MOLTEN PLUTONIUM CHLORIDE FAST BREEDER REACTOR COOLED BY MOLTEN URANIUM CHLORIDE

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Abstract—A fast breeder reactor of 2000 ~MWt output using molten chlorides as fuel and coolant is discussed. Some of the most significant characteristics are: The liquid fuel contains only $PuCl_3$/NaCl. The coolant is molten UCl_3/NaCl and also forms the fertile material along with the blanket system, again UCl_3/NaCl: the coolant blanket system is divided into 2 or more independent circuits. The fuel circulates through the core by forced convection; the core is not divided. The thermal stability of the reactor is very good. Power excursions or fuel temperature transients are quickly damped by the phenomena of fuel expansion pushing part of the fissile material out of the critical zone. The loss of coolant accident results in a loss of half (or ⅓, ⅔) of blanket which, without relying on a reactor scram, results in an automatic adjustment of the reactor power level. Corrosion effects form the most difficult problem. Thermodynamic studies suggest the use of molybdenum alloys as structural materials.

Breeder reactors differ from other reactor types in that they are not only power-producing devices but also a source of fissile material and therefore should be considered as part of a complex "breeding system" which includes both the power reactor (producing electricity and heat) and the reprocessing and fuel preparation plant. From this point of view the fast reactor with molten fuel seems to be better adapted to the long-term secure breeding system than are the solid-fuel reactors.

The reactor system described here being a continuation of a long-term development study of a molten-chlorides breeder (Bulmer, 1956; Chasanov, 1965; Nelson *et al*., 1967; Taube, 1961) demonstrates some of these advantages, among others a very high inherent safety (without relying on engineered devices) against changes of power level and loss-of-coolant accident.

1. GENERAL DESCRIPTION OF THE REACTOR

In this paper a molten-chloride fast breeder reactor (MCFBR) with in-core cooling using fertile material as coolant is discussed. The most important features are shown in Table 1 (Taube, Ligou 1972).

Liquid fuel consisting of (mol %) 16% $PuCl_3$ and 84% NaCl.

Liquid fertile material and core cooling agent (mol %) 65% $^{238}UCl_3$, 35 % NaCl.

The flow of the blanket-coolant agent is as follows—through U tubes in the core—heat exchanger—pump—blanket region—return to core (Figure 1).

The coolant blanket circuits are in the form of at least two independent systems: the core vessel is not divided.

The core is directly connected with: overflow buffer vessel (for thermal expansion of fuel), emergency fuel drainage tank, reprocessing and fuel preparation plant, and an off-gas system (with continuous purification) for removing the volatile fission products and for control of corrosion processes.

The structural material is molybdenum and iron; the corrosion phenomena might be controlled in-situ by means of a volatile reagent added to the off-gas system.

The coolant is circulated through the U-tubes with a velocity of 9 m/s, the coolant pump is out-of-core; the fuel is circulated within the core itself with a velocity of 2 m/s (the pump being in-core).

The advantages brought about by this arrangement are as follows.

No separate 'foreign' cooling agent in the core results in an improvement in the neutron balance and the unique behavior of the system.

The unification of the blanket material and cooling agent results in a system which has a very strong negative reactivity in the case of loss-of-coolant accident (LOCA), which in this system equals the loss of blanket material accident. This results in a spontaneous 'self-reduction' of the criticality thus stopping the fission reaction without any 'engineered' safeguards (control rods, valves etc.) since the reactivity decrease by $8\%/N$ where N is the number of independent circuits only one remaining intact.

Table 1. Molten chloride fast breeder reactor (MCFBR) "CHLOROPHIL"

Electrical power, approximate	MWe	~800
Thermal power, total/in core	MWt	2050/1940
Core volume	m^3	8.75
Specific power	MW/m^3	220
Core geometry	m	height 2.0/2.36 dia.
Fuel: liquid PuCl$_3$/NaCl	mole %	16/84
Liquidus/boiling point	K	~960/~1770 (approx)
Fuel mean temperature	K	1260
Fuel volume fraction in the core		0.386
Power form-factors radial/axial		0.60/0.78
Fast flux, mean across core	n/(cm·s)	7×10^{15}
Fuel density at 984°C	kg/m^3	2344
Heat capacity 984°C	kJ/(kg·K)	0.95
Viscosity 984°C	g/(cm·s)	0.0217
Thermal conductivity, at 750°C	W/(cm·K)	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(t)/kg	0.67
Mean plutonium specific power in entire system	MW(t)/kg	0.62
Coolant: liquid ^{238}UCl$_3$/NaCl	mole %	65/35
Liquidus/boiling point	K	~980/~1970
Coolant temperature inlet/outlet	K	1023/1066
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
Thermohydraulics		
Fuel (shell side, pumped), velocity	m/s	2
Coolant, velocity	m/s	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		8.5/10.5

The division of this coolant system into (at least) two totally independent circuits with a "crossed over" lattice of U-tubes in the core plus the circulation of liquid fuel (even when assumed to be low) makes possible the removal of residual fission product heating; only one undamaged circuit being necessary to achieve this.

The off-gas system makes it possible to continuously remove part of the noble gases and other volatile fission products from the core (such as iodine) and to control the corrosion processes on the core structural materials.

The plutonium-containing salt (fuel) is in general separate from the uranium-containing salt (coolant and blanket material) which simplifies the technology of reprocessing, improves the corrosion problems and at the same time diminishes the problem of a leak in either direction particularly when the continuous reprocessing removes the "foreign" component in both the fuel and blanket coolant. The compatibility of fuel and blanket coolant is excellent.

The very strong negative temperature coefficient of reactivity arising out of any power excursion we get an increase of fuel temperature causing a decrease in density and an ejection of part of the fuel material from the core region through specially arranged tubes.

The continuous reprocessing plant (using salt-liquid metal transport processes) means a reduced plutonium inventory and an increased load factor for the plant and enhances the effective doubling time of the system. However, the disadvantages are also numerous.

The first and most important disadvantage is of course corrosion. The molten chloride medium, especially in neutron and gamma fields at high temperatures and velocities with virtually free chlorine present from the fission of plutonium chloride, presents
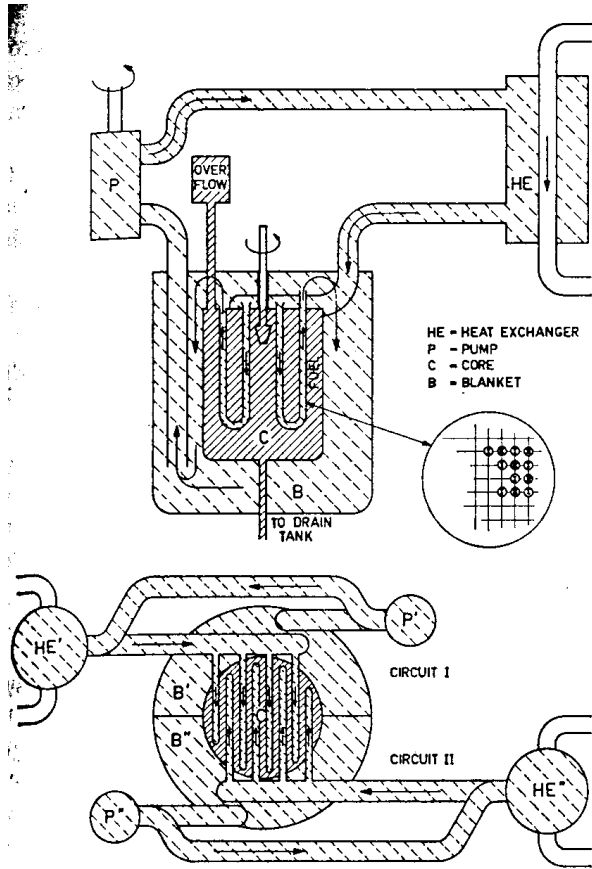
a very serious problem which has to be solved (perhaps by means of continuous control of the chemistry in-situ by means of volatile agents).

The most likely structural material seems to be a molybdenum alloy which among other things gives rise to parasitic absorption of neutrons (see the neutron balance: Table 2).

The fuel is circulated by a pump which must be located in or close to the core which increases the corrosion problems. Other possibilities for increasing the heat transfer in the fuel are probable, but not easy to realise.

The high fuel and coolant velocities result in high pumping costs and could cause erosion and increase of hydrodynamic pressure.

2. REACTOR PHYSICS

In order to define the neutron spectrum in this reactor a 22-group cross-section set has been prepared; the code GGC3 (Adir *et al.*, 1967) which allows 99 group calculations for a rather simple geometry has been used for this condensation. Most of the GGC3 library data were evaluated by GGA before 1967 but some are more recent.

All whole reactor calculations have been made on the basis of transport theory using the 22-group cross-section set in spherical and cylindrical geometry. The transport methods were aimed at getting a better accuracy for some reactivity coefficients.

HE = HEAT EXCHANGER
P = PUMP
C = CORE
B = BLANKET

Figure 1: Circuit Schematic

Table 2: Neutron balance (%)

Region		Atoms (cm^3 x 10^{21})	Absorption		Leakage	Production
Core	^{238}U	3.5629	25.50	22.51 (n,γ) 2.99 (n,f)		8.23
	^{239}Pu	0.66796	34.56	5.58 (n,γ) 28.98 (n,f)		85.55
	^{240}Pu	0.16699	3.78	2.24 (n,γ) 1.54 (n,f)		4.72
	Na Cl	6.3017 19.495	0.26 3.16	1.10 (in fuel) 2.06 (in coolant)		
	Fe Mo F.P.	5.078 0.7386 0.0679	1.30 2.04 0.50			
Total core			71.10		27.40	98.50
Blanket	^{238}U	6.42	23.70	23.15 (n,γ) 0.55 (n,f)		1.50
	Na Cl	3.457 22.72	0.08 2.22			
Total blanket			26.00		2.9	1.50

The detailed neutron balance is given in Table 2; for more detailed information see Ligou (1972). The spectrum of the MCFBR compares favorably with that of a LMFBR.

From Table 2 one can deduce that k = 1.385. If δk is the loss of reactivity due to some parasitic absorption the corresponding behaviour of the breeding ratios are:

$$C_{core} \approx 0.716 - 1.72\delta k$$
$$C_{blanket} \approx 0.670 - 1.05\delta k$$
$$C_{total} \approx 1.386 - 2.77\delta k$$

Taking into account the latest data the problem of parasitic absorption of the chlorine is not dramatic so there appears to be no need to enrich the chlorine (^{37}Cl) which is consistent with the conclusion of Nelson et al. (1967). The neutron spectrum shows a peak which corresponds to the chlorine cross section minimum (65% of neutrons are in an energy range where $\sigma_{Cl} < 5$ mb). This fact was perhaps not recognized 15 yr ago when fine spectrum calculations were not possible; it would perhaps explain the pessimistic conclusions of several eminent physicists (Weinberg and Wigner, 1958) about the use of natural chlorine in this type of reactor.

For the chosen structural material (20% Mo) the reactivity cost of 2% is quite acceptable (see Table 2). However, the cost could rapidly become rather prohibitive if the volume of the structural material and/or molybdenum content should increase for technological reasons. Using the same structural materials (20% Mo) the loss of reactivity due to the vessel is δk(%) = 0.72le where e is the vessel thickness in cm.

Five complete transport calculations have been made for different coolant or fuel densities, and different temperatures (Ligou, 1972). If one considers that all the density modifications come from thermal expansion (liquid phase only) one can define general temperature coefficients

$$\frac{\delta k}{k}(\%) \cong -0.038\delta T_{fuel} + 0.0129\delta T_{coolant}$$

In the second term the part played by the Doppler coefficient (0.00048) is quite negligible. For +100K in the isothermal core the loss of reactivity is –2.5% which is very important from the safety point of view. Compared to any other type of power reactor (even the BWR) the advantage of this kind of reactor on the grounds of inherent safety is quite evident.

The "feedback effect" which is very important for kinetics studies is as follows

$$\frac{\delta k}{k}(\%) \cong 60\left(\frac{\delta\rho}{\rho}\right)_{fuel} - 15\left(\frac{\delta\rho}{\rho}\right)_{coolant} - 0.00048\delta T_{coolant}$$

The void coefficient of the fuel (1st term) is strongly negative; 1% void

$$\frac{\delta\rho}{\rho} = -10^{-2}$$

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

3. THERMOHYDRAULICS

This type of reactor, has fuel circulating in the core only, with a relatively low temperature gradient of approximately 35°C and a high heat capacity, and a high velocity coolant ~9 m/s with again a low temperature gradient of approximately 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 is too low giving decrease of a specific power which is 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; minimization of the fuel leakage, minimization of the auxiliary power, optimization of the fuel flow regulation—all point 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.).

4. FISSION PRODUCTS AND CORROSION PROBLEMS

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 constituents 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 (see Figure 2). 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 behavior 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/mole 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.

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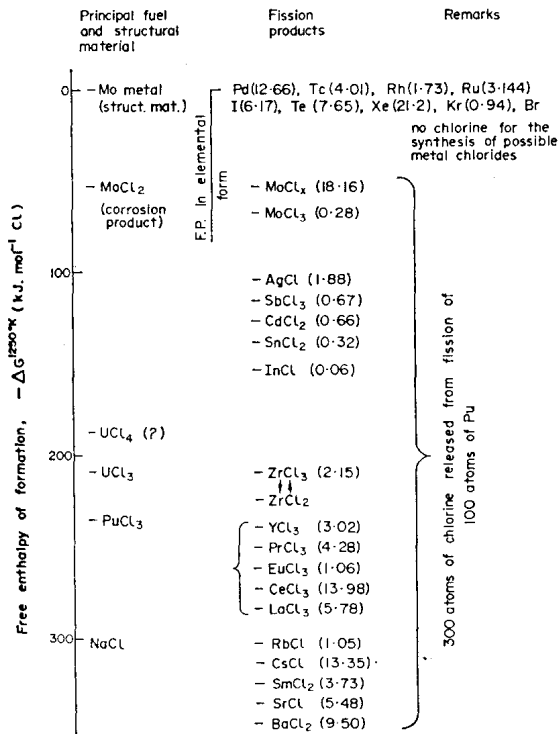

Figure 2: The behavior of fission products in the molten-chloride fuel. (Yields given represent products for 100 Pu atoms fissioned.)