## Eidg. Institut für Reaktorforschung Würenlingen Schweiz

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

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## Abstract

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

- a) the precursors of the delayed neutrons nearly all remain
- b) iodine-131 in the steady-state core is reduced approxby a factor 1000
- c) caesium-137 concentration in the core decreases only by a factor 7, but the volatility is relativ low due to chloride
- caesium isotopes 135 and 133 can be separated from Cs-137 by a factor of which make easier the Cs-137 management
- e) xenon isotopes are extensively extracted

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

## 1. General Remarks

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

The design philosophy of a safe reactor is guided by the statements of the following type: (WASH 1250, 1973) "The measures against the <u>escape</u> of radioactivity from nuclear facilities (nuclear power plants, fuel reprocessing waste disposal facilities, shipping, storage etc.) should use design features inherently favourable to safe operation e.g. by selection of fuel, coolant and core structural materials which will have inherent stability and safe characteristics."

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

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

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

## 2. Continuous gas extraction

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

- a) the reactor is fuelled with molten salt fuel in this case molten chlorides  $PuCl_{3} UCl_{3} NaCl etc.$  (fig. 1)
- b) the chemical properties of the fission products in this molten media are characterised by the scheme shown in (fig. 2)
- c) the balance of the fission products especially for chlorine is given in (fig. 3)
- d) the thermodynamic stability of all the important irradiated fuel components are given in fig. 3
- e) the volatility (partial pressure) of some selected crucial fission products: e.g. caesium in the form of element, oxide and chloride is given in fig. 4



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

Kr	54	Хе				/	~	fast	
Br	53	I				$\langle$	Gas Extraction	slow	
Se	52	Те						very	slow
As	51	Sb				/	Volatile ch	nlorid	es
Ge	50	Sn							
Ga	49	In				/	Low volati	le	
Zn	48	Cd					chlorides		
	47	Ag							
	46	Pd				/	Non		
	45	Rh				$\langle$	volatile		
	44	Ru					metals		
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	41	Nb				$\langle$	volatile		
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	<b></b>		64	Gd					
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Fig. 2 Fission Products in Molten Chlorides Media







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Fig. 5 Scheme of the independent Yield Calculation



Fig. 6 Scheme of Gas-Extraction

- f) the history of each individual fission product is calculated on the basis of the independant yields: see fig. 5 (according to Crouch 1973)
- g) the in-core molten salt medium is purged by means of heliumhydrogen gas bubbles which greatly influence the chemical behaviour of some fission products (particularly iodine). The assumptions concerning the size of this stream is given in Appendix 1
- 3. Scheme of gas extraction and possible technology of gas separation

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

## 4. Scheme of calculation

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



The only assumption arbitrarily made was the gas extraction rate of the volatile fission products.

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

### Table 1 Core dwell time - (seconds)

	Variant								
	1	2	3	4					
Se and Te	10 <sup>6</sup>	109	10 <sup>4</sup>	10 <sup>3</sup>					
Br and I	10 <sup>6</sup>	lo <sup>4</sup>	10 <sup>2</sup>	102					
Kr and Xe	10 <sup>6</sup>	10 <sup>3</sup>	10 <sup>2</sup>	lol					
All other elements *)	106	106	106	10 <sup>6</sup>					
*) 10 <sup>6</sup> seconds equals	11.57	days which	is	postulated as the					

reprocessing period of the total liquid fuel

## 5. Results of the calculations

The most important results are:

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

- 2. Short lived bromine isotopes A = 88, 89, 90, being also the probable precursors of delayed neutrons are in practice not extractable (no loss of delayed neutrons) (fig. 9 - linear vertical scale)
- 3. The very high retention of the heavy bromine isotopes in the molten fuel is almost independant of the rate of gas extraction. For the highest gas 'extraction' only a small extraction of bromine occurs (fig. 9).
- 4. The main problem for krypton isotopes is the Kr-85 (fig. 10). The gas extraction is successful for the lowest case (2) and results in a reduction of the amount of Kr-85 by approx 1000 times.

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

- 5. The gas extraction has only a small effect on the two radioactive strontium isotopes (fig. 12). They are decreased only by a factor of 7.
- 6. The case of the iodine isotopes (fig. 13 and 14) is critical for two reasons
  - I) the iodine-131 is an important factor in the core accident hazard.

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Fig. 8

The figures 8-17 are made directly by the CDC 6500/6700 prin-

ter as output from GASEX; If \* appears then at least two

curves are superimposed.

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Fig.

GRAPH	1	ELEMENT J AT.N	VO. 53	ISOTOPE 127	び手点もしう		
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GRAPH	2 -	ELEMENT J AT.N	NO. 53	ISOTOPE 129	HALF-LIFE 1.TOBE+07	YRS	
1.00E-08		1.008-86		1.008-04	1.088-02	1.09E+1	1.002+02
GRAPH	3 -	ELEMENT J AT.N	NO. 53	ISOTOPE 130	HA.F-LIFE 1.239E+01	HRS	
1.00E-08		1.008-36		1.902-04	1,008-02	1.03641	1.002+02
GRAPH	ių	ELEMENT J AT.N	VO. 53	ISOTOPE 131	HALF-LIFE 8.050E+00	DAY	
1.00E-08		1.007-36		1.00E-04	1.008-82	1.002+1	00 1.00E+02
GRAPH	5 -	ELEMENY J AT.N	NO. 53	ISOTOPE 132	HALF-LIFE 2.400E+00	HKS	
1.00E-08		1.00E-06		1.108-04	1.00E-02	1.008+1	1.00E+82
GRAPH	6 -	ELEMENT J AT.N	10. 53	ISOTOPE 133	HALF-LIFE 2.080E+01	HRS	
1.00E-08		1.00E-06		1-90E-04	1.088-02	1.001.+1	00 <b>t.00F+02</b>
GRAPH	7 -	ELEMENT J AT.N	NO. 53	ISOTOPE 134	HALE-LIFE 5.200E+01	MIN	
1.00E-08		1-00E-06		1.00F-04	1-485-02	1.005+0	1.00E+62
GRAPH	8	ELEMENT J AT N	ND. 53	ISOTOPE 135	HALF-LITEE 6.700E+00	HeS	
1.008-08		1.00F-06		1-00F-04	1-80E-02	1.096+1	1.005+02
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GRAPH	1 -	ELEMENT XE AT.NO.	54	ISOTOPE 129	SIDOLE					
1.00E-08		1.00E-06		1.00E-04	1.00E-	02	1.00€+	00	1.00E+02	
GRAPH	2 -	ELEMENT XE AT.NO.	54	ISOTOPE 130	STABLE					
1.00E-08		1.00E-06		1.00E-04	1,00E-	02	1.00E+	0 0	1.002+02	
GRAPH	3 -	ELEMENT XE AT.NO.	54	ISOTOPE 131	STABLE					
1.00E-08		1.00E-06		1.00E-04	1.00E-	02	1.00E+	0.0	1.00E+02	
GRAPH	4 -	ELEMENT XE AT.NO.	54	ISOTOPE 132	STABLE					
1.008-08		1.00E-96		1.00E-04	1.00E-	0.2	1.00E+	00	1.00E+02	
GRAPH	5 -	ELEMENT XE AT.NO.	54	ISOTOPE 133	HALF-LIFE 5.650	E+D0 DAY				
1.00E-08		1=00€−06		1.U0E-04	1.008-	02	1.00E+	0 0	1.00E+02	
GRAPH	6 -	ELEMENT XE AT.NO.	54	ISOTOPE 134	STABLE					
1.00E-08		1.00E-06		1.00E-04	1.00E-	02	1.00E+	00	1.00E+02	
GRAPH	7 -	ELEMENT XE AT.NO.	54	ISOTOPE 135	HALF-LIFE 9.150	E+00 HRS				
1.00E-08		1.008-06		1.00E-04	1.008-	02	1.00E+	00	1.00E+02	
GRAPH	8 -	ELEMENT XF AT.ND.	54	ISOTOPE 136	STABLE		-			
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ELEMENT XE WITH AT.NO. 54 - ISOTOPE 144 HALF-LIFE 1.000E+00 SEC HAS RM = INDEF.

Fig.

GRAPH 1 -	ELEMENT XE AT.NO. 54	ISOTOPE 137 H	ALF-LIFE 3.900E	+00 M1N	
1-00E-08	1.00F-06	1.00E-04	1.00E-0	2 1.00E+	00 1+00E+02
GRAPH 2 -	FIENENT XE AT-10. 54	TSOTOPE 138 H	ALF-LIFE 1.410E	+01 HIN	
1 005-08	1-005+06	f = 0.0E = 0.4	1.00F-8	2 1.00E+	00 1.00E+02
COADU 7	ELEMENT VE AT NO SA	TSOTOPE 139 H	ALE-1 TEE 4.100E	+01 SEC	
4 005-00	4 00E-06	1 005-04	1-00E-0	2 1-0DE+	0.0 1.00E+02
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GRAPH 4 -	ELEMENT XE AT.NU. 54	ISUIDPE 140 H	ALF-LIFE 1.350E		0.0 1_00E+02
1.00E-08	1.30E-06	1.0UE+04		1.000	
GRAPH 5 -	ELEMENT XE AT.NO. 54	IS010PE 141 H	ALF-LIFE 1./UUE	+UU SEU	
1.00E-08	1.09E-06	1.00E-04	1.UUE-0	2 1.UUE+	UU 1.UUC+U2
GRAPH 6 -	ELEMENT XE AT.NO. 54	ISOTCPE 142 H	ALF-LIFE 1.200E	+00 SEC	
1.00E-08	1.00E-06	1.00E-04	1.00E-0	2 1.00E+	00 1.00E+02
GRAPH 7 -	ELEMENT XE AT.NO. 54	ISOTOPE 143 H	ALF+LIFE 1.000E	+00 SEC	
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Fig. 16

ALL ISOTOPES OF ATOMICNUMBER 55

GRAPH	1 - ELEMENT CS	AT.NO. 55 IS	OTOPE 13	3 STABLE					
1.00E-08	1.00E-0	6	1.00E-04	1.	90E-02	2	1.00E+0	1.00E	+02
GRAPH	2 - ELEMENT CS	AT.NO. 55 IS	OTOPE 13	5 HALF-LIFE 2	2.000E+	⊧06 ¥RS			
1.00E-08	1.00E-0	6	1.00E-04	1.	.00E-02	2	1.00E+0	1.00E	+02
GRAPH	3 - ELEMENT CS	AT.NO. 55 IS	OTOPE 13	7 HALF-LIFE 3	3.000E+	+01 YRS			
1.00E-08	<b>1.</b> 00E-0	6	1.00E-04	1.	00E+02	2	1.08E+0	10 <b>1.00E</b>	+02
GRAPH	4 - ELEMENT CS	AT.NO. 55 IS	OTOPE 13	8 HALF-LIFE 3	3.230E+	+01 MIN			
1.00E-08	1.00E-0	6	1.00E-04	1.	00E-02	2	1.00E+0	1.00E	+82
GRAPH	5 - ELEMENT CS	AT.NO. 55 IS	OTOPE 13	9 HALF-LIFE 9	9.000E+	+00 MIN			
1.00E-08	1.00E-0	6	1.00E-04	1.	006-02	2	1.00E+0	1.00E	+02
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II) The isotopes I-136, I-137, I-138, I-139 and I-140 are probably the most efficient precursors of delayed neutrons.

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

- 7. The xenon isotopes are also an important factor in the core accident hazard. Gas extraction decreases the amount of xenon in the core significantly including the long lived Xe-133 and Xe-135 (fig. 15 and 16). The short life xenon isotopes are only slightly influenced by the gas extraction.
- 8. The next controlling radionuclide is Cs-137. From fig. 17 it is seen that the gas extraction does not aid in the removal of this nuclide from the molten fuel, a factor of 7 is all that can be obtained. But the amount is not the only important factor, the volatility of caesium is also of interest.

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

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

#### ALL ISOTOPES WITH HALF-LIFE BETWEEN 0.3 AND 3 Y

GRAPH 1 + ELEMENT RU	AT.NO. 44 ISOTOPE 10	6 HALF+LIFE 1.000E	+OO YRS	
1.005-08 1.005-0	6 1.00E-04	1.005-03	2 1.00E+	00 1.006+02
GRAPH 2 - ELEMENT SO	AT.NO. 51 ISOTOPE 12	5 HALF-LIFE 2.700E	+00 YRS	
1.00E-08 1.00E-0	16 1.00E-04	1.00E=03	2 1.00E+	00 1.00E+02
GRAPH 3 - ELEMENT CE	AT.NO. 58 ISOTOPE 14	4 HALF-LIFE 2.840E	+O2 DAY	
1.00E=08 1.00E=0	16 1.00E-04	1.00E-0	2 1.00E+	00 1.006+02
GRAPH 4 - ELEMENT PM	AT.NO. 61 ISOTOPE 14	7 HALF-LIFE 2.620E	+00 YRS	
1.00E-08 1.00E-0	6 1.00E-04	1.006-03	2 1.00E+	00 1.00E+02
GRAPH 5 - ELEMENT EU	AT.NO. 63 ISOTOPE 15	5 HALF-LIFE 1.810E.	+00 YRS	
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GRAP	H 1 - ELEMENT KR	AT.NO. 36 ISOTOPE	85 HALF-LIFE 1.076	E+U1 YRS	
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GRAP	H 2 - ELEMENT XE	AT.NU. 54 ISOTOPE	135 HALF-LIFE 9.150	E+00 HRS	
1.00E-0	8 1.0nE-	06 1.00E-	04 1.00E+	02 1.00E+	0u 1.00E+02
GRAP	H 3 - ELEMENT CS	AT.NU. 55 ISOTOPE	137 HALF-LIFE 3.000	E+01 YRS	
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#### NJ6 WITH 1885, N54 WITH IS135, N55 WITH IS137

26

Fig. 19

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

I) the neutron absorption in other isotopes

II) the increased volume.

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

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

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

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

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

- 9. Fig. 19 shows the most hazardous nuclides together Kr-85, Sr-90, Xe-135, Cs-137.
- 10. Fig. 20, 21, 22 show the estimated values of radioactivity for some selected nuclides in a 2000 MW(th) fast reactor for different cases
  - a) without continuous gas extraction and with 85 days irradiation period.
  - b) with gas extraction at the highest rate
  - c) with gas extraction at the lowest rate.

#### Conclusions

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

- The extraction rate of the precursors of the delayed neutrons can be reduced to an insignificant value - even if these are very volatile. e.g. Kr, Xe, Br, I etc.
- 2. The extraction rate of I-131, the most hazardous fission product, prominent in any severe core-accident, is so high that the steady state concentration of this nuclide is reduced by more than 1000 times which significantly improves the reactor safety.













Seconds

- 3. The extraction rate of Cs-137 is rather small and results in the diminishing of the caesium concentration by only 7 times when compared to the normal liquid fuel reprocessing having a time constant of  $10^6$  seconds or  $\approx 10$  days mean dwelling time of the liquid fuel in the reactor. In the solid fuel case the same amounts of Cs-137 are accumulated after approx 100 days that is ~3.5 months dwell time in the core. A special feature of the molten chlorides fuel is that the vapour pressure of CsCl is much lower than CsO<sub>2</sub>/Cs metal in solid oxide or carbide fuels, decreasing the amount of vapourised caesium in the case of a core accident. Another important point is that it appears possible to separate the caesium isotopes 135 and 137.
- 4. Further hazardous fission products such as Xe-133 and Xe-135 can also be effectively extracted and the steady state level is >100 times lower than in the unextracted core.
- 5. The gas extraction rate influences also the oxygen extraction from the molten fuel which considerably improves the corrosion behaviour of metallic molybdenum and also the stability of the uranium and plutonium chlorides (Taube 1974).

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## Appendix I

Calculation of the gas stream rates. The 2000 MW(th) reactor has a fission rate of about  $2 \times 10^9$  W x 3.1 x  $10^{10} \frac{\text{fiss/s}}{W} = \frac{1}{6.02 \times 10^{23}} \approx 100 \,\mu\text{M Pu/s}$ 

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

The relative amounts of volatile nuclides is estimated very roughly here as 0.2, that is 2 x 20  $\mu$ M/<sub>s</sub> of gaseous volatile elements. The volume of these elements at ~950 <sup>O</sup>C and ~10 bars pressure is

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

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

The total volume of fuel in the core equals  

$$8.75 \frac{m^3}{m} \times 0.386$$
 fuel fraction = 3.377 m<sup>3</sup>  
core

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

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