it appears that the problem
of the possible existence of fission product oxides states was not
sufficiently discussed.

On the basis of the fission yields of Pu-239 in a fast reactor after
10 days irradiation and without cooling time the yield of the indi-
vidual elements are as shown in table 4

The most difficult problem is to predict the probable valency
(oxidation state) of some metallic elements and metalloids. In this
paper the oxidation states have been determined on the basis of the
free enthalpy of formation of the chlorides (according to Veryatin
et al. 1965) and are shown below.

TOP

200

2·32 2·33 2·34 2·35 2·36   3·98 3·99 4·00 4·01 4·02

980°C

T=793°C

998°C

Height
of
core
[cm]

Coolant

Fuel

T=769°C

T=984°C

100

2 ms⁻¹

9 ms⁻¹

Power Output
1939 MW(t)

$\overline{\Phi} \cong 0.8 \times 10^{16}$ n cm⁻² s⁻¹

150

967°C

50

25

750

BOTTOM 0

980°C

2·34 2·35 2·36   3·98 3·99 4·00 4·01 4·02

DENSITY OF SALT   [g·cm⁻³]
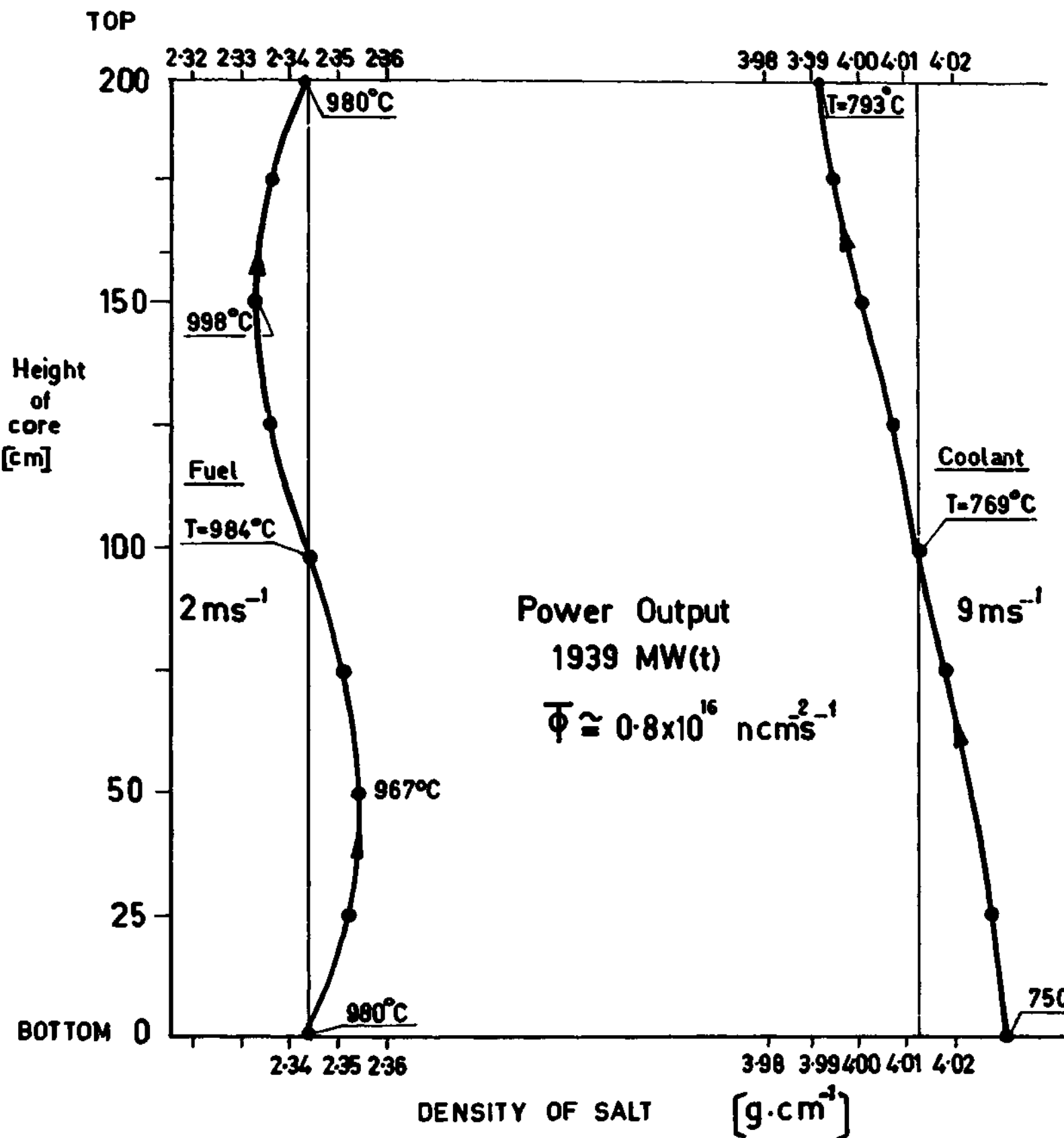
Fig 15

Density of fuel and coolant for

Reference Core

Fuel velocity 2ms^{-1}
Coolant velocity 9ms^{-1}

Fuel velocity 0·8ms^{-1}
Coolant velocity 1ms^{-1}

Power MW(t)

Temperature coefficient of density $\frac{\Delta\rho}{\rho}$ deg^{-1}
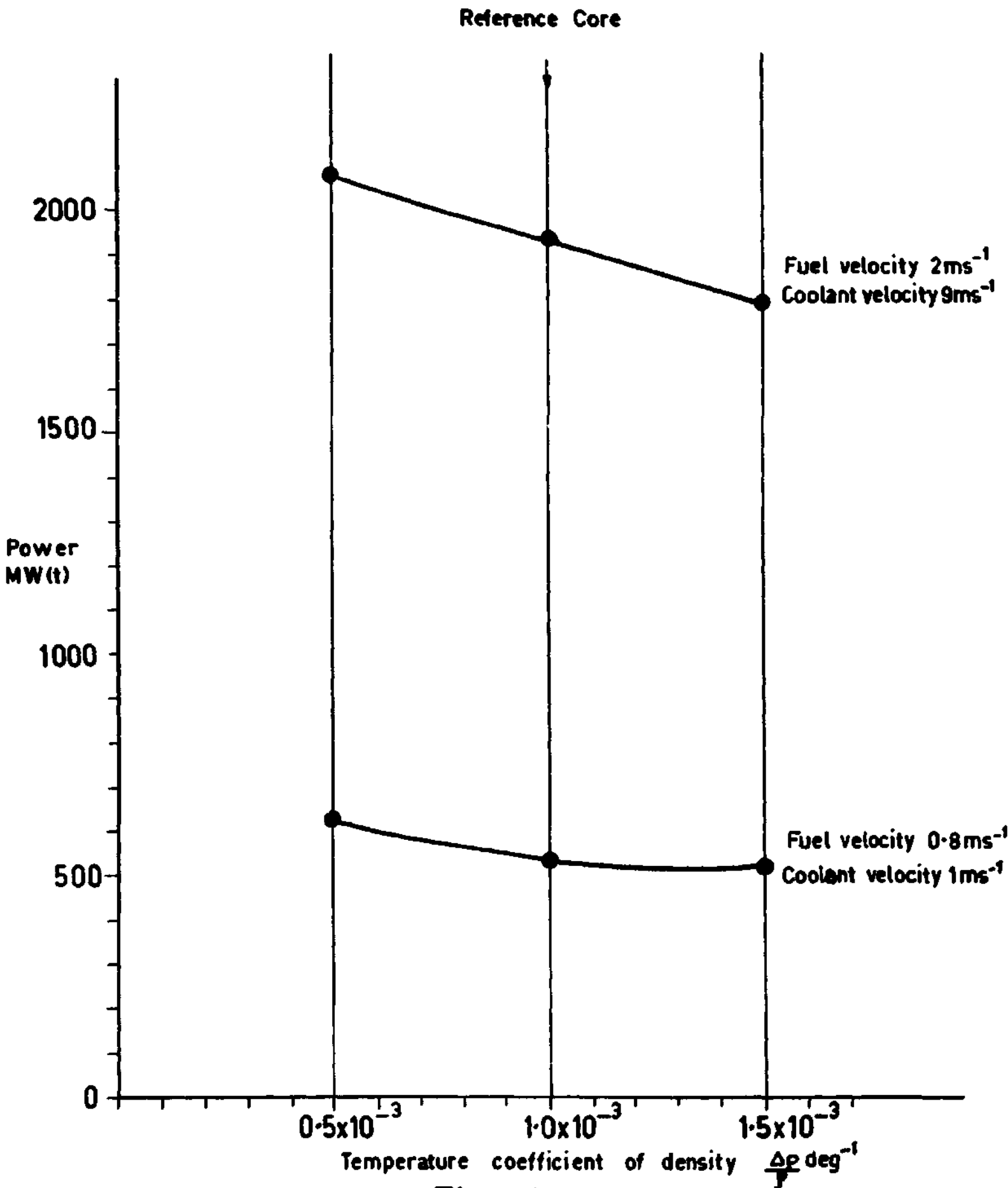
Fig 16

Impact of the density temperature coefficient
of the core power output

Table 4

Fission products in irradiated chloride fuel

Oxidation state	Fission products	Remarks
0	Kr, Xe	noble gases
0	Se, Br, I	volatile metalloids
0	Tc, Ru, Rh, Pd	semi-noble metals
0	Mo (partially 30 %)	-------
+ 1	Rb, Cs, Ag	the only stable oxidation state
+ 1	In	the most stable (but unimportant)
+ 2	Sr, Cd, Ba	the only stable oxidation state
+ 2	Sn, Mo (70 %)	the most probable oxidation state in this medium
+ 3	Y, La, Ce, Pr, Nd, Pm, Dc	the only stable state
+ 3	Sm, Eu	the most probable oxidation state
+ 3	Sb	the lowest oxidation state
+ 5	Nb	the only stable oxidation state
+ 3	Zr	the most probable state:

On the basis of this assumption the balance of chlorine is fully realised: no free chlorine is to be expected in this molten chloride system. The following elements are fully or partially in a nonoxidized state, that is in metallic form:

Tc, Ru, Rh, Pd

The total amount of these metals equals approximately 50 atoms per 100 fissioned atoms of Pu-239 (see also fig. 17).

(Note that all these considerations have been made for standard free enthalpy; but even a change in the thermodynamic activity from γ = 1 to γ = 0,001 which means a change in free enthalpy of 14 kJ$\cdot$mol^{-1}, thus appears insignificant in these rough considerations).

Table 5

Fission products in a molten chloride plutonium fuel after 10 days irradiation in a fast reactor (no cooling).

Yields per 100 atoms of Pu-239 fissioned. (Burris, 1957)

Element	Yield	Oxidation state	Chloride
Se	0,008	0	no
Br	0,003	0	no
Kr	0,942	0	no
Rb	1,050	+ 1	RbCl
Sr	5,487	+ 2	$SrCl_2$
Y	3,028	+ 3	YCl_3
Zr	21,520	+ 3	$ZrCl_3$
Nb	0,289	+ 5	$NbCl_5$
Mo	18,160	+ 2	$MoCl_2$
Tc	4,014	0	no
Ru	31,445	0	no
Rh	1,736	0	no
Pd	12,657	0	no
Ag	1,88	+ 1	AgCl
Cd	0,66	+ 2	$CdCl_2$

Element	Yield	Oxidation state	Chloride
In	0,06	+ 1	InCl
Sn	0,324	+ 2	$SnCl_3$
Sb	0,674	+ 3	$SbCl_3$
Te	7,654	+ 2	$TeCl_2$
I	6,177	0	no
Xe	21,234	0	no
Cs	13,355	+ 1	$CsCl_2$
Ba	9,502	+ 2	$BaCl_2$
La	5,79	+ 3	$LaCl_3$
Ce	13,986	+ 3	$CeCl_3$
Pr	4,278	+ 3	$PrCl_3$
Nd	11,870	+ 3	$NdCl_3$
Pm	1,44	+ 3	$PmCl_3$
Sm	3,737	+ 3	$SmCl_3$
Eu	0,595	+ 3	$EuCl_3$
Gd	0,028	+ 3	$GdCl_3$
Total	200	Mean +15	200 M $Cl_{1.5}$

Fission products in fertile material

The most important reactions in the fertile material are

fission process $\quad UCl_3 \rightarrow$ Fiss. products $+ 3$ Cl

oxidation process $\quad UCl_3 + 1/2 Cl_2 \rightarrow UCl_4; \Delta G^{1250\ °K} = 25$ kJ$\cdot$mol^{-1}

disproportionation $UCl_3 + 3\ UCl_3 \rightarrow 3\ UCl_4 +$ Umet $\quad$ (see also: Harder, 1970)
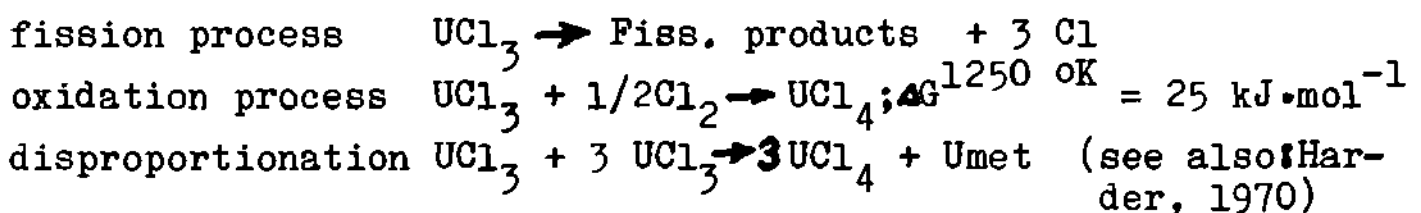
Because some of the fission product chlorides have a free enthalpy
of formation of the same order of magnitude as the oxidation
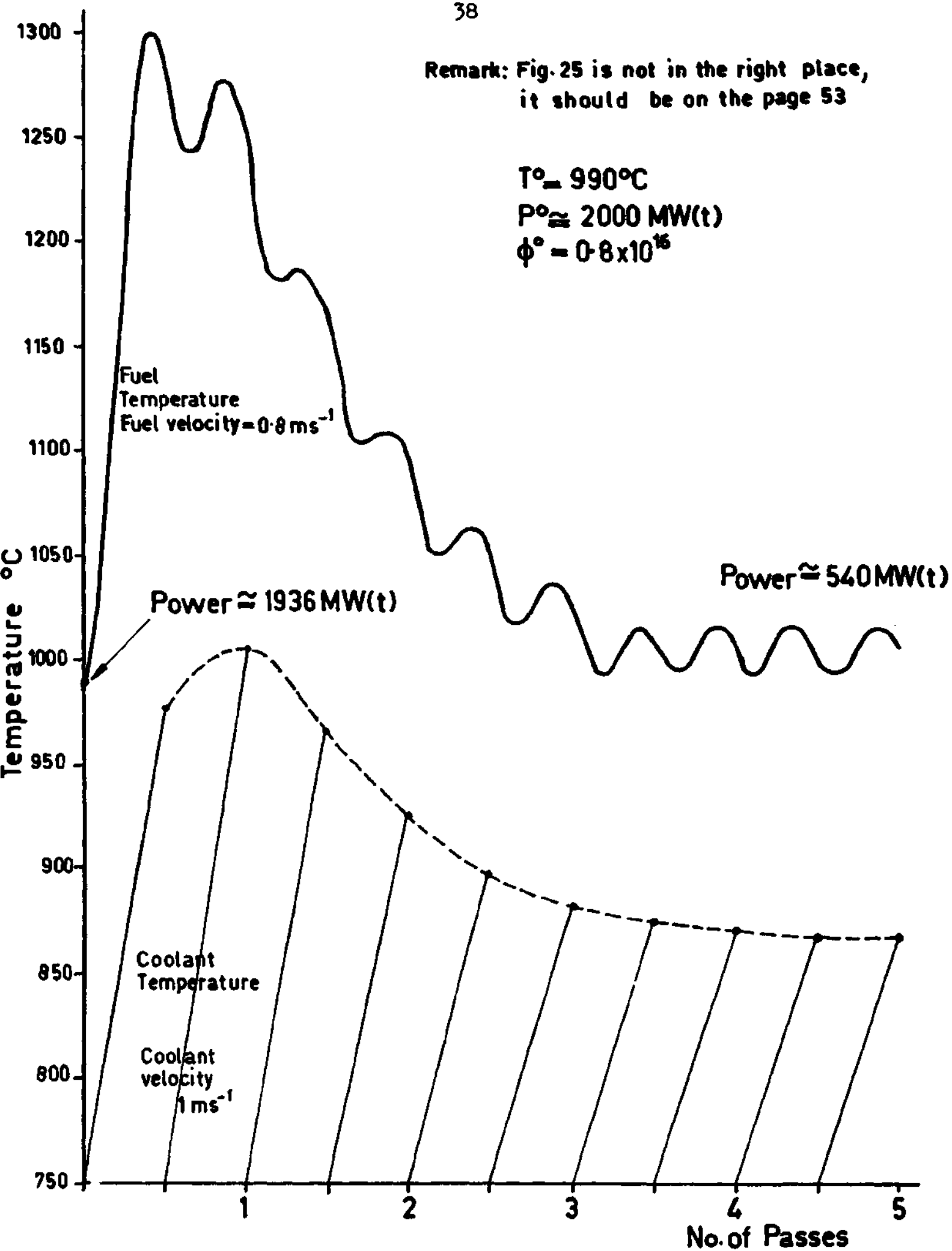
Selfdamping mechanisms of the reactor
following step reductions of coolant and

process of $UCl_3 \longrightarrow UCl_4$, a reaction of the following type is possible

$$UCl_{3(s)} + 1/2\ MoCl_{2(s)} \longrightarrow UCl_{4(s)} + 1/2\ Mo_{(m)}; \Delta G^{1000\ ^{O}K} = 0$$
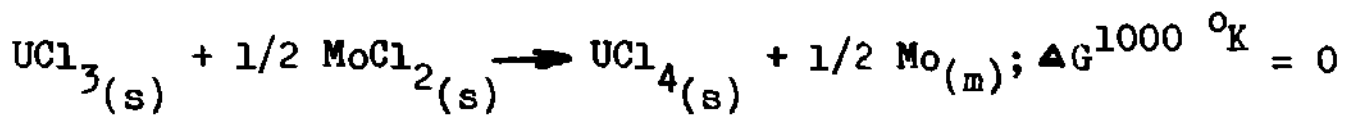
The corrosion aggressivity of UCl_4 is of course similar to that of $MoCl_2$.

7. Rep: 'cessing

Breeder reactors as we already know 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.

The separation of plutonium and/or uranium from the irradiated fuel by means of pyrochemical techniques could be carried out, for example, in the following way

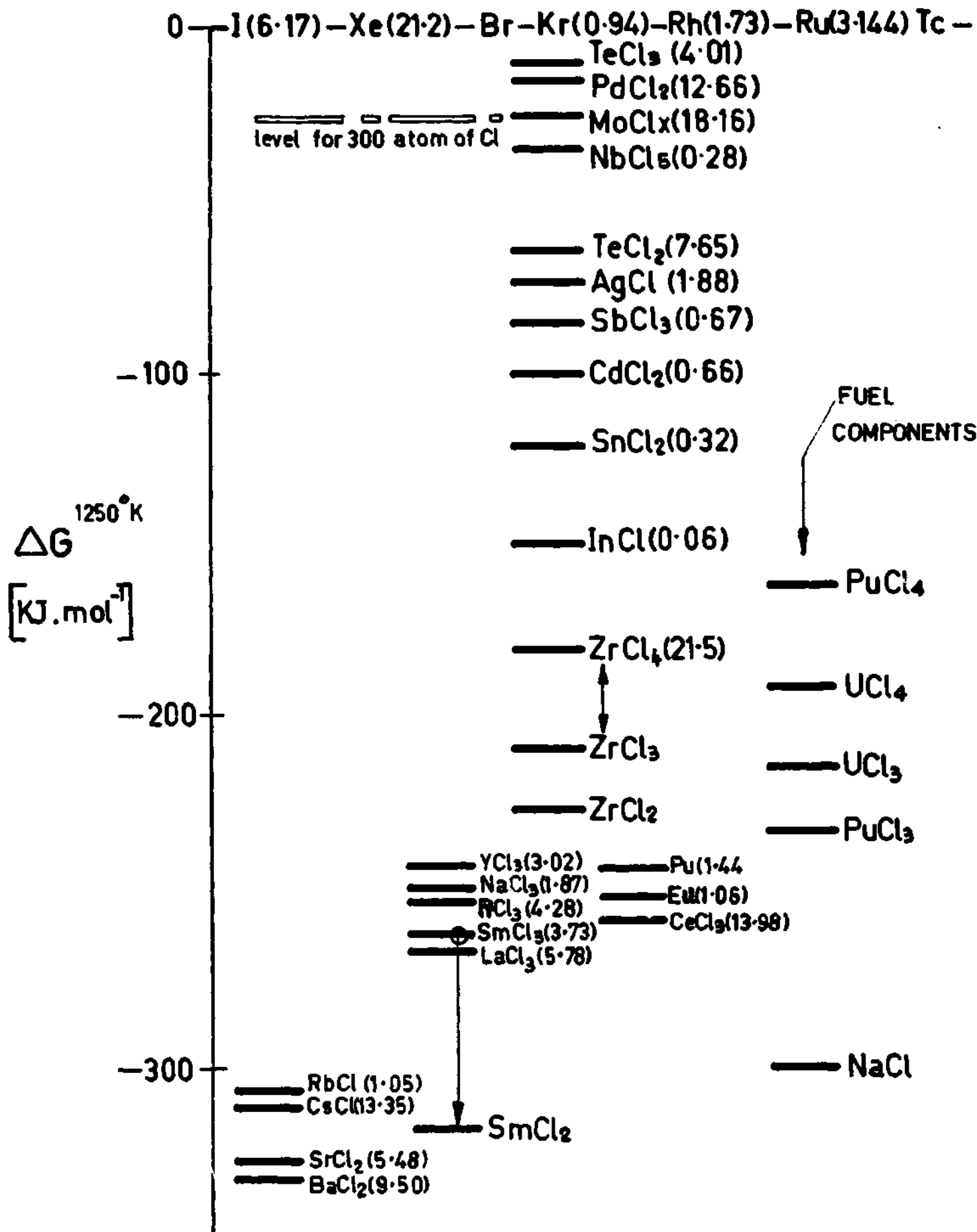Molten salt, primary phase Pu, FP (part of FP remains)
            Transport of Pu and part of FP.

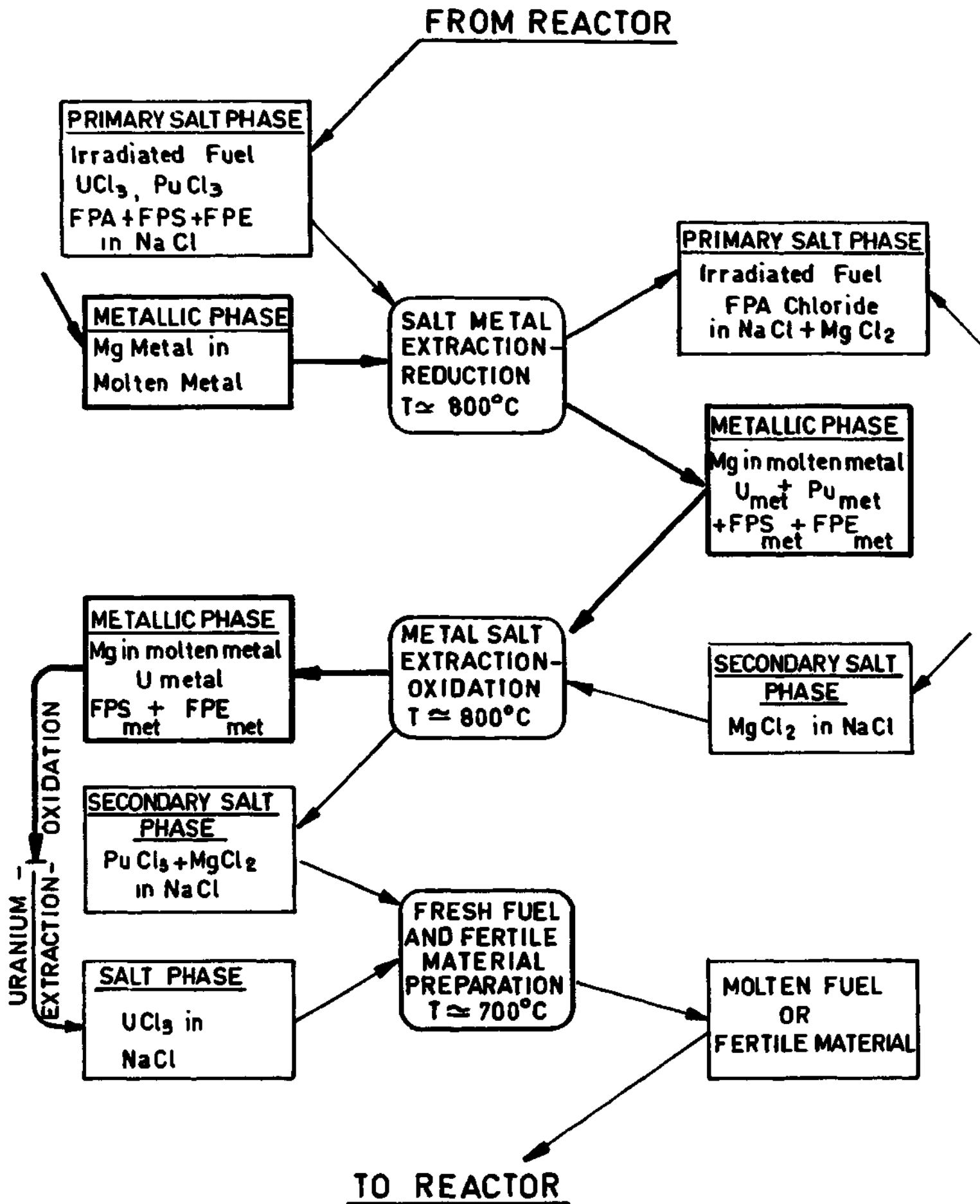Metallic phase (part of FP remains)
            Transport of Pu

Molten salt, secondary phase containing only Pu.

This is the so called 'metal transport' process (fig. 18)

Free Energy of formation for fission product
chlorides [Yields shown in brackets]

Fig. 17

FROM REACTOR

PRIMARY SALT PHASE
Irradiated Fuel
UCl_3, $PuCl_3$
FPA + FPS + FPE
in $NaCl$

METALLIC PHASE
Mg Metal in
Molten Metal

SALT METAL EXTRACTION-REDUCTION
$T \simeq 800°C$

PRIMARY SALT PHASE
Irradiated Fuel
FPA Chloride
in $NaCl + MgCl_2$

METALLIC PHASE
Mg in molten metal
$U_{met} + Pu_{met}$
+ FPS_{met} + FPE_{met}

METALLIC PHASE
Mg in molten metal
U metal
FPS_{met} + FPE_{met}

METAL SALT EXTRACTION-OXIDATION
$T \simeq 800°C$

SECONDARY SALT PHASE
$MgCl_2$ in $NaCl$

URANIUM – EXTRACTION– OXIDATION

SECONDARY SALT PHASE
$PuCl_3 + MgCl_2$
in $NaCl$

SALT PHASE
UCl_3 in
$NaCl$

FRESH FUEL AND FERTILE MATERIAL PREPARATION
$T \simeq 700°C$

MOLTEN FUEL OR FERTILE MATERIAL

TO REACTOR

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.

From fig. 19 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 formulations smaller than those of $PuCl_3$.

FPE = fission products existing in elementary form because of small free enthalpy of chloride formation or negative balance of chlorine.

The proposed schema of the separation processes utilizing metal transport is given in fig. 20
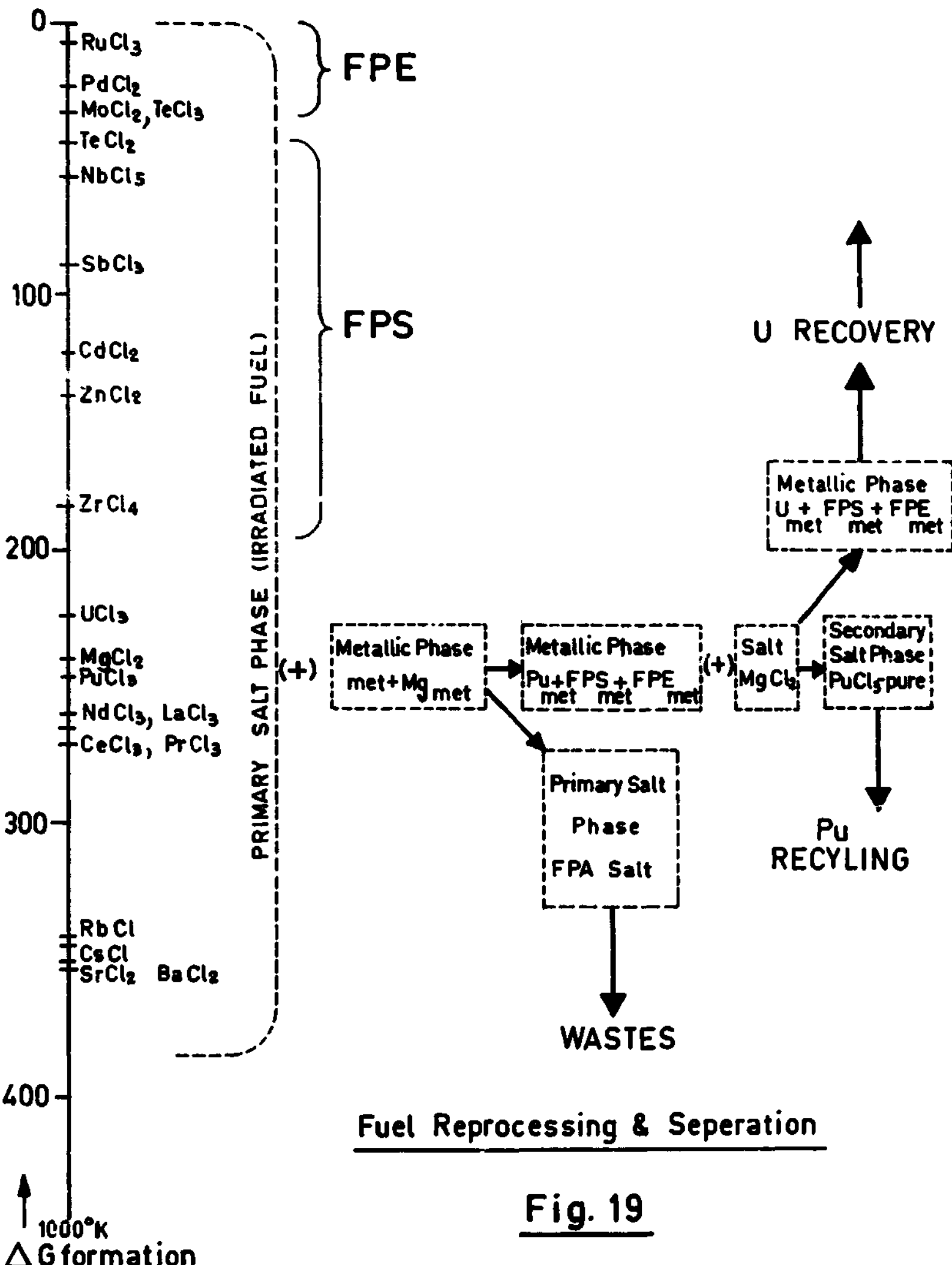
8. Aluminium trichloride as turbine working agent

One of the most important features of the proposed reactor is the relatively high total thermal efficiency of the power station.

Such a high thermal efficiency is possible only under two conditions:
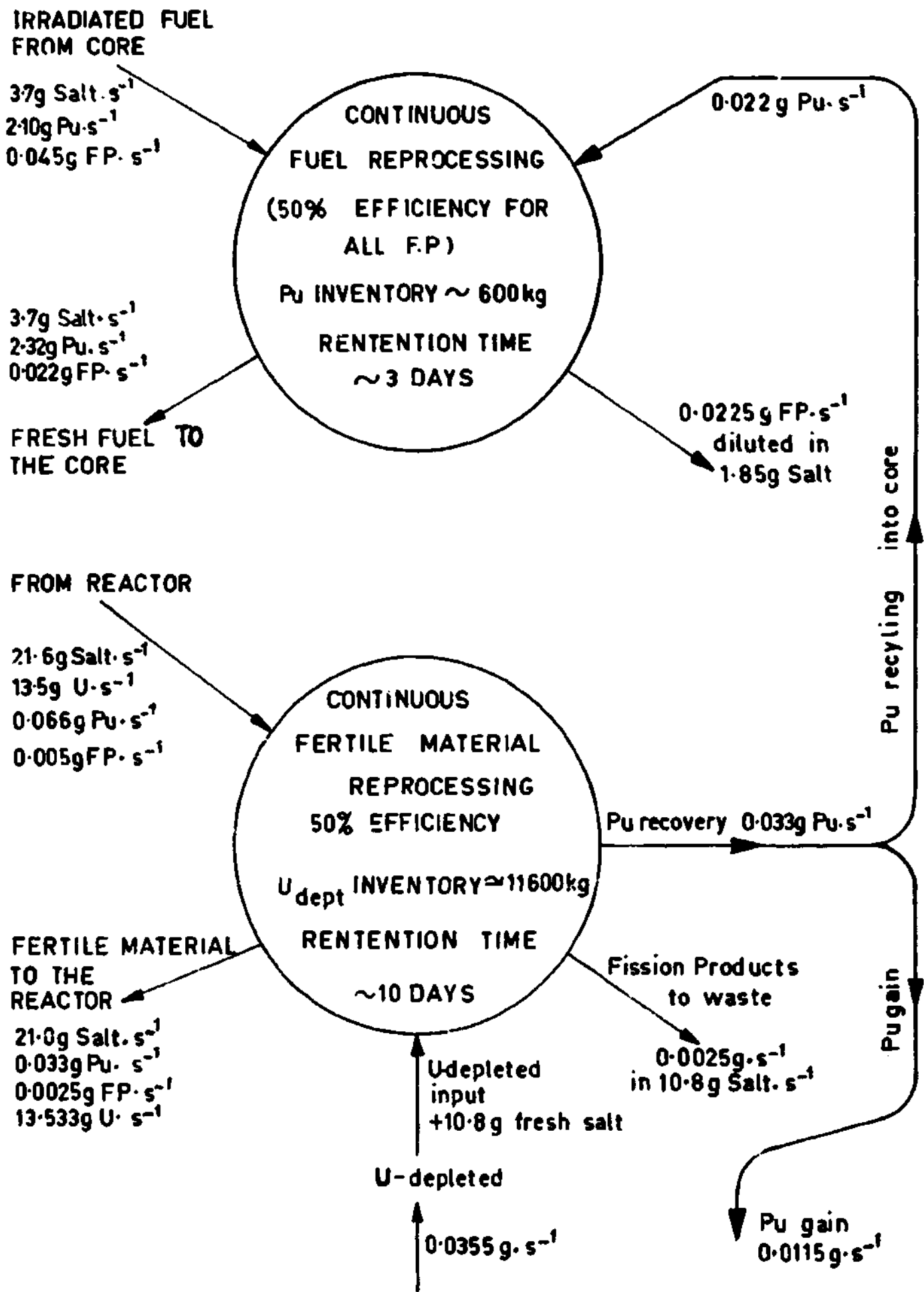
- working agent at a higher temperature

- certain required thermodynamic properties of the working agent must be met.

Among several possibilities- aluminium trichloride, a rather exotic working agent is proposed here.

43

Fuel Reprocessing & Seperation

Fig. 19

IRRADIATED FUEL
FROM CORE

3.7g Salt·s⁻¹ → $3.7g\ Salt\cdot s^{-1}$
2.10g Pu·s⁻¹ → $2.10g\ Pu\cdot s^{-1}$
0.045g F.P.·s⁻¹ → $0.045g\ F.P.\cdot s^{-1}$

0.022 g Pu·s⁻¹ → $0.022\ g\ Pu\cdot s^{-1}$

CONTINUOUS
FUEL REPROCESSING
(50% EFFICIENCY FOR
ALL F.P)
Pu INVENTORY ~ 600 kg
RENTENTION TIME
~ 3 DAYS

3.7g Salt·s⁻¹ → $3.7g\ Salt\cdot s^{-1}$
2.32g Pu·s⁻¹ → $2.32g\ Pu\cdot s^{-1}$
0.022g FP·s⁻¹ → $0.022g\ FP\cdot s^{-1}$

FRESH FUEL TO
THE CORE

0.0225 g FP·s⁻¹ → $0.0225\ g\ FP\cdot s^{-1}$
diluted in
1.85g Salt

Pu recyling into core

FROM REACTOR

21.6g Salt·s⁻¹ → $21.6g\ Salt\cdot s^{-1}$
13.5g U·s⁻¹ → $13.5g\ U\cdot s^{-1}$
0.066g Pu·s⁻¹ → $0.066g\ Pu\cdot s^{-1}$
0.005g FP·s⁻¹ → $0.005g\ FP\cdot s^{-1}$

CONTINUOUS
FERTILE MATERIAL
REPROCESSING
50% EFFICIENCY
U_{dept} INVENTORY ~ 11600 kg
RENTENTION TIME
~ 10 DAYS

Pu recovery 0.033g Pu·s⁻¹ → $0.033g\ Pu\cdot s^{-1}$

FERTILE MATERIAL
TO THE
REACTOR

21.0g Salt·s⁻¹ → $21.0g\ Salt\cdot s^{-1}$
0.033g Pu·s⁻¹ → $0.033g\ Pu\cdot s^{-1}$
0.0025g FP·s⁻¹ → $0.0025g\ FP\cdot s^{-1}$
13.533g U·s⁻¹ → $13.533g\ U\cdot s^{-1}$

Fission Products
to waste

0.0025g·s⁻¹ → $0.0025g\cdot s^{-1}$
in 10.8g Salt·s⁻¹ → $10.8g\ Salt\cdot s^{-1}$

Pu gain

U-depleted
input
+10.8g fresh salt

U-depleted

0.0355 g·s⁻¹ → $0.0355\ g\cdot s^{-1}$
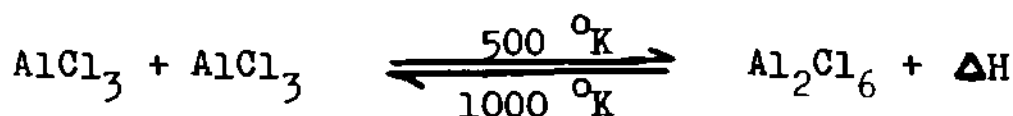
Pu gain
0.0115 g·s⁻¹ → $0.0115\ g\cdot s^{-1}$

FUEL REPROCESSING MATERIAL BALANCE

Fig 20

The most important property of this substance is its spontaneous dimerisation at lower temperatures

$$AlCl_3 + AlCl_3 \xrightleftharpoons[1000\ ^{o}K]{500\ ^{o}K} Al_2Cl_6 + \Delta H$$

$\Delta H = 125$ kJ/mol

This reaction results in a two fold decrease in volume but also release of some amount of heat energy.

The physico-chemical properties of aluminium trichloride are very well known (see Blander 1957, Krasin 1967). The phase diagram is given in fig. 21.

Table 6

Physical and chemical properties of dissociating-gas systems Al_2Cl_6 and N_2O_4

|	Al_2Cl_6	N_2O_4
Molecular weight (g·mol^{-1})	266,7	92,02
Normal boiling point (^{o}C)	193	21,5
Critical temperature (^{o}C)	352,7	158,3
Critical pressure (bar)	26	1033
Melting point (^{o}C)	195	- 11
	(at 2,46 bar)	
Heat of evaporation (kJ·kg^{-1})	150	415
Heat of dimerisation (kJ·kg^{-1})	470	620
Type of reaction	Al_2Cl_6 $\Updownarrow$ $2AlCl_3$	N_2O_4 $\Updownarrow$ $2\,NO_2$ $\Updownarrow$ $2\,NO + O_2$

5. Corrosion

From the point of view of corrosion the following regions can be distinguished, see Table 7.

|	Region F (fuel in core)	Region B (Blanket and coolant)	Region A (outside core)
Components	NaCl	NaCl	-----
	$PuCl_3$	$PuCl_3$ (little)	-----
	no UCl_3	UCl_3	-----
	FP	FP (little)	-----
	-----	-----	$AlCl_3$
Temperature gradient	$965-998^{\circ}C = 33^{\circ}$	$750-793^{\circ}C = 73^{\circ}$	$750-400^{\circ}C = 350^{\circ}$
Neutron dose	very high	high	none
Gamma dose	very high	high	very small
Velocity of medium	$2\ m.s^{-1}$	$9\ m.s^{-1}$	$40\ m.s^{-1}$
State	liquid	liquid	gaseous/liquid

The selected structural material is molybdenum.

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

$$Mo_{(m)} + \frac{2}{X} MeCl_{X(s)} \longrightarrow MoCl_{2(s)} + \frac{2}{X} Me_{(m)}$$
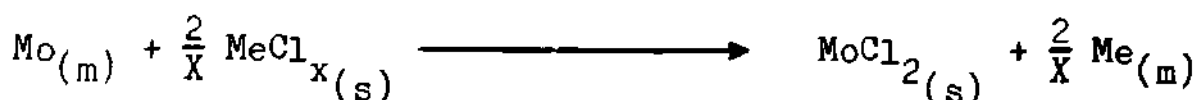
For the reaction in region F of fresh fuel $PuCl_3$ in NaCl, the most likely reaction is (1250 $^{\circ}K$)

$$Mo_{(m)} + 2/3\ PuCl_{3(s)} \longrightarrow MoCl_2 + 2/3\ Pu_{(m)}$$

$$\Delta G^{1000\ ^{\circ}K} = +\ 450\ kJ/mol\ Cl$$
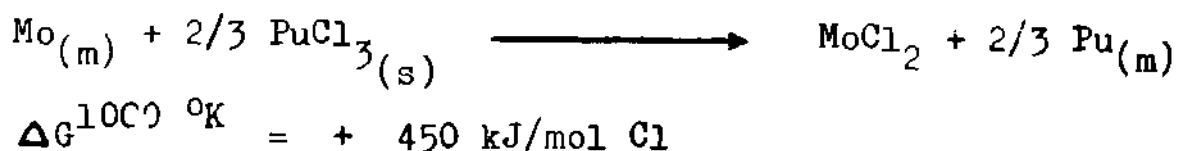
The equilibrium constant of this reaction is so small and equals 10^{-17} that this reaction has no practical meaning.

In region B the most dangerous reaction is connected with uranium tetrachloride, the product of the oxidation of uranium trichlorides (chlorine from fission of $PuCl_3$):

$$UCl_{3(s)} + 1/2\ Cl_{2(s)} \longrightarrow UCl_{4(s)}$$

$$2\ UCl_{4(s)} + Mo_{(m)} \longrightarrow 2\ UCl_{3(s)} + MoCl_{2(s)}$$
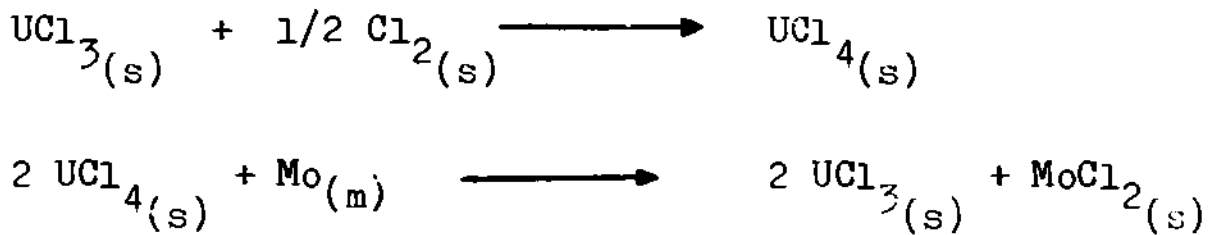
The control of the UCl_3/UCl_4 ratio in the fertile-coolant material might be feasible due to the continuous reprocessing of this material together with the control of zirconium from the fission products oxidation state. In the region A that is in the external heat exchanger the main corrosion process results from the action of gaseous aluminium chloride (the secondary working agent). (see fig. 22)

$$AlCl_{3(g)} + \frac{1}{X}\ Mo_{(m)} \longrightarrow \frac{1}{X}\ MoCl_{2x(g)} + AlCl_{(g)}$$

This reaction was discussed in previous publications (Blander 1957) but unfortunately not all the thermodynamic data is known. Molybdenum forms four compounds with chlorine: $MoCl_2$, $MoCl_3$, $MoCl_4$, $MoCl_5$ (fig. 22). The stability of these chlorines is strongly influenced by the concentration of free chlorine and also by the temperature.

A more detailed calculation of metallic molybdenum corrosion in the aluminium trichloride is needed, see fig. A7. These calculations are very sensitive to the vapour pressure of chlorides (fig. 23). In connection with corrosion problems mention must also

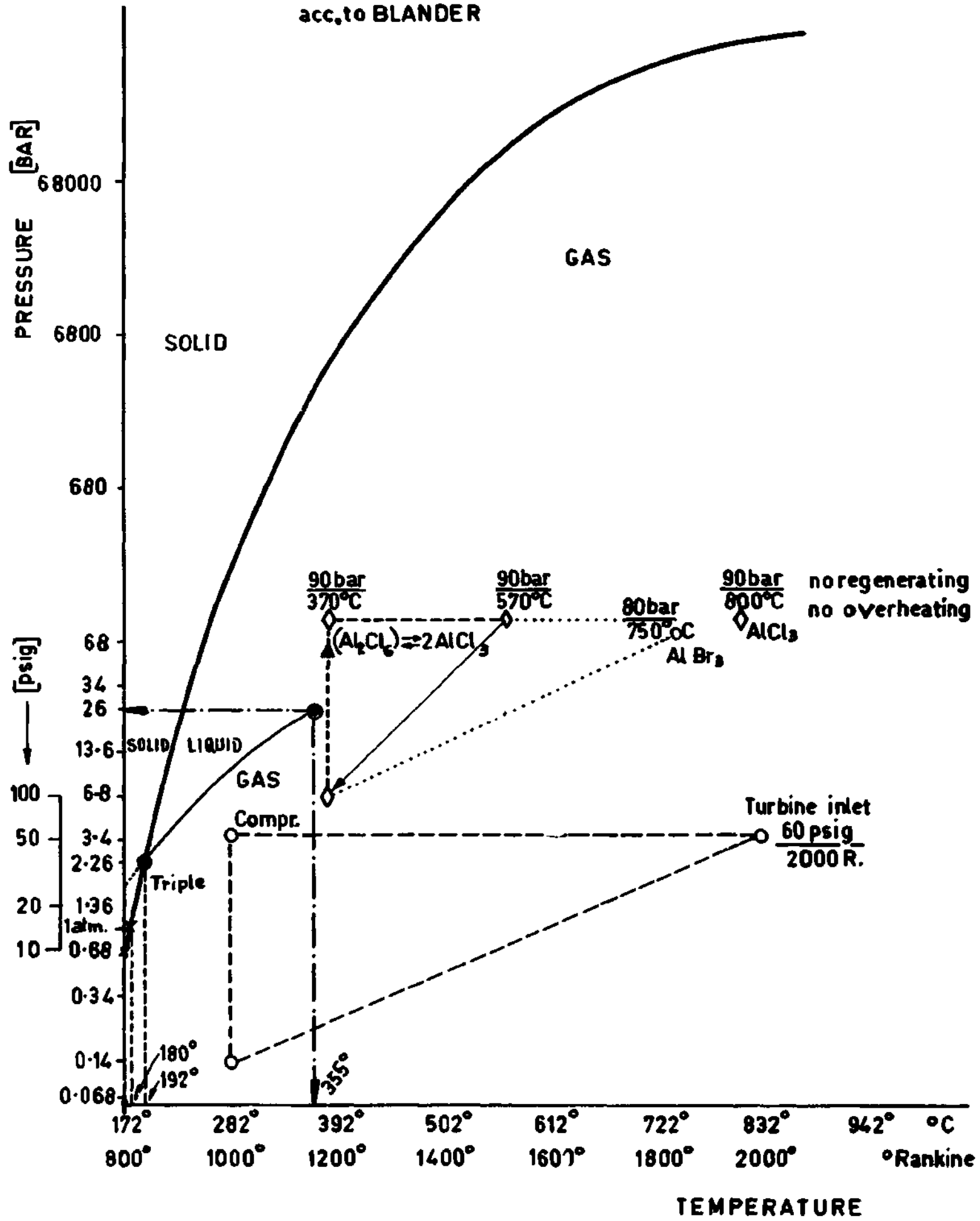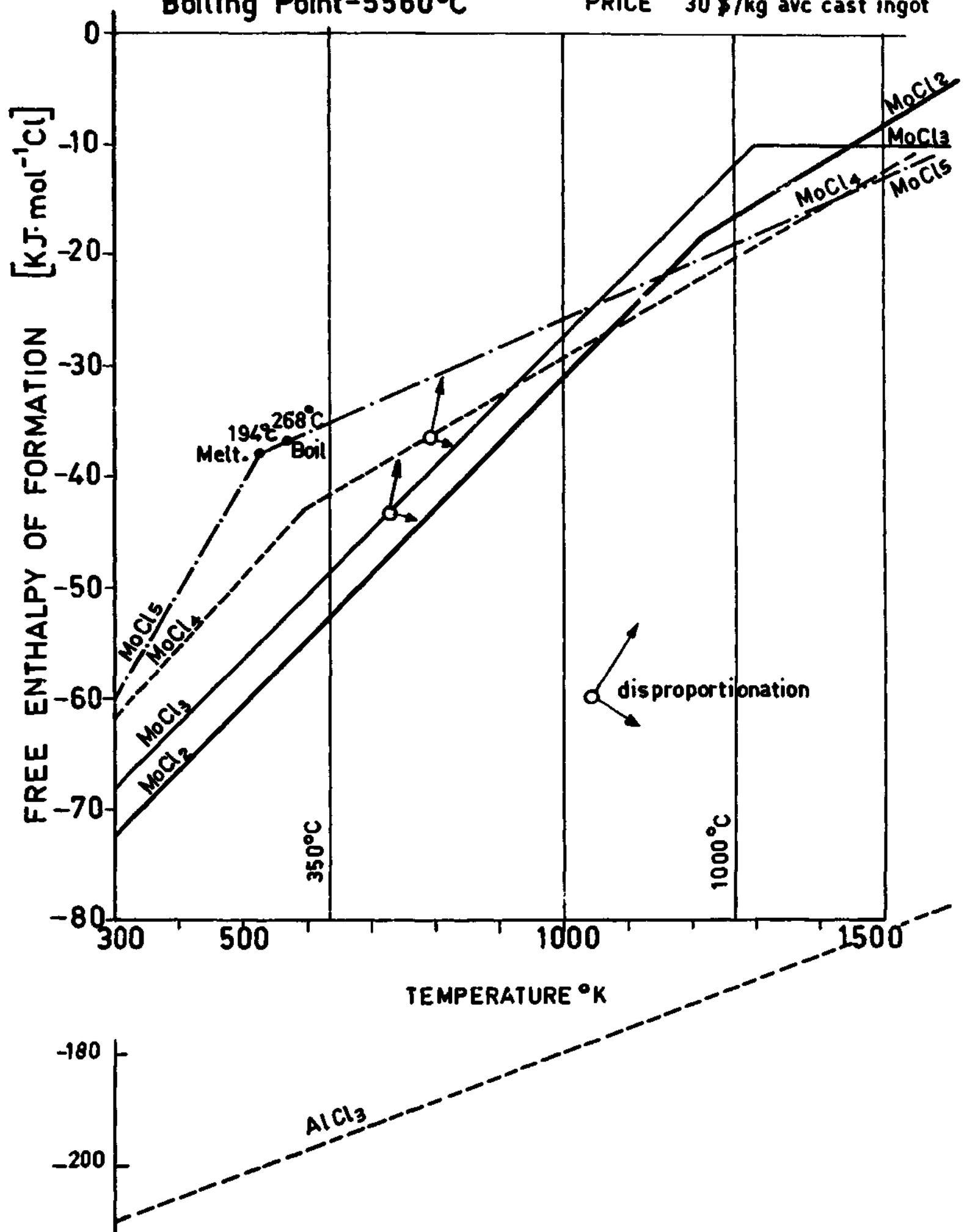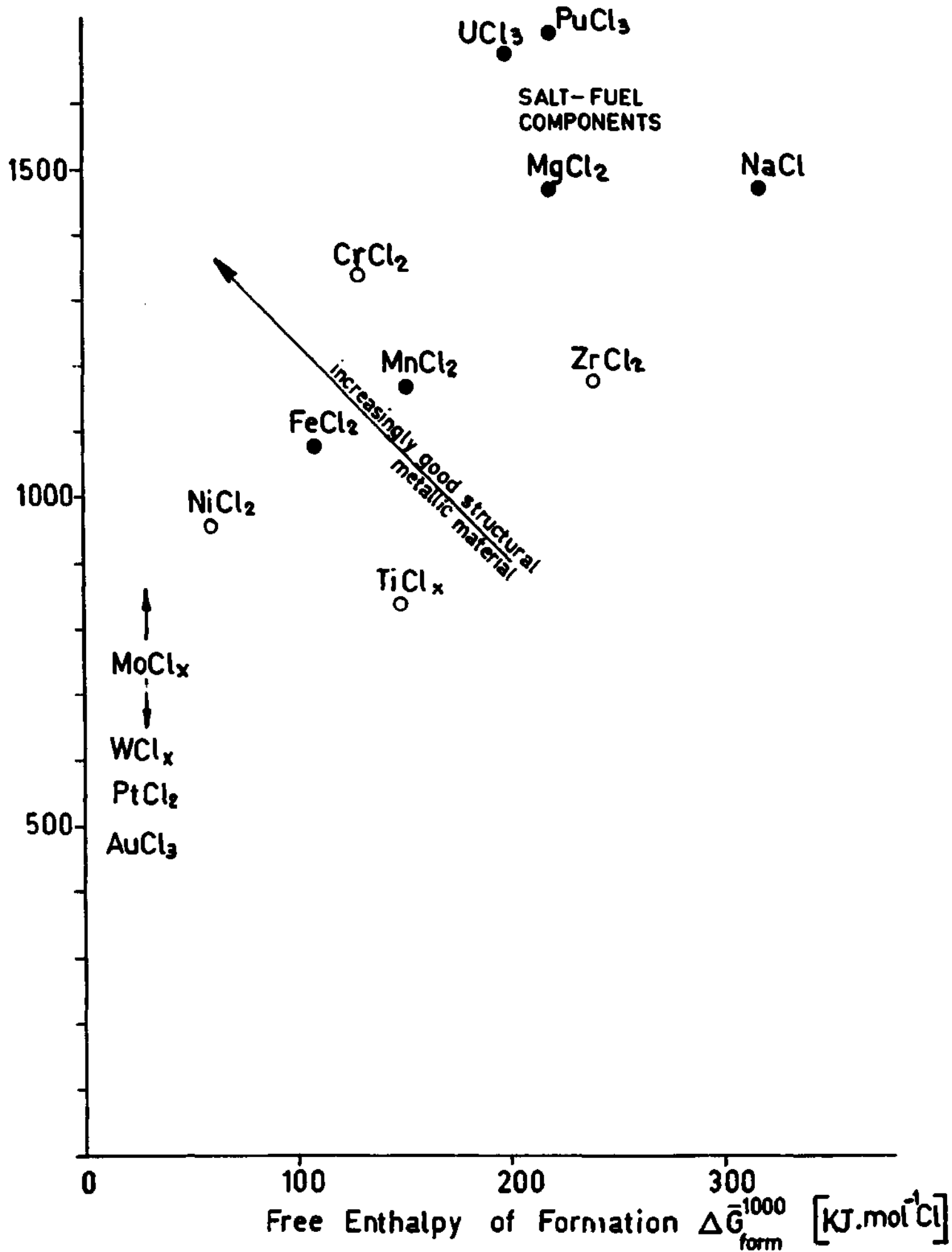$$lg P_{(atm)} = -\frac{6360}{T} + 3.77T - 0.00612T + 6.78$$

acc. to BLANDER

PRESSURE [BAR]

68000

GAS

6800

SOLID

680

68

34

26

13·6 — SOLID   LIQUID

[psig]

100

50

20

10

6·8

3·4

2·26

1·36

1 atm.

0·68

0·34

0·14

0·068

90 bar / 370°C

(Al₂Cl₆) ⇌ 2 AlCl₃

90 bar / 570°C

80 bar / 750°C

Al Br₃

90 bar / 800°C

AlCl₃

no regenerating
no overheating

GAS

Compr.

Triple

180°
192°

355°

Turbine inlet
60 psig
2000 R.

172°   282°   392°   502°   612°   722°   832°   942°   °C
800°   1000°   1200°   1400°   1600°   1800°   2000°   °Rankine

TEMPERATURE

Molybdenum – Metal
Melting Point – 2610°C
Boiling Point – 5560°C

$\rho = 10.22\ g \cdot cm^{-3}$

PRICE   30 $/kg avc cast ingot

FREE ENTHALPY OF FORMATION $[kJ \cdot mol^{-1}\ Cl]$

MoCl2
MoCl3
MoCl4
MoCl5

194°C 268°C
Melt.   Boil

disproportionation

MoCl5
MoCl4
MoCl3
MoCl2

350°C

1000°C

TEMPERATURE °K

AlCl3

Free Enthalpy of Formation $\Delta\bar{G}_{form}^{1000}$ $[KJ.mol^{-1}Cl]$

CHLORIDES – BOILING POINT   V

be made of the problem of the reaction between metal chlorides and oxygen and water.

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

$$MeCl_2 + H_2O \longrightarrow MeO + 2HCl$$

$$MeCl_2 + O_2 \longrightarrow MeO + Cl_2$$
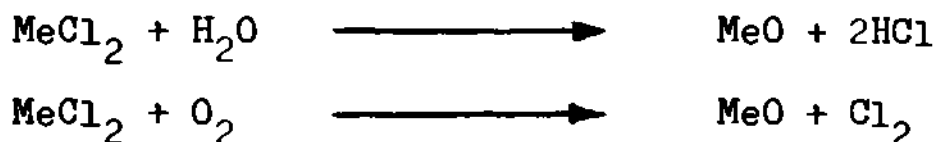
The metal oxides are mostly insoluble in molten chlorides, which results in a serious disturbance of the fuel system. From this point of view the metallic elements could be divided into three classes: (see fig. 24)

- those which are stable against H_2O and O_2, that is the chlorides are more stable than the oxides (eg. Na, Cs, Ba) and partially Ca.

- those which are not stable against H_2O and O_2 and the resulting product is a mixture of chloride, oxychloride and oxide (eg. 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 (eg. Mo, W).
  Metals of this class seem to be not so numerous as in the other two classes.

This property causes the rapid elimination of traces of water or oxygen in the molten salts of Pu and U chlorides. It is also well known that traces of H_2O and O_2 have a very big influence on corrosion rate.
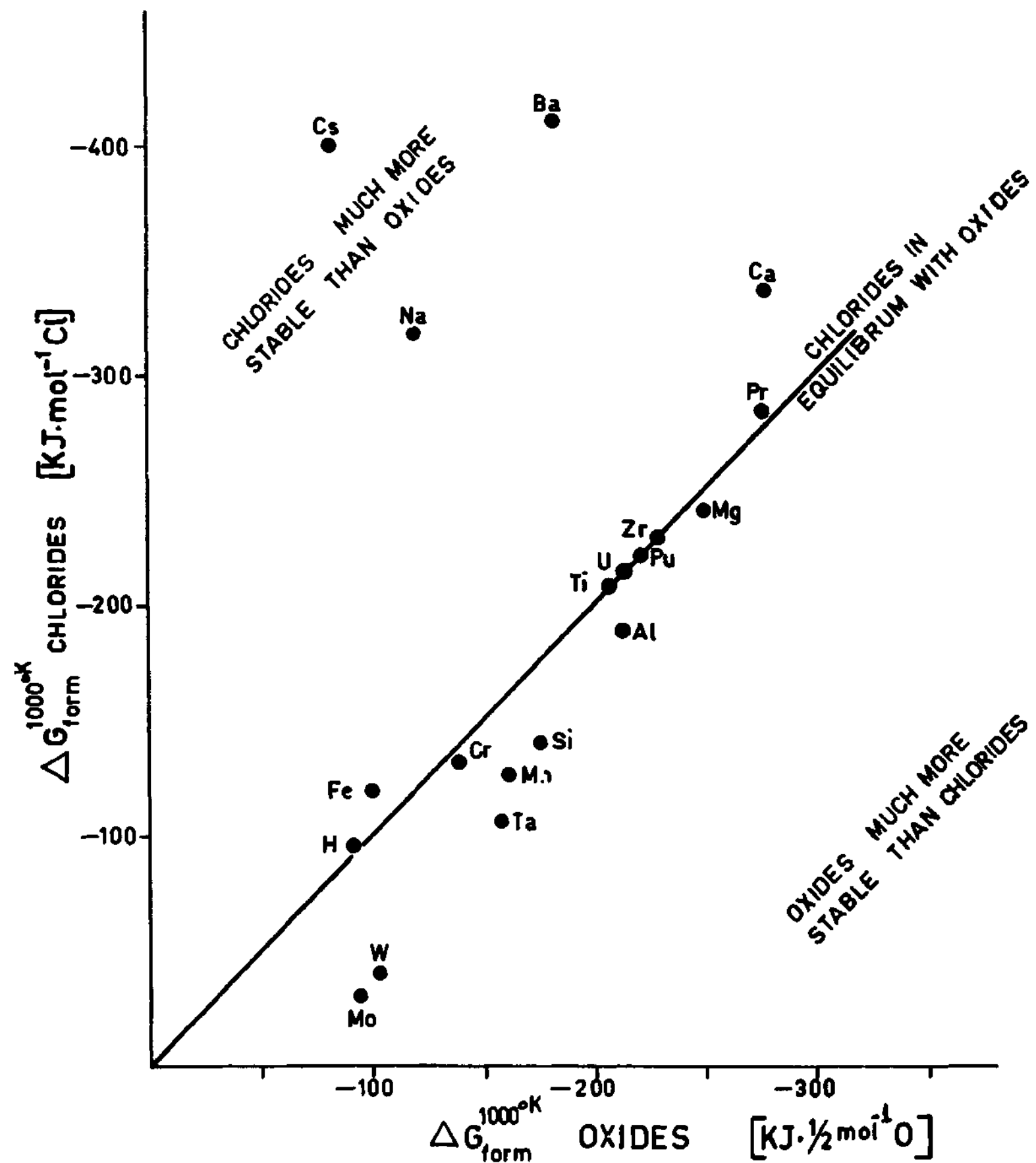
Fig 24 <u>CHLORIDES—OXIDES EQUILIBRUM DIAGRAM</u>
at 1000°K

The same property may be advantageous in establishing a thin coating of oxide on molybdenum surfaces. This suggestion must be proved thermodynamically and experimentally.

It must be stressed that the problem of removal of oxygen and water and other oxygen containing substances from the salts may be crucial for the corrosion problem as well as for long term fuel stability.

10. Safety Problems

The molten chlorides reactor seems to be a relatively safe system because of the following reasons

- an extremely high negative temperature coefficient of reactivity, because 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. (fig. 25) <u>Fig. 25 is on the page 38.</u>

- 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 leak 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 rather adverse property of such a molten fuel reactor is the necessity of initially heating the solidified fuel in a non critical geometry with external power. (eg. from the electrical grid).

11. Economics

It is not possible, when considering the econimics of this type of
reactor, at such an early stage to make realistic statements of
costs and predictions of economic performance for a station in
full operation. Here we merely indicate the main areas in which
this type of reactor can be expected to have an economic advantage
including some comments on the possible attractions offered by a
molten fuel reactor in utilizing the abundant but low grade sources
of uranium which may become available when economic or national
requirements dictate the need.

The most important advantage of the molten chloride breeder power
system (power reactor including reprocessing plant) is of course
due the part it could play in reducing the costs of power produc-
tion. The possible eccnomic advantages are caused by the following
features

- in relation to the 'classical' solid fuel fast breeder reactor,
  the molten chlorides fast breeder reactor system removes the
  need for the following operations: cooling of the irradiated
  fuel, transport, decladding, liquefaction, manufacture of solid
  fuel, cladding, fuel pin assembly, transport etc.

- the amount of fuel outside the core is, in the molten chloride
  reactor only a few percent of the fuel core inventory. In the
  solid fuel reactor the out of core amount equals the fuel core
  inventory. The capital costs for the out of core fuel are of
  significance.

- the doubling time for these reactors _s shorter than those for
  sodium cooled solid fuel reactors and, being equal to 7 years,
  gives a good doubling time.

- the mean burn up in the molten fuel, continuously reprocessed
  could be a factor 3-5 lower than that for the solid fuel system.

- in a 'closed' system of power station and reprocessing plant the safeguards are much simpler and easier to apply.

In addition the molten chlorides reactor has further advantages:

- a higher mass specific power ($MW(t)/kgPu$) than the solid fuel reactor which decreases the fuel inventory capital costs.

- a high power density ($MW(t)/m^3$ of core) than the solid fuel reactor, which may decrease the capital costs of core, blanket and shielding, perhaps in the future an increasingly important part of power production costs.

- more attractive, from the point of view of future conditions when the costs of uranium recovery will probably increase and when independance from a foreign market may become an important factor.

This last point is here developed further as of being of particular relevance to the Swiss economy and national interests but which may become more and more relevant to the world uranium market in the future.

As with the classical fast breeder reactor, the molten chlorides reactor can be used for 'burning the rocks' (according to A.M. Weinberg), that is for utilization of the dispersed uranium present in granites in amounts of the order of 10 ppm (the mean value of uranium concentration in the entire earths crust is 4 ppm).

The continuous reprocessing of irradiated fuel and the relatively simple preparation of fresh fuel and fresh fertile material as suggested for the molten chlorides reactor seems to be ideally suited for 'rocks burning'.

Let as make some simple calculations:

1 m^3 of granite equals 2500 kg of minerals, and with a uranium content of 10 ppm we have 25 g of uranium per 1 m^3 of granite.

As is known in the Swiss Alps in Piz-Giuv (Aar-Massif) the uranium content in the syenite equals 15 - 30 ppm (Prof. Hügi 1971) so in this case for 30 ppm.

1 m^3 of syenite weighs: 2.5×10^3 kg

2.5×10^3 kg $\times 30 \times 10^{-6} = 75 \times 10^{-3}$ kg = 75 g Uranium.

In breeding

    75 g U-239 $\longrightarrow$ 70 g Pu-239

    for power production

    70 g Pu $\times$ 1 MWd $\times 10^3$ kw $\times 24 \times 3.6 \times 10^3$ s $= 6 \times 10^9$ KJ

in other words

1 m^3 of syenite is equivalent to 6×10^9 KJ = 6 TJ or 6×10^{12}

                                                                                  Joules.

It is interesting to compare this energy source with oil:

1 m^3 of petroleum product, with a specific weight og 900 kg/m^3 and heat of combustion $\sim$ 12000 kCal/kg gives:

900 kg $\times 12 \times 10^3 \times 4.18 = 4.5 \times 10^7$ kJ $= 4.5 \times 10^{10}$ Joules

that is 6/0.045 = 130 times less than the energy content of 1 m^3 of granite (syenite).

In terms of volumes, 1 m^3 of petroleum product is equivalent to 7 dm^3 of granite (syenite) (or a cube 19 cm x 19 xm x 19 cm) and in addition 1 m^3 of oil requires approx. 3000 kg of oxygen or 10'000 m^3 air for combustion.

In the future output of the Swiss nuclear power industry will reach 10'000 MW(e).

For this amount of electrical energy approx. 10'000 kg Pu must be burnt annually. In a steady state nuclear power industry this amount of plutonium could be obtained from 20'000 kg of natural uranium (allowing for losses etc.). So for the Swiss nuclear industry it would be sufficient to recover the uranium from approximately 1 million cubic meters of granite annually - in other words the volume of a tunnel 30 m x 10 m x 3000 m or an open mine 200 x 200 x 20 m.

The cost of this rocks burning may be estimated as follows

The present price of natural uranium is 20 $/kg U for the case where uranium is recovered from 0.1 % uranium ores.

In the future, when uranium is recovered from granites with only 0.001 % (10 ppm) uranium the price of uranium may increase to 500 $/kg that is 25 times.

Let use assume that the price of plutonium (currently and in the future) equals 8000 $/kg.

At the present time the proportion of uranium raw material costs appearing in this plutonium price is only

$$\frac{20 \times 2}{8000} = 0,5 \% \qquad (2 \text{ kg U for 1 kg Pu})$$

In the future for the very expensive uranium from granites this part will equal

$$\frac{500 \times 2}{800} = 12.5 \ \%$$

Or in other words an increase in the cost of recovery of natural uranium by a factor of 25 will only raise the plutonium price from 8000\$/kg to 9000\$/kg.

The present cost of electrical energy with an optimistic figure of 4 mills/kWh(e) gives 1,1 mills for Pu per kWh(e) which at the higher price would give energy (electrical) costs of 4,27 mills/kWh(e) that is only 6,7 % more expensive than the current price.

Thus to summarize:

- The molten chlorides reactor is an attractive candidate for utilization of low grade ores such as from granite.

- Even with the greatly increased cost of recovery of uranium from granite, the influence on electrical energy costs is small.

- From a resource point of view, the energy content (per m^3) of granite is greatly superior to say fuel oil (130 times).

-- The use of the abundant supplies of granite is abviously an attractive possibility from the point of view of the economy and interests of countries such as Switzerland.

12. Some other selected breeders, thermal and fast

For sake illustration a critical review concerning four reactors has been made.

Type of reactor		Fuel	
		liquid	solid
		(molten salt)	(oxide)
	thermal	molten fluorides breeder (Oak Ridge)	------
	fast	molten chloride breeder (Argonne)	plutonium oxide breeder (Argonne)
		molten chloride breeder (EIR) (this paper)	

The main parameters of these reactors are given in the Table 8

Table 8 I

Some selected breeder reactors

Reactor type	Thermal reactor		Fas
Fuel	Molten fluoride	Solid oxide	Molten p
Laboratory	Oak Ridge N.L.	Argonne N.L.	Argonne Na
Reference year	⊥971	1970	(A)
Cooling system	external to core	in the core	external to core	i
Coolant medium	fuel	sodium	fuel	s
Blanket material	Th-232 fluoride in the fuel	U-238 oxide in the fuel	U-238 chloride in the fuel	U- r f U- b
Core volume (m^3)	55,5	5,8	10	1
External fuel holding (m^3)			15	
Specific power $\frac{MW(e)}{Kg\ fiss}$	0.66	0.26	0.132	
Specific power $\frac{MW(e)}{m^3}$	18	170		2
Fuel composition mol%	3% $^{233}UF_3$ in fluoride	12% PuO_2 in UO_2	30% $(Pu_xU_{1-x}Cl_3)$	5 (
Fuel max. temp. (^{o}C)	704	2800		6
Cladding material	graphite	steel	HastelloyF	H
Fuel density $\frac{kg}{m^3}$	3280 (at 704°)	11000	3000	3
Coeff expan $\frac{1}{^{o}C}$	6.7 x 10^{-4}			3

Table 8 / II

Reactors	Thermal Molten Fluoride	Fast oxide	Homogen (A)
Specific heat $\left(\frac{J}{g \cdot deg}\right)$ of fuel	1.357		0.84
Fuel velocity $\left(\frac{m}{s}\right)$	2.6 (fuel)	solid fuel	high
Coolant velocity $\left(\frac{m}{s}\right)$	no coolant	8.2 (sodium)	no coolant
Neutron flux $\left(\frac{n}{cm^2 s}\right)$	2.6×10^{14}	10^{16}	
Fissile material in entire system (kg)	3000	3860	7500
Fertile material	Th fluoride	U-238 oxide	U-238 chloride
Breeding ratio (total)	1.06	1.27	1.48
Doubling time (years)	22	13	9 (estimated)

Appendix

A1. Physical and chemical properties of salt components

The search for fuel, fertile material and for coolant is limited by following criteria:

- thermodynamic and kinetic stability of plutonium and uranium compounds

- melting point not higher than $700^{\circ}C$, in pure state or in dissolved state (fig. A1, A2)

- boiling point not lower than $1500-1600^{\circ}C$ both for pure and dissolved states (low vapour pressure) (fig. A3)

- high solubity, at least 15 mol % of plutonium and 70 mol % uranium compounds

- small neutron capture cross-section for fast neutrons

- small elastic scattering for fast neutrons

- small inelastic scattering

- good heat-transport properties; specific heat capacity, (low viscosity, high conductivity etc.) (fig. A4)

- good corrosion properties (if possible) (fig. A5)

- sufficient technological or laboratory experience

- relatively low price.

All these manifold criteria are sufficient well fullfilled by following compunds:
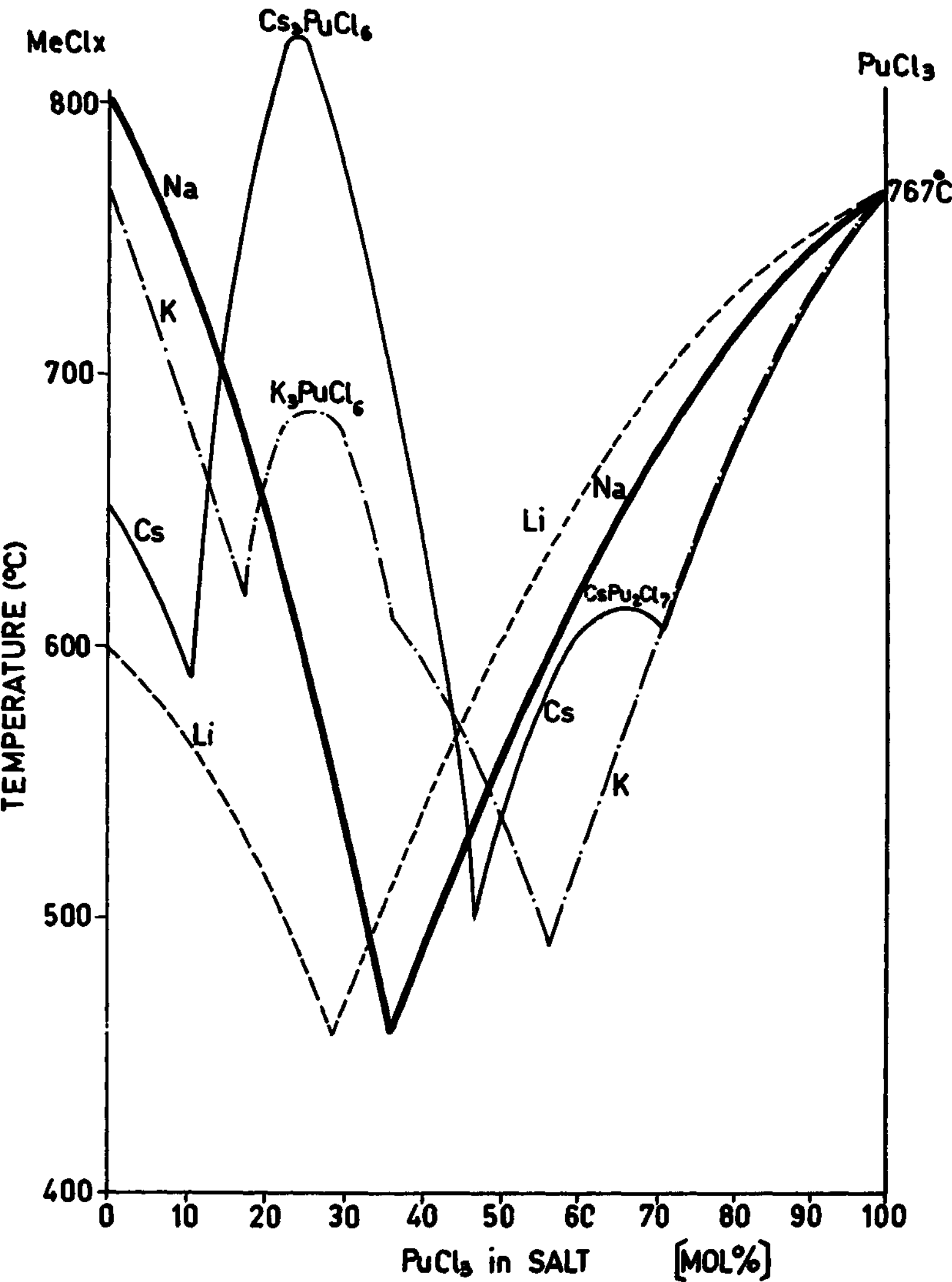
$PuCl_3$, UCl_3, NaCl and as coolant $AlCl_3$.

Table I

Properties of fuel components

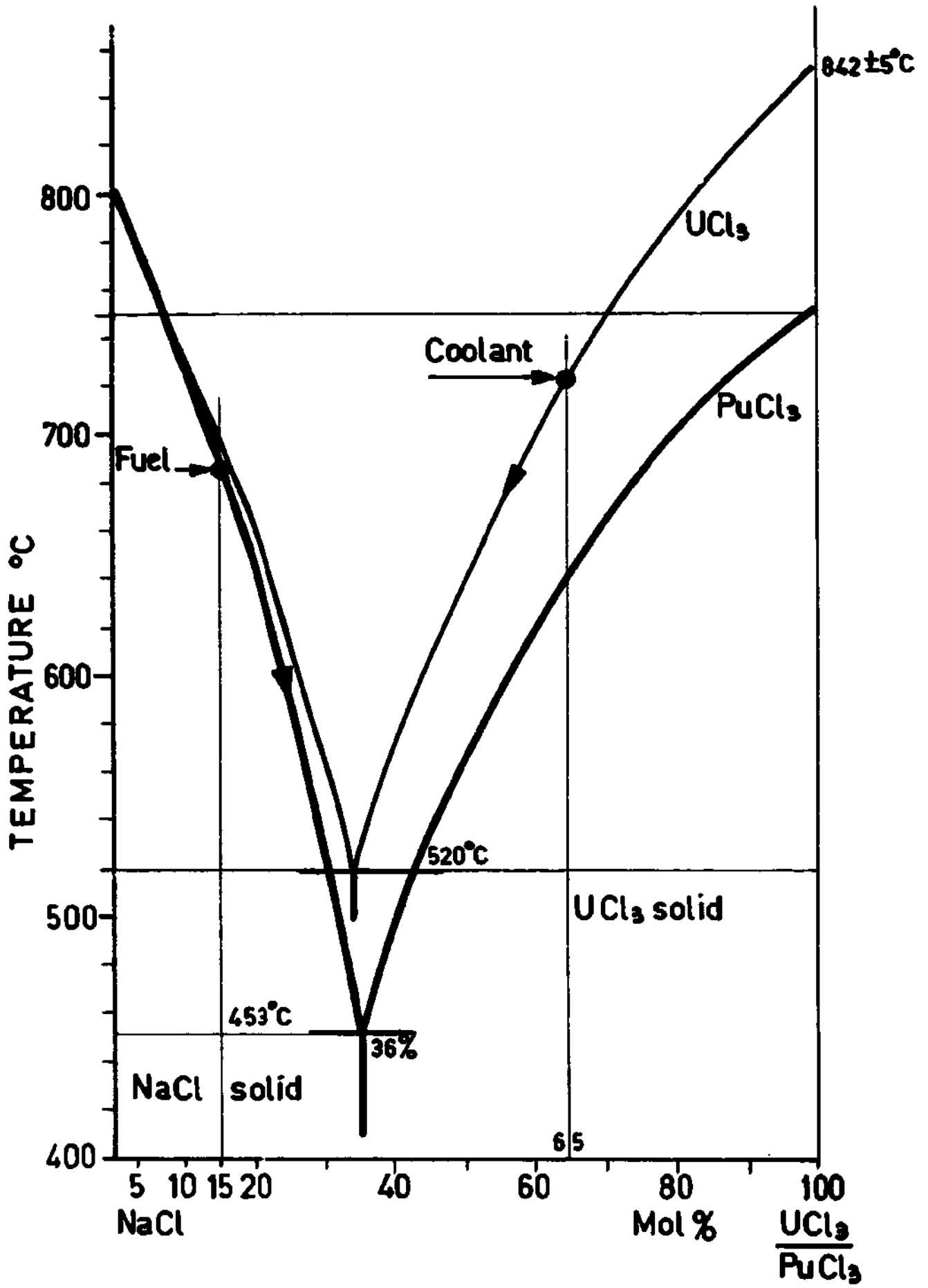|	$PuCl_3$	UCl_3	$NaCl$
Molecular weight	348.3	347	58.4
Postulated molar ratio-fuel	0.15	---	0.20
- blanket material	---	0.65	0.20
Density solid state $(kg.m^{-3})$	5.7	5.57	2.14
Melting point $(^{\circ}C)$	767	835	800
Boiling point at atmospheric pressure $(^{\circ}C)$	1730	1720	1465
Melting enthalpy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

Table A2

Other chlorides of plutonium, uranium and alumin...

	Plutonium	Uranium		
	PuCl$_4$	UCl$_4$	UCl$_5$	UCl$_6$
Melting point^{o}C	All efforts to produce pure solid PuCl$_4$ have been unsuccesful; only in gaseous	590	(287)	178
Boiling point^{o}C	state with free chlorine, or in molten salt solution or in aqueous solution as complexes	792	(417)	(372)
Free enthalpy of formation at 1000°K (kJ/mol) at 2000°K	4 x -180 = -760	4 x -182 = -768	5 x -165 = -825	6 x -: = -'

Fig A2 PHASE DIAGRAM
for Pu Cl₃/NaCl and UCl₃/NaCl

MoCl5
268

AlCl₃
180

ZrCl4
333

TiCl₄
136

NbCl5
245

FeCl₃
319

CrCl₂
815

NiCl₂
987

MnCl₂
1190

ZrCl₂
1207

CuCl
1490

1750

1026

1428

1627

17

FeCl2

MgCl₂

NaCl   CaCl₂

UCl₃

BaCl₂

PuCl₃

Maximum
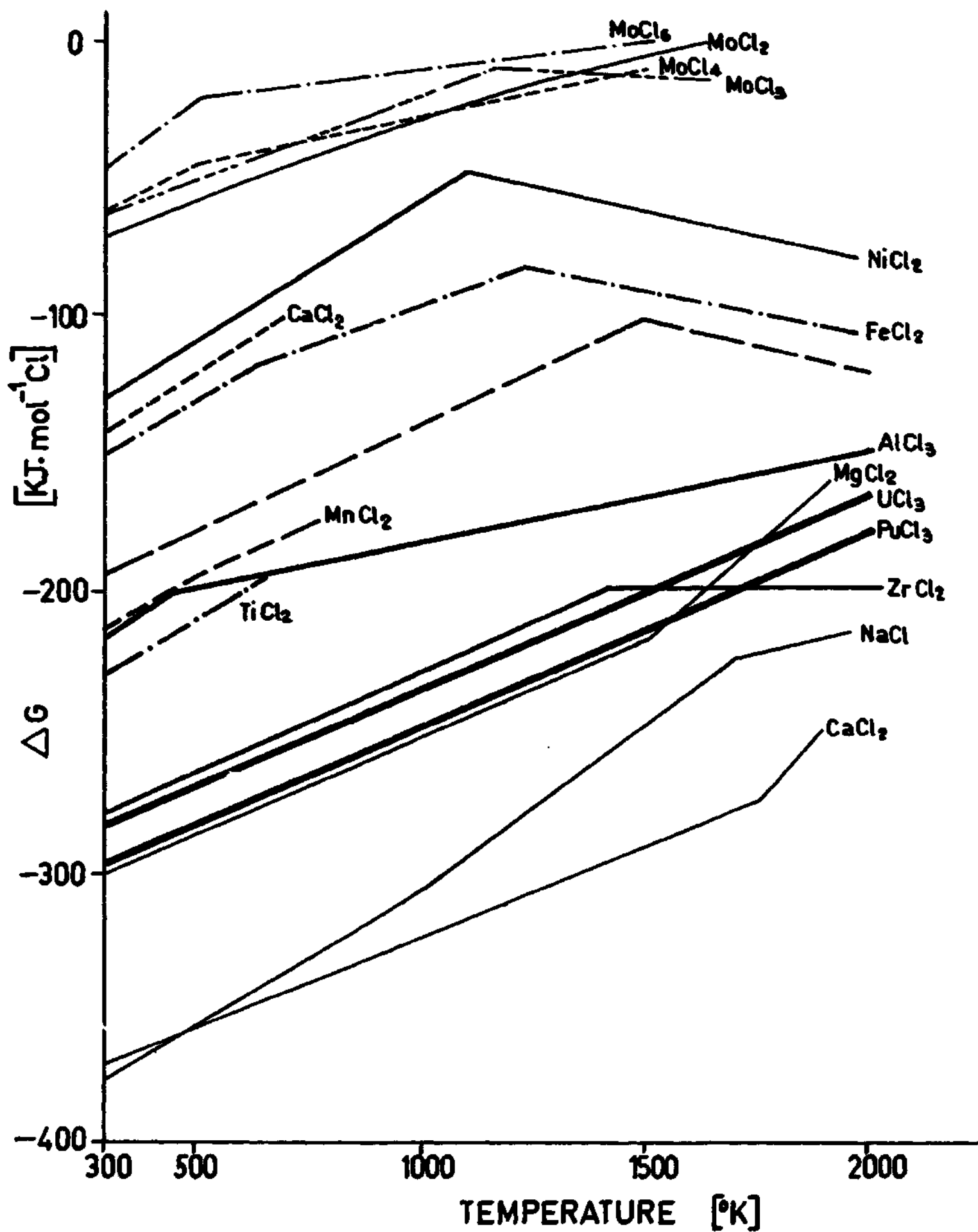fuel temperature
in this reactor

PRESSURE   [Bar]

TEMPERATURE   [°C]
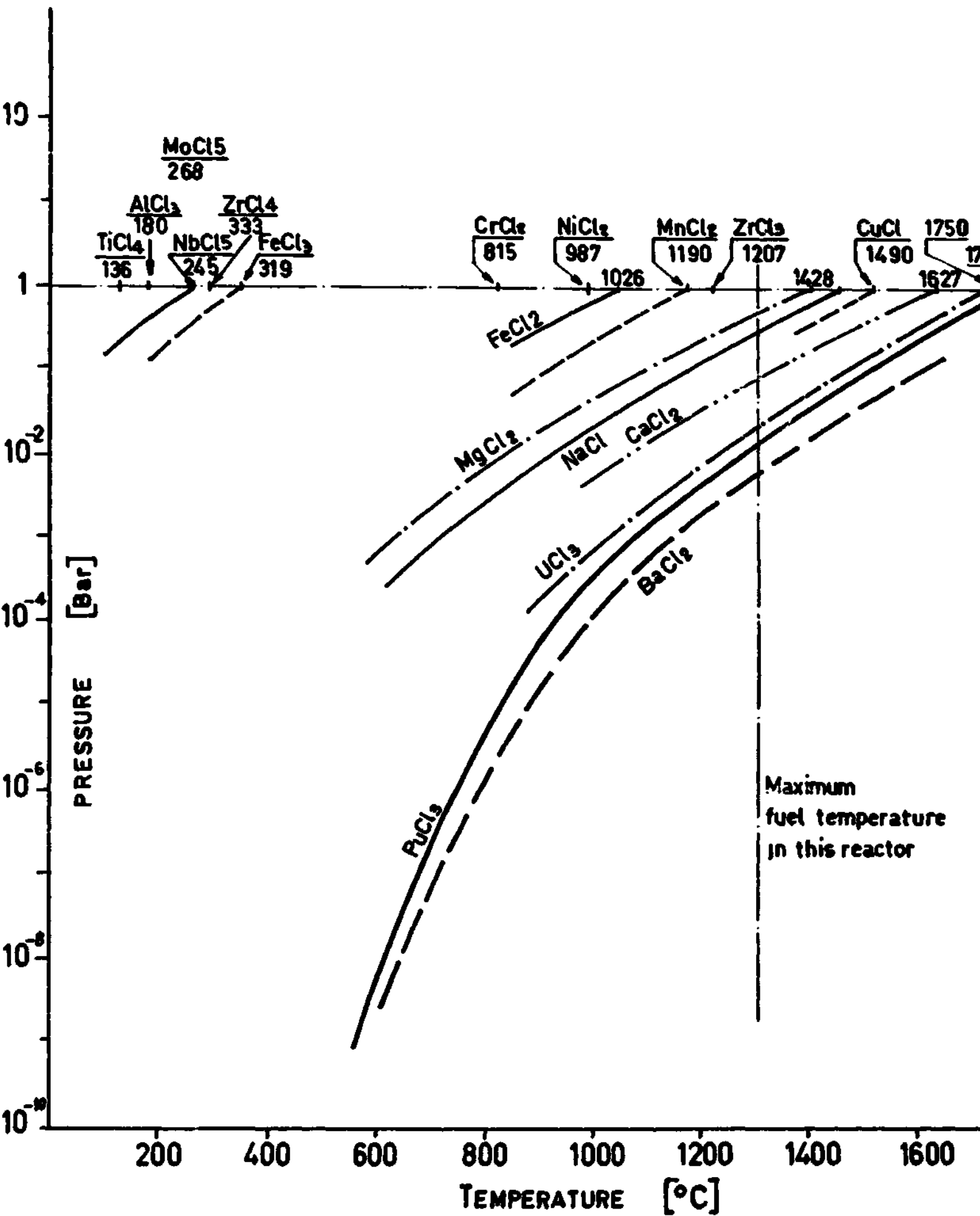
VAPOUR   PRESSURE — METAL CHLORIDES
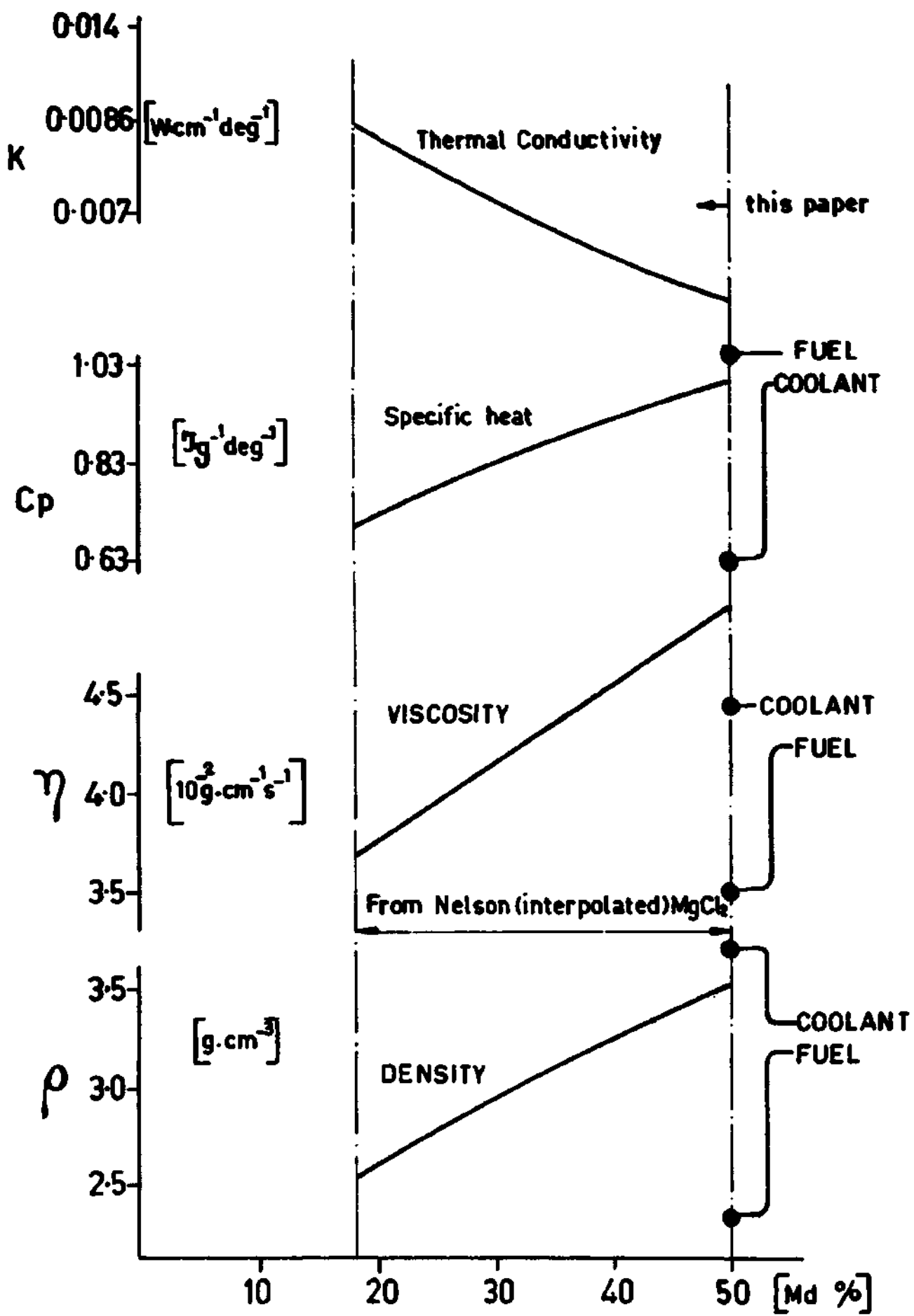
Salts properties at 650°C
v. Chemical composition
Data derived from Nelson
for UCl₃/PuCl₃/MgCl₂/NaCl

Fig. A4

FREE ENTHALPY OF FORMATION
CHLORIDES

Acknowledgement

The authors would like to acknowledge the valuable assistance given by R. Stratton in the preparation of the text and O. Koller in the preparation of the figures.

Authors' Note

During the time in which this report was prepared and printed. one of us (J.L.) has made more sophisticated neutronics calculations which result in the following

- for the ratio Fertile/Fissile material given in this paper the critical buckling was overestimated.

- An improved figure for the core results when the ratio Fertile/Fissile material is reduced from 6.60 to 5.65. This is possible by a slight change in the tube/cell ratio and increase in the coolant velocity.

Recent data shows that the total cross section for chlorine used here was overestimated by a factor 2.

The results of the revised neutronics calculations will be given in a further paper.

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