

# EUR 5010 e

COMMISSION OF THE EUROPEAN COMMUNITIES

## REPROCESSING OF FAST BREEDER REACTOR FUEL USING AQUEOUS REPROCESSING TECHNOLOGY

by

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1973



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Industrial, Technological and Scientific Affairs - Brussels

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**Commission of the  
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D.G. XIII - C.I.D.  
29, rue Aldringen  
L u x e m b o u r g

September 1973

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Directorate General for Industrial, Technological and Scientific Affairs,  
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As a basis for the analysis and comparison, light water reactor fuels with a burn-up of 30 000 Mwd/ton and fast breeder core fuel of 80 000 Mwd/ton burn-up have been chosen. The reactor Na(1 on which information and

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## **ABSTRACT**

The report is meant to be an inventory of the most important problems which will be encountered during the processing of Fast breeder reactor fuels using aqueous reprocessing technique. It has been the objective to analyze the origin of the different problems and establish their importance in comparison with the actual practice in processing light water reactor fuels.

As a basis for the analysis and comparison, light water reactor fuels with a burn-up of 30 000 Mwd/ton and fast breeder core fuel of 80 000 Mwd/ton burn-up have been chosen. The reactor Na(1) on which information and calculated provisional data were readily available to the authors has been used as a specific example.

The annexes to the report contain detailed figures on fuel characteristics used in the study.

## **KEYWORDS**

REPROCESSING  
FBR TYPE REACTORS  
WATER  
SPENT FUELS  
FISSION PRODUCTS

DECAY  
HEATING  
URANIUM DIOXIDE  
PLUTONIUM OXIDES

## INTRODUCTION \*)

Aqueous reprocessing is the generally adopted technique for the reprocessing on an industrial scale of metallic and oxide type power reactor fuels.

Fast breeder fuels of the oxide type will appear under the same general form as the thermal oxide fuels. The high burn-up foreseen for fast breeder fuels will result in characteristics of the irradiated fuel which are much more extreme than those encountered with thermal fuels.

An analysis of the different steps in the reprocessing process is necessary in order to evaluate the consequences of the more extreme characteristics.

The present report is meant to be an inventory of the most important problems which will be encountered during the processing of Fast breeder reactor fuels using aqueous reprocessing technique. It has been the objective to analyze the origin of the different problems and establish their importance in comparison with the actual practice in processing light water reactor fuels. The report reflects the situation of different aspects of the problems as they were seen in 1970.

As a basis for the analysis and comparison, light water reactor fuels with a burn-up of 30.000 Mwd/ton and fast breeder core fuel of 80.000 Mwd/ton burn-up have been chosen. The reactor Na-1 on which information and calculated provisional data were readily available to the authors has been used as a specific example (Ref. - annexes I, II & III).

The annexes to the present report contain detailed figures on fuel characteristics used in the study.

## CHARACTERISTICS OF IRRADIATED FAST BREEDER REACTOR FUEL

At this moment the sodium cooled type F.B.R. is certainly the most advanced in development.

The core fuels for this reactor type will consist of mixed plutonium dioxide-uranium dioxide clad in stainless steel. Although this type of fuels has disadvantages as far as breeding ratio is

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\*) Manuscript received on April 2, 1973

concerned, they are preferred at this stage because of the experience gained with oxide fuels and stainless steel cladding in light water reactor fuels. Other compounds such as carbides or nitrides as well as other cladding materials may eventually replace the stainless steel clad oxide fuels in second generation reactors.

The core fuel of the first generation fast breeder will contain of the order of 20% plutonium dioxide. The blanket fuel will be natural or depleted uranium oxide.

The very high investment in fissile material (see table 1) for fast breeder reactors results in high charges for the fuel inventory. As a result of these high inventory charges, the economics of the fuel cycle call for minimum out of reactor time.

TABLE 1

Pu CONTENT OF UNLOADED NaI FUEL

Discharge rate *		Pu content at discharge	Total quantities. of Pu
Zone 1	1/3	18,9 %	515,5 kg
2	1/3	22,2 %	620,7 kg
3	1/3	2,9 %	73,9 kg
4	1/3	1,8 %	45,9 kg
5	1/3	1,6 %	92,6 kg
6	1/6	0,7 %	<u>43,9 kg</u>
			Tot. 1392,5 kg

\* fraction of fuel loading discharged every 8 months

Minimization of the out of reactor time will affect in the first place the cooling time and will require the handling of short cooled fuels throughout the different processing operations beginning with transport.

High specific power and high burn-up of F.B. Reactor fuel (table 2) result in very high levels of fission product activity in the discharged fuels.

TABLE 2

COMPARISON OF BASIC FUEL CHARACTERISTICS

Reactors	Light water reactor fuel		Fast breeder core fuel	
	BWR (Würgassen KKW)	PWR (Stade KKS)	Na 1	A.I.
Specific power Kw/kg	22	33	146	148
Burn-up Mwd/ton	27.000	32.000	85.000	80.000

(A.I.: Atomics International Fast breeder project)

In first approximation the short lived fission product content of the irradiated fuel when discharged is directly proportional to the specific power, while the long lived fission product content is directly proportional to the burn-up.

Consequently it can be said that for fuel characteristics as cited in the table above, fast breeder core fuels will contain, at the moment of discharge from the reactor, roughly 5 times more short lived fission product activity per ton than light water reactor fuels. This higher fission product content results in a very important heat generation in these fuels (see annex III and table 3). The heat generation becomes a problem of primary importance when the handling of short cooled fuels is considered.

At this moment it is the tendency of those engaged in fast breeder development to consider total cooling times before reprocessing ranging between 30 and 60 days. Consequently, the different operations leading to the recovery of the fissile material are analysed in view of handling short cooled fuels.

IRRADIATED FUEL TRANSPORT

Decay heat generation is the main fuel parameter of concern in the fuel transport operation as it has a determining influence on the transport cask characteristics and cask loading. The decay heat release is for an important part dependent on the short lived fission product content which in turn is directly related to the specific power and the cooling time.

Table 3 compares calculated heat generation data for the fuel of two light water reactors under construction and for the core fuel of two fast breeder reactor projects. The basic fuel characteristics for this comparison are given in Table 2.

TABLE 3

COMPARISON OF THE DECAY HEAT GENERATION FOR FUELS OF LIGHT WATER AND FAST BREEDER REACTORS W/kg fuel

Cooling time (days)	Fast breeder core fuel		Light water reactor fuel	
	Na-1	A.I.	KKW(BWR)	KKS (PWR)
30	197	162	53	70
60	139	111	36	48
90	104	88		
100			26	34
120	85	73		
150	70	62	21	28

From the above table it can be seen that the transport of 30 or 60 days cooled fast breeder core fuels will have to cope with a heat generation rate which is more than five times greater than the heat generation which is encountered during transport of 150 days cooled light water reactor fuels.

The safety regulations with which irradiated fuel transport has to comply, impose limits of temperature and radiation levels, criteria for the cooling system, as well as tests for the transport cask.

The regulations worked out by the International Atomic Energy Agency "Regulations for the Safe Transport of Radioactive Materials" are of application in the different European countries, in addition to the general transport regulations in each of these countries.

At present the packaging and transport of short cooled fast breeder core fuel is being studied in relation with the various fast breeder projects. The most advanced study is probably the shipping cask design of Oak Ridge National Laboratory (ref. 1). The overall characteristics of this cask could be summarized as follows :

- exceptionally rugged containment system
- energy absorbing members to protect the cask from high impact loads
- steel as the biological shield
- sodium as primary coolant
- arrangement of multiple fuel elements in close array inside a single fuel cavity.

Although this design study is not complete and additional work is required the author concludes that the available test data indicate that the approach is feasible.

In this study only rough indication is given of shielding to fuel weight ratio as function of the total cask capacity. In the case of twenty days cooled fuel the ratio of shielding weight to fuel weight would vary from 100 down to 40 for cask-loads from 6 to 36 A.I. subassemblies respectively. An increase in cooling time from 20 to 90 days would reduce the shielding to fuel weight ratio from 40 to 28 in the case of 36 A.I. subassemblies.

It is further made clear that the total system weight would be 30 to 40 % greater than the shielding weight alone.

## FUEL CLEANING

The fuel assemblies discharged from sodium cooled reactors will be stored under sodium for partial cooling before being transported to the reprocessing installation. The transport of short cooled fuel from the reactor to the reprocessing installation is envisaged to be done in sodium cooled containers.

Before entering the aqueous processing scheme, sodium adhering to the assemblies will have to be removed. Longer fuel cooling times at the reactor site could allow the transport to be done in water cooled containers but this would not eliminate the assembly cleaning operation, which would then have to be executed at the reactor.

Various cleaning methods for reactor parts (including fuel assemblies) have been envisaged and experimented. A number of these methods such as water cleaning (ref.2;3); steam cleaning (ref. 4;5;6;7;8) and wet gas cleaning are based on the reaction of sodium with water. The chemical reaction is exothermic and produces hydrogen which can react explosively with oxygen under certain concentration conditions and must therefore be closely controlled.

- Water cleaning which is the most straight forward method but presents favorable conditions for the explosive hydrogen oxygen reaction to occur. The chemical reaction of sodium with water is very rapid and cannot be controlled.

This fuel cleaning method has been experimented at S.R.E. During such a cleaning operation an explosion occurred and damaged the fuel assembly (ref. 9).

- Steam cleaning assures a rapid and complete reaction but necessitates a temperature of the assembly above 100°C to avoid condensation. It allows a certain control of the chemical reaction and of the gas phase conditions. The limited cooling capacity and the rapid reaction of sodium and water vapour could lead to local overheating of fuel assemblies.

- Wet gas cleaning permits lower and better controlled reaction rates through regulation of the humidity content of an inert gas. It presents no danger of explosion as an inert gas carries the humidity or vapour. The equipment to be cleaned does not need to be heated like it is the case for steam cleaning. Next to these advantages it should be pointed out that the major drawback of this cleaning method is the possibility of incomplete reaction, specially in case of complicated geometries and for this reason the method will probably have to be used in combination with water cleaning or steam cleaning.
- Distillation of sodium has been experimented and has given good results. The equipment to be cleaned has to be heated in a vacuum chamber. The major difficulty of such a cleaning method is the evacuation of the decay heat, problem which is not solved at this moment.
- Sodium evaporation under argon gas flow at temperatures over 400°C has given good results on simple geometries, but the problems for fuel assembly cleaning have not been solved yet.
- Dissolution of sodium in different media is being evaluated in connection with the following reactors :

SRE	Mineral oil
PFFBR	Al-Si alloy
RAPSODIE	Pb-Sb alloy
DFR	Lead

From available information it can be said that although various methods of cleaning of reactor parts are possible it seems that for fuel assembly cleaning at this state of the development most attention is given to steam and wet gas cleaning. Both methods are rather simple and do not necessitate elaborate and refined equipment.

FUEL STORAGE

On arrival at the reprocessing installation, reactor fuel has to be unloaded and stored for some time. Several media such as sodium, gas or water can be envisaged for both the unloading and storage.

The unloading medium will mainly depend on the cask cooling medium used. Sodium cooled transport casks will have to be unloaded under inert gas cover in a shielded cell. The unloaded fuel elements with adhering sodium will need to be stored under sodium awaiting fuel cleaning. Water or gas cooled transport casks could be handled in a way similar to the way thermal reactor fuel is handled, making use of water as shielding medium.

As existing reprocessing plants will have to handle the first fast breeder fuels, water pool storage can be expected to be the preferred system for reasons of simplicity. This presupposes that the physical state of the fuel assemblies is compatible with water and that the necessary heat dissipation is provided for short cooled fuels.

The calculated heat generation for assemblies from the different reactor zones is given in table 4 hereafter

TABLE 4

HEAT RELEASE PER FUEL ASSEMBLY (REACTOR NA-1) KW/ASSEMBLY

<u>Reactor zones</u> \ <u>Cooling time</u>	30	60	90	120	150	180
Zone 1 + 3	17,5	12,31	9,20	7,52	6,22	5,31
Zone 2 + 4	13,63	9,59	7,17	5,86	4,85	4,14
Zone 5	3,96	2,76	2,04	1,68	1,38	1,17
Zone 6	1,89	1,37	1,04	0,87	0,73	0,65

A fuel discharge from the Reactor Na-1 would be composed  
Of :

35 assemblies from zone 1 + 3 (core + axial blanket)

- 35 assemblies from zone 2 + 4 (core + axial blanket)
- 30 assemblies from zone 5 (radial blanket)
- 18 assemblies from zone 6 (radial blanket).

This would lead to a total heat release of all the assemblies discharged in one discharge operation as indicated in table 5.

TABLE 5

HEAT RELEASE OF DISCHARGED FUEL AS A FUNCTION OF COOLING TIME

Reference Reactor : Na-1

Cooling Time days	Zones 1 + 3 (35 ass) kw	Zones 2+4 (35 ass) kw	Zone 5 (30 ass) kw	Zone 6 (18 ass) kw	TOTAL kw
30	622	477	119	34	1322
60	431	336	83	25	875
90	322	251	61	19	623
120	263	205	50	16	534
150	218	170	41	13	442
180	186	145.	35	12	378

MECHANICAL PROCESSING

The practice in the head-end part of L.W.R. fuel processing has evolved from chemical decladding and dissolution to the now generally accepted chop-leach process.

Chemical decladding produces rather important volumes of liquid radioactive wastes, which have to be stored safely awaiting further treatment for final storage and disposal.

The chop and leach process has the advantage of producing metallic decladding waste in much smaller volume and offers the possibility of further volume reduction by compaction. Experience with chopping equipment has demonstrated the workability of the method and the equipment.

Other processes based on the dissolution of cladding materials in molten metals and alloys such as Zn and Sb-Cu are under study and evaluation. Interesting results have been obtained with these methods but at this state, these processes have not been tried any further than on bench scale. Apart from the potential advantages of these systems a number of operational problems, mainly on the separation of the fuel and the molten metal phase remain to be solved.

The practice of the chopping process for L.W.R. fuels consists of removing the fuel element end-pieces and cutting through the whole fuel bundle as such, at distances of about 5 cm. The pieces of the fuel pins are collected in a basket which serves as charging basket for the dissolver.

As the burn-up of to-days L.W.R. fuel is rather low (20.000 to 25.000 Mwd/ton) and the cooling time relatively long (150 days or more) there is no cooling problem during the mechanical processing phase and the operation is conducted in a normal shielded cell. In the case of F.B.R. fuels, specially short cooled fuels (30 to 60 days), the mechanical processing could be more complicated as a result of the decay heat generated in the fuel.

A report of Dr. Watson & C<sup>o</sup> from Oak Ridge (Ref. 10) (table 6) indicates the temperature attained by the centermost tube of different arrays of fuel pins at different cooling times for A.I. reference fuel. The calculated heat generation of the Na-1 reference fuel is somewhat higher than the corresponding figures for the A.I. reference fuel (table 3 and 7).

Since F.B. fuel assemblies will consist of some 150 to over 300 fuel pins the heat generation and the temperatures attained in the center of the bundle will be of the same order of magnitude as in the equivalent array (see table 6).

TABLE 6 (reference 10)

Temperatures attained by arrays of tubular fuel grouped for shearing,

Basis : A.I. reference oxide core and axial blanket fuel.

Burn-up : 80.000 Mwd/T.

Specific power : 148.15 kw/kg

Decay time (days)	Envelope temperature for center most fuel tube ° C				
	5 X 5 array	8 X 8 array	10 X 10 array	15 X 15 array	20 X 20 array
30	476	601	680	858	1.010
60	418	527	597	756	890
90	383	482	546	693	820
120	358	449	508	646	764
150	335	421	477	606	719

TABLE 7

fission product decay heat release values

Cooling time days	Kw/ass(core)		W/rod/core		W/cm of fuel rod (core)		W/kg(core)	
	A.I.	Na-1	A.I.	Na-1	A.I.	NA-1	A.I.	Na-1
30	9,97	17,5	45,1	50,9	0,37	0,52	162	197
60	6,86	12,31	31,6	35,3	0,25	0,37	111	139
90	5,42	9,2	24,9	26,7	0,20	0,27	88	104
120	4,51	7,52	20,8	21,9	0,17	0,22	73	85
150	3,83	6,22	17,6	18,1	0,14	0,18	62	70

Carbide cutting tools can operate up to temperatures of 600°C, so that the problem of cutting such assemblies will be centered mainly on the behaviour of the cladding material at these high temperatures. Forced cooling of the assemblies could be used to limit the temperature of the cladding, but this will introduce operational complication and have a repercussion on the volatile fission product collection and retention.

Apart from longer decay times, which seem to be economically unacceptable, the other alternative to forced cooling is disassembly of the fuel assemblies and regrouping of the single core and blanket fuel pins into smaller bundles for cutting.

Several fuel assembly concepts for fast reactors have been worked out to permit disassembly. For other types such as S.N.R. and Na-1, disassembly cannot be envisaged. It could however be argued that the fuel from prototype fast reactors will not need to be reprocessed shortly after discharge from the reactor and that long cooling times will eliminate the heat generation problem.

The high burn-up of fast breeder core fuels (80.000 to 100.000 Mwd/ton) may result in such embrittlement of the cladding material, that fuel pins will tend to break during the cutting operation. Such breakage could lead to small cladding fragments which could constitute a problem in the operation of the cutting machine.

Another aspect of short cooled F.B.R. fuel, which differs in an important way from L.W.R. fuel, is its volatile fission product content and the liberation of these products during the cutting operation. Short cooled (30 to 60 days cooled) F.B.R. core fuel will contain important quantities of  $I^{131}$ ;  $Kr^{85}$ ;  $Xe^{133}$  and  $H^3$ . These gaseous fission products are contained in the fuel tubes under pressures of several atmospheres.

An important part of the original content of these gaseous fission products is estimated to be set free when opening the fuel cladding, the remainder being liberated in the successive processing steps, mainly during dissolution of the fuel. The cell atmosphere in which these fission products are set free has to be treated for the removal of the activity or stored for decay before it can be released through the chimney. Hence it will be important to minimize the volume of air or gas to be treated for gaseous f.p. retention.

A peculiar aspect of the F.B.R. fuel is the presence of sodium bonding between fuel pellets and cladding material, and the possible sodium in-leakage in ruptured fuel pins. The presence of sodium inside the fuel pins could make it necessary to use an inert atmosphere inside the mechanical processing cell or at least in a part of the equipment. The use of an inert gas might lead to the recycling of the gas over a F.P. retention treatment and a cooling system. This could then result in less stringent requirements for the iodine retention yields as there would be practically no release of this gas to the atmosphere.

In actual practice for L.W.R. fuels the cut fuel fragments are collected in stainless steel baskets which serve as charging baskets for a batch dissolver. In case of fast breeder core fuel the collected fuel fragments will need to be cooled due to the high heat generation, or will have to be charged in a continuous way to the dissolver unit.

From the above considerations on heat generation it would seem preferable to disassemble the fuel assemblies and to regroup the fuel pins into smaller bundles for chopping rather than to use forced cooling on complete assemblies and eventually cool the fuel fragments.

#### FUEL DISSOLUTION AND FEED PREPARATION

The nitric acid dissolution scheme for core material of L.W.R. fuels will most probably also be of application for F.B.R. reactor fuels as extraction with T.B.P. in nitric acid medium is the most likely extraction scheme.

It had been shown that  $\text{PuO}_2$  does not dissolve in nitric acid as does  $\text{UO}_2$ . Consequently, a dissolution scheme using Hydrofluoric acid as a catalyst in the nitric acid was developed.

From later results it seems however that solid solutions of  $\text{PuO}_2$  in  $\text{UO}_2$  dissolve like pure  $\text{UO}_2$  but mixtures of  $\text{PuO}_2$  in  $\text{UO}_2$  need HF as catalyst to assure the dissolution of  $\text{PuO}_2$ . It has further more been found that irradiation of the fuel up to 99.000 Mwd/ton has no adverse effect on the dissolution rate of solid solutions of  $\text{UO}_2$  - 20 %  $\text{PuO}_2$ . As for mixtures of  $\text{UO}_2$  - 0,5 %  $\text{PuO}_2$ , irradiation results in an increase in dissolution rate which was attributed to the formation of solid solutions during irradiation. The question whether a fuel mixture of  $\text{UO}_2$  -  $\text{PuO}_2$  can become a real solid solution during irradiation is not answered yet, but this could come in the next few years.

The high  $\text{I}^{131}$  content of the fuel makes it necessary to obtain very high decontamination factors of the process off-gasses for this isotope. The technical difficulties for achieving this have led to envisage a fission gas release treatment whereby iodine, tritium, xenon and krypton are released from the fuel in such a way that they undergo a minimum dilution. Such fission gas release treatment can be an oxidation of the fuel material by which  $\text{UO}_2$  -  $\text{PuO}_2$  is transformed into  $\text{U}_3\text{O}_8$  -  $\text{PuO}_2$ . The change in crystallographic structure of the uranium compound permits the release of trapped fission gasses. It is not yet clear whether during such an oxidation of the fuel material, the dissolution characteristics of  $\text{PuO}_2$  are altered.

Several continuous or semi-continuous dissolver and leacher concepts are under development at the moment. As a number of problems remains to be solved for each of these concepts, additional work is required. The selection between dissolution and leaching will depend to some extent on the outcome of the study on the fuel oxidation process for fission gas release.

Fast breeder fuels irradiated to a burn-up as high as 100.000 Mwd/ton will contain important quantities of fission products such as Mo, Zr, Ph, Rh, Nd, Ru (see Annex IV). Some of these elements will not dissolve in nitric acid, while others have limited solubility in this medium. Calculations show that in the core fuel the quantity of fission products likely to form precipitates during dissolution of the fuel, reaches the ten kgr per ton level. Valuable elements such as Rd & Rh could be recovered the precipitate after clarification of the dissolver solution. Fuel treatment before dissolution could alter the chemical form and dissolution rate of these elements.

Attention should also be paid to iodine present in the dissolution solution, and iodine removal from the solution by such methods as oxidation and air-sparging may be necessary as part of the feed preparation step.

The presence of Macro amounts of Pu in the dissolver solution will require important quantities of reagents to be added to the solution to assure Pu valence adjustment. The reagent used up to now for the small quantities of Pu present in L.W.R. fuels was  $\text{NaNO}_2$ . Research work on the use of  $\text{HNO}_2$  for Pu adjustment has led to introduction of No gas in the solution producing in situ the reduction reagent  $\text{HNO}_2$  (Ref. 11).

The high plutonium content of F.B.R. core fuel ( 20 %) will result in geometric restrictions of the processing equipment for criticality safety. The dissolver unit, specially the batch type, is very critical in this respect as the equipment has to be safe for an heterogeneous as well as for an homogeneous configuration.

The use of soluble neutron poisons to assure criticality safety has been considered for some time, and highly enriched Uranium fuels have been processed using criticality safety control through soluble poison (ref. 12). The neutron poison in the form of a chemical compound is added to the dissolution reagent and ends up with the fission product solution after the fissile materials has been extracted. The generalisation of this technique will mainly depend on its compatibility with the

handling and treatment of the fission product solutions. The use of soluble poisons would permit much higher concentrations of plutonium in the dissolver solution and the extraction feed solution but it does not solve the problem in the following processing steps where plutonium is extracted into an organic solvent.

#### EXTRACTION AND SEPARATION PROCESS

Uranium and plutonium extraction from nitric acid solutions is a well proven process for the processing of thermal reactor fuels. It is generally accepted, that a similar extraction process will be used for fast breeder fuel processing at least in the early period, as the basic separations to be performed are the same as for thermal reactor fuels. The application of T.B.P. extraction to fast breeder fuels requires however careful consideration of the characteristics of these fuels as some of these characteristics may require adaptation of the standard Purex flowsheet conditions.

The Pu/U ratio in F.B. core fuel is of the order of 1/4. Even when considering the mixing of core and breeder pins in the same proportion as they are discharged from the reactor, the average of Pu/U ratio in the dissolution solution will be around 1/20, which is still five to ten times higher than the Pu/U ratio in light water reactor fuels. Most of the data on extraction of Pu and U have been obtained from solutions with Pu concentrations in the range 0,1 % to 1 % of the uranium concentration, it is therefore felt that more extraction data will be needed on solutions containing macro concentrations of Pu.

Uranium-Plutonium separation in thermal reactor fuel is based on the reduction of Plutonium IV to Plutonium III using such reagents as ferrosulfamate, Uranium IV and Hydrazine. For the larger amounts of Plutonium present in F.B. fuels the quantities of reagents to be added will be important because these reduction reactions are being carried out in the presence

of a large excess of reductant, the quantities of reagent to be introduced in the solution may become excessive. In situ electrochemical reduction or reduction with hydrogen in the presence of platinum seem to be valuable alternatives.

Solutions containing high Plutonium concentration show increased tendency to form Plutonium polymers that can not be extracted. The tendency to form polymers increases with decreasing acidity of the solution. This phenomenon will have to be understood well before flowsheet conditions can be established.

The high radiation level of the fast breeder core fuel will result in appreciable degradation of the solvent. The degradation products of T.B.P., dibutyl and monbutylphosphate have not only a negative effect on the decontamination factor but they contribute also to form insoluble organo-metallic compounds with certain elements such as Zr. These compounds tend to accumulate at the interphase in the extraction equipment. This last effect is all the more important as the fission products concentration is high. The accumulated insoluble organo-metallic compounds also act as substrate on which other solid impurities collect. Such unpurity accumulation could seriously disturb the operation of the extraction equipment and adds to the solvent radiation damage.

The higher Plutonium content of the fuels will result in a higher Plutonium loading of the solvent and this in turn will contribute to the solvent radiation damage.

Iodine present in the extraction feed solution accumulates in the solvent, where it contributes in an important way to the solvent radiation damage. The chemical and radiation effects of such accumulation will affect the solvent behaviour and the solvent regeneration.

In order to try to minimize solvent degradation short contact time extraction equipment has been developed and experimented. This equipment, centrifugal contactors, is now

available and will probably replace the usual mixer settlers and pulse columns in those parts where important solvent degradation is to be feared. Short contact time of the aqueous and organic phases will limit the radiation dose absorbed by the solvent as well as the formation of degradation products. The high concentration of fission products in core fuel solution will still tend to form the solid organo-metallic compounds so that the centrifugal contactors could have to handle a certain amount of solids, which may constitute one of the problems in their utilisation.

The increased fission product content of the extraction feed solutions will require a good knowledge of fission product behaviour and their possible inter influence in the extraction process.

The specific extraction and separation problems resulting from high burn-up and short cooling as well as those resulting from the high plutonium content of the core fuel could be overcome by mixing the high burn-up, high plutonium content core fuel with the low burn-up, low Pu content blanket fuel. The principle of mixing core and blanket fuel pins supposes however that the fuel elements are disassembled into single pins and regrouped before cutting which constitutes an additional operation in the head end. Although such an additional operation adds its own particular problem to the whole, the resulting average burn-up and average Pu concentration of the mixed fuel pins (see table 8) is interesting from the point of view of heat output, volatile fission product release during cutting, criticality restrictions as well as for the chemical and operational aspects of the process. It should therefore be considered and evaluated in comparison with the processing of core fuel alone.

TABLE 8

AVERAGE BURN-UP SPECIFIC POWER AND Pu CONTENT OF MIXTURES OF FUEL PINS AS THEY ARE DISCHARGED FROM Na-1

	Core + axial blanket Zones 1-2-3-4	Core + axial + radial blanket	
		Zones 1-2-3-4-5	Zones 1-2-3-4-5-6
Average burn-up $10^3$ Mwd/T.	~ 45	~ 24	~ 18
Average specific power Kw/Kg	75		35
Average PuO <sub>2</sub> content % of total oxide	~ 11,5	~ 7	~ 5,5

The final plutonium purification requires the handling of considerable quantities of plutonium and the criticality problems involved in this part of the reprocessing process are at their highest. The amounts of plutonium to be handled appear to be around 50 kg/ton of mixed core and blanket fuel.

Losses of plutonium experienced in light water reactor fuel reprocessing have frequently been in the order of 2%. For a small plutonium through-put these losses may seem acceptable. In the case of plutonium concentrations in the fuel of some 5% such losses represent about 1 Kg of plutonium per ton of fuel processed. Most of the lost plutonium appears in the waste streams. Apart from the loss of financial value the accumulation of important quantities of Plutonium in the fission product solutions may raise criticality problems in the storage of high level waste solutions. It is evident that an important reduction in the losses of plutonium (measured and unmeasured) will be required before big scale reprocessing of fast breeder fuel can be undertaken.

The usual anion resin system and the more recent amine extraction system have been developed to handle the small quantities of plutonium resulting from thermal reactor fuel processing. The application of these processes in handling great quantities of plutonium from fast breeder fuels will require adaptation and adjustment.

The high  $\alpha$  activity of F.B.R. plutonium and the associated neutron generation through ( $\alpha ; \gamma$ ) reaction on oxygen could lead to remote fabrication of the fuels. Such remote fabrication may further result in less stringent requirements for fission product decontamination and affect in this way the whole reprocessing process.

#### GASEOUS EFFLUENTS

The gaseous fission product content in fast breeder core fuel is much higher than in the light water reactor fuel as a result of higher burn-up, higher specific power and shorter cooling time.

The isotopes of primary importance are  $I^{131}$  ( $t_{1/2}$  8,05 d)  $Xe^{133}$  ( $t_{1/2}$  = 5,3 d)  $Kr^{85}$  ( $t_{1/2}$  = 10.8 y) and  $H_3$  ( $t_{1/2}$  = 12,3 y).

The concentration of these isotopes in the core fuel can be expressed as follows : (ref. 13)

$Kr^{85}$	: 0,3 Ci/Mwd burn-up
$H^3$	: 0,025 Ci/Mwd burn-up
$Xe^{133}$	: 1.200 Ci/Mw specific power )
$I^{131}$	: 2.200 Ci/Mw specific power ) after 30 days cooling time

#### - IODINE

A comparison of the calculated  $I^{131}$  content in light water reactor fuels and fast breeder reactor core fuels given in table 9 shows the importance of the difference between both types of fuels.

TABLE 9

I<sup>131</sup> CONTENT OF FUELS IN Ci/T

	<u>L.W.R. fuels</u>		Core fuels			<u>F.B.R. Na-1</u> Mixed core + axial blanket Zones 1 + 2 + 3 + 4			Mixed core + axial + radial blanket Zones 1 + 2 + 3 + 4 + 5 + 6		
Durn-up: Mwd/ton	25.000		80.000			45.000			18.000		
Specific power : Kn/Kg	15		150			75			35		
Cooling time : days	30	150	30	60	90	30	60	90	30	60	90
I <sup>131</sup> ac- tivity Ci/Ton	$3,3 \cdot 10^4$	1,0	$3,3 \cdot 10^5$	$2 \cdot 10^4$	$1,6 \cdot 10^3$	$1,6 \cdot 10^5$	$1,10^4$	$8 \cdot 10^2$	$8 \cdot 10^4$	$5 \cdot 10^3$	$4 \cdot 10^2$

In the case of fast breeder fuels, cooling times as short as 30 and 60 days are envisaged, this allows only a very limited reduction of the  $I^{131}$  content which is for the F.B.R. core fuels 10 times higher than in light water reactor fuels at the moment of discharge of the fuel. The overall Iodine decontamination factor to be obtained is dependent on the allowed continuous release limit which is in turn dependent on the stack dilution factor.

As an example under conditions of an average dilution factor of  $7 \cdot 10^6 \text{ m}^3/\text{sec}$  (case of Eurochemic) a release of 0,1 Ci/day of  $I^{131}$  would not result in a breathing air concentration above the MPC (population) corrected by the factor 1/700 for intake by the milk pathway ( $1,4 \cdot 10^{-13} \text{ Ci/m}^3$ ).

On the basis of a release of 0,1 Ci  $I^{131}$ /day the decontamination factor to be obtained has been derived for L.W.R. fuels, F.B.R. core fuels and mixtures of F.B.R. core and blanket fuel (Table 10).

TABLE 10

DECONTAMINATION FACTORS FOR  $I^{131}$  TO BE OBTAINED FOR LWR FUEL AND FBR FUEL

release of 0,1 Ci  $I^{131}$ /day

Plant capacity	1 ton/day				5 Tons/day			
	30 d.	60 d.	90 d	150 d	30 d	60 d	90 d	150 d
L.W.R. fuels	$3,3 \cdot 10^5$			10	$1,6 \cdot 10^6$			50
F.B.R. core fuel	$3,3 \cdot 10^6$	$2 \cdot 10^5$	$1,6 \cdot 10^4$		$1,6 \cdot 10^7$	$1 \cdot 10^6$	$8 \cdot 10^4$	
F.B.R. core + axial blanket Zones 1+2+3+4	$1,6 \cdot 10^6$	$1 \cdot 10^5$	$8 \cdot 10^3$		$8 \cdot 10^6$	$5 \cdot 10^5$	$4 \cdot 10^4$	
F.B.R. mixed core & Blanket fuel (Reactor Na-1) Zones 1+2+3+4 +5+6	$8 \cdot 10^5$	$5 \cdot 10^4$	$4 \cdot 10^3$		$4 \cdot 10^6$	$2,5 \cdot 10^5$	$2 \cdot 10^4$	

From published information it can be concluded that decontamination factors for  $I^{131}$  of  $10^2$  to  $10^3$  have been achieved in presently operating reprocessing plants.

The iodine removal methods in use are mainly : scrubbers using sodium hydroxide with or without sodiumthiosulfate addition to the scrub solution and silver reactors in which Iodine is fixed on a packing coated with Silver Nitrate. Adsorption on activated charcoal and molecular sieves has been extensively studied and is still being tested to clarify the efficiency in the presence of other gases such as oxygen and oxides of nitrogen.

From Table 10 it is clear that  $I^{131}$  release limits can be rather easily met for 150 days cooled light water reactor fuels even in the case of processing plants of 5 to 10 tons/day throughput. In the case of short cooled (30 or 60 days) F.B.R. fuels the iodine content is so high that decontamination factors in the order of  $10^5$  and  $10^6$  would have to be reached for core fuels. We can expect that the higher iodine content in the off-gas and a less diluted form of the off-gas stream will result in an increased overall efficiency of the removal methods. Nevertheless the required decontamination factors will only be achieved through improvement and combination of existing methods or the development of new methods. It is to be noted that processing mixed core and blanket fuels would result in lower average iodine release over short periods and would distribute more evenly the release of the total iodine contained in a batch of discharged Fast Breeder fuel.

It should also be considered that gaseous fission products in general and iodine in particular are released from the fuel at different stages of the processing line.

The pressure in the fuel tubes and the temperature at which short cooled fast breeder fuels will have to be handled indicates that an important part of the gases will be released when the fuel tubes are opened. When released from the fuel the iodine and other gaseous fission products will be diluted in the gas constituting the cell or equipment atmosphere. The cell or equipment volume in which the release occurs constitutes an important factor in the dimensioning of the iodine removal equipment.

A second part of the iodine content will be released during fuel dissolution and will follow the dissolution off-gas which will have to be treated for iodine removal. The dissolution off-gas contains important quantities of Nitrogen oxides which are recombined to form nitric acid. In the process part of the Iodine is also returned to the solution. After recombination of nitric acid the off-gas stream contains still small quantities of nitrogen oxides as well as the excess oxygen or air from the acid recombination. The presence of nitrogen oxides and the dilution obtained by the excess air or oxygen do not simplify the Iodine retention.

In order to assure maximum decontamination it would be advantageous to release all or as much as possible of the fission gases in one and the same step, preferably before dissolution of the fuel as this would avoid to mix the fission gases with nitrogen oxides from the dissolution off-gas. These considerations lead to a reevaluation of the existing head end treatments and possibly to the incorporation of a fission gas release treatment before dissolution of the fuel.

#### NOBLES GASES

The noble gas fission products  $Kr^{85}$  and  $Xe^{133}$  have up to now been released to the atmosphere as the environmental concentration resulting from stack dilution remains well below the permissible concentration. In the case of fast breeder core fuels the concentration of  $Kr^{85}$  and  $Xe^{133}$  are much higher than in light water reactor fuels. A comparison of these concentrations is given in Table 11.

Considering an atmospheric dilution factor of  $1.10^6 \text{ m}^3/\text{sec}$  the calculation shows that the release of the total content of  $Kr^{85}$  and  $Xe^{133}$  of 60 days cooled fuel, processed at a rate of 5 T/day is still feasible without any special arrangements.

Table 11

Kr<sup>85</sup> and Xe<sup>133</sup> ACTIVITY IN LIGHT WATER REACTOR AND FAST BREEDER REACTOR FUELS

	<u>L.W.R. fuels</u>		<u>F.B.R.</u>								
			Core fuels			Mixed core + axial blanket Zones 1 + 2 + 3 + 4			Mixed core + axial + radial blanket Zones 1 + 2 + 3 + 4 + 5 + 6		
Burn-up (Mwd/Ton)	25.000		80.000			45.000			18.000		
Specific power (Kw/Kg)	15		150			av.75			av.35		
Cooling time (d)	30	150	30	60	90	30	60	90	30	60	90
Kr <sup>85</sup> con- tent (Ci/T)	$7,5 \cdot 10^3$	$7,5 \cdot 10^3$	$2,4 \cdot 10^4$	$2,4 \cdot 10^4$	$2,4 \cdot 10^4$	$1,3 \cdot 10^4$	$1,3 \cdot 10^4$	$1,3 \cdot 10^4$	$5,4 \cdot 10^3$	$5,4 \cdot 10^3$	$5,4 \cdot 10^3$
Xe <sup>133</sup> content (Ci/T)	1,8.10	< 1	$1,8 \cdot 10^5$	$3 \cdot 10^3$	$6 \cdot 10^1$	$9 \cdot 10^4$	$1,5 \cdot 10^3$	$,3 \cdot 10^1$	$4,2 \cdot 10^4$	$7 \cdot 10^2$	1,5.10

ENVIRONMENTAL  $\text{Xe}^{133}$  AND  $\text{Kr}^{85}$  CONCENTRATIONS RESULTING FROM PROCESSING OF DIFFERENT TYPES OF FUELS UNDER -  
 CONDITIONS OF TOTAL RELEASE

Basis : figures of table 11

atmospheric dilution factor  $7 \cdot 10^6 \text{ m}^3/\text{sec}$

	Plant capacity	LWR fuels cooling time 150 d	F.B.R. Core fuel Cooling time			F.B.R. Core + axial blanket Zones 1 + 2 + 3 + 4 Cooling time			Mixed Core & total blanket Cooling time fuel		
			30 d	60 d	90 d	30 d	60 d	90 d	30 d	60 d	90 d
$\text{Kr}^{85}$ $\text{Ci}/\text{m}^3$	1 T/day	$1,3 \cdot 10^{-8}$	$4 \cdot 10^{-8}$	$4 \cdot 10^{-8}$	$4 \cdot 10^{-8}$	$2 \cdot 10^{-8}$	$2 \cdot 10^{-8}$	$2 \cdot 10^{-8}$	$9 \cdot 10^{-9}$	$9 \cdot 10^{-9}$	$9 \cdot 10^{-9}$
	5 T/day	$6,5 \cdot 10^{-8}$	$2 \cdot 10^{-7}$	$2 \cdot 10^{-7}$	$2 \cdot 10^{-7}$	$1 \cdot 10^{-7}$	$1 \cdot 10^{-7}$	$1 \cdot 10^{-7}$	$5 \cdot 10^{-8}$	$5 \cdot 10^{-8}$	$5 \cdot 10^{-8}$
$\text{Xe}^{133}$ $\text{Ci}/\text{m}^3$	1 T/day	$< 10^{-11}$	$3 \cdot 10^{-7}$	$5 \cdot 10^{-9}$	$1 \cdot 10^{-10}$	$1,5 \cdot 10^{-7}$	$2,5 \cdot 10^{-9}$	$5 \cdot 10^{-11}$	$7 \cdot 10^{-8}$	$1 \cdot 10^{-8}$	$2,5 \cdot 10^{-11}$
	5 T/day	$< 10^{-11}$	$1,5 \cdot 10^{-6}$	$2,5 \cdot 10^{-8}$	$5 \cdot 10^{-10}$	$6,5 \cdot 10^{-7}$	$1,2 \cdot 10^{-8}$	$2,5 \cdot 10^{-10}$	$3,5 \cdot 10^{-7}$	$5,10^{-7}$	$1,2 \cdot 10^{-10}$

## WASTES

### Low activity effluents

The radioactive waste solutions produced during the aqueous processing of irradiated fuel constitute an important volume of liquids. As the fission products have to be stored, be it under liquid or solid form, there is a distinct interest in concentrating the waste solutions. The concentration steps produce highly active concentrate to be stored and low active effluent which can be released to the environment. The concentrate retains practically all of the fission product elements except Tritium. Tritium appears as Tritiated water in the aqueous solutions throughout the process. It distributes during the waste concentration steps over the low activity effluent and the concentrate. The low activity effluent, after eventual further treatment to decrease its activity content, is released to the environment.

In actual practice with L.W.R. fuels of up to 20.000 Mwd/Ton burn-up, the low activity effluent volume represents approximately 95% of the total waste volume, so that finally about 95% of all tritium generated is released to the environment as tritiated water. As a result of the rather long halflife of Tritium (12,3 year) its content in irradiated fuel is directly proportional to the burn-up of the fuel and is not influenced by cooling. A thermal fission yield of  $\sim 1,10^{-4} \text{ H}^3$  atoms per fission for  $\text{U}^{235}$  and a fast fission yield of  $\sim 2,5 \cdot 10^{-4} \text{ H}^3$  atoms per fission for  $\text{Pu}^{239}$ , are cited in the literature (ref.14). Tritium formation in the fuel takes also place through an  $(n; \alpha)$  reaction with  $\text{Li}^6$  [ $\text{Li}_3^6 (n; \alpha) \text{H}_1^3$ ]  $\text{Li}^6$  being present in the fuel as an impurity (ref. 14; 15).

In the case of thermal neutron irradiation the Tritium production through  $n; \alpha$  reaction with  $\text{Li}^6$  is quoted to be approximately twice as high as through fission. The Tritium content of L.W.R.fuels with 20.000 Mwd/Ton burn-up is considered to be around 700 Ci/Ton (ref. 14).

Other sources evaluate the Tritium content of L.W.R. fuels at  $\sim 300 \text{ Ci-/Ton}$  (ref. 15), neglecting the Tritium generation through activation of  $\text{Li}^6$  as the concentration of the  $\text{Li}^6$  impurity in the fuel is uncertain.

For F.B.R. fuel irradiated to 100.000 Mwd/Ton the Tritium content is calculated to be around 3.300 Ci/Ton. In this case practically all of the Tritium is generated through fission, as the cross section for the  $(n;\alpha)$  reaction with  $\text{Li}^6$  impurities is rather low for fast neutrons.

Considering that fuel discharged from a F.B.R. (example Na-1) is approximately 1/5 core fuel and 4/5 blanket fuel, the average Tritium content can be estimated to be around 600 to 700 Ci/Ton. This content of Tritium is very much comparable to the Tritium content of 700 Ci/Ton for L.W.R. fuels irradiated to 20.000 Mwd/Ton.

The quantities of Tritium indicated above are to be released to the environment or to be stored.

In case of release to the environment two possibilities are to be considered :

- release to surface waters in which case the dilution at the release point has to reduce the concentration of Tritium below the MPC for the population ( $3 \cdot 10^{-3}$  Ci/ml) such a dilution would require an important water flow to handle the Tritium release from a fuel processing plant handling one ton/day core fuel or 5 tons/day mixed core and blanket fuel.
- dispersion into the atmosphere is another method for Tritium release which has been put forward. It consists of evaporating the Tritium containing water and evacuating it through the stack. This method takes advantage of the greater dispersion coefficient, obtained through stack release at a certain height.

Lately consideration has been given in the American literature to the storage of Tritium. This method of Tritium handling could be of interest when the fuel is subjected to a fission gas release treatment. The gaseous fission products could then be collected in a relatively small volume from which Tritium could be separated.

The public acceptance of nuclear energy is often related to its environmental aspects. Hence it might be worthwhile to consider the release concept "as low as practical" for reprocessing plants as well as for reactors.

Such release concept for the gaseous wastes implies the liberation and collection of gaseous fission products from the fuel before dissolution. For the aqueous wastes it would require the thorough decontamination of the low level wastes to be released or the recycling of distillates and possibly other low level wastes as make up water for fresh reagent solutions.

#### High level wastes

The concentration factor which can be reached in the concentration step of the fission product solutions is limited by the content of dissolved salts and activity of fission products. Considering that dissolved salts content is low and does not limit the concentration factor, the maximum activity of the concentrate is usually limited to around 5.000 Ci/l for reasons of decay heat evacuation during storage of the concentrates.

The long lived fission product ( $T_{1/2} > 1$  year) content of irradiated fuel is directly proportional to the burn-up. Considering that the fast fission yield of  $\text{Pu}^{239}$  and the thermal fission yield of  $\text{U}^{235}$  are not so much different on the whole, the fast breeder core fuel of 30.000 Mwd/Ton would have a 4 times higher long lived fission product content. Mixed core and blanket fuel (1/5 core - 4/5 blanket) will have approximately the same average content of long lived fission product as light water reactor fuels irradiated to 20.000 Mwd/Ton.

As fast breeder fuel will have to be processed at short cooling times there will be an important contribution of fission products of intermediate half lives (a few weeks). The production of these fission products is proportional to the specific power. For fast breeder fuel the specific power in the core is 10 times higher than the specific power for L.W. reactors, consequently fast breeder core fuel will have a ten times higher content of these fission products than L.W.R. fuel of the same cooling time. Mixing of core and blanket fuel will reduce this by a factor of 4 to 5.

The activity of the specific nuclides as well as the total activity of the fuel as function of the cooling time is presented in annex IV. The data indicate clearly that the volume reduction of fission product solutions resulting from processing of short cooled fuel will be limited by the activity contained and the decay heat generated.

Economic studies on waste disposal have indicated the advantage of liquid storage for a period of three to five years before application of solidification treatment and final disposal. The economic advantage of interim storage stems from reduction of activity and heat output which allows a greater volume of liquid waste to be incorporated per unit volume of solidified product (f.i. glass).

The interim storage being an economic advantage for fission product solutions from L.W.R. fuel that were cooled for about 150 days before processing, the advantage will even be greater for less cooled fast breeder fuels. Depending on the effective cooling time before processing it will have to be examined what could be gained by reconcentrating the wastes after a first decay storage.

A unique aspect of fast breeder core fuel is its high content of noble metal fission products. The calculated content fission products for 80.000 Mwd/Ton irradiated core fuel (annex IV) shows that kilogram per ton quantities of Rhodium and Palladium are present. It is to be expected that the recuperation of these elements will become economically interesting. Although the chemical behaviour of these elements under the conditions of fuel dissolution has not been studied specifically, the major part of these elements can be expected to remain in the dissolver precipitate formed during dissolution of the fuel. They would then be separated from the fuel solution in the feed clarification step, which should allow the eventual recuperation of Rh & Pd to start from a minimum volume and a reduced amount of other activity.

## Conclusion

The aqueous fuel reprocessing technique is at this stage the most probable choice for the processing of the fast breeder fuel because of experience gained with this technique in the processing of oxide fuel from thermal reactors.

The process will however need to be adapted to the characteristics of the fuel and at some stages of the process new process steps may even be needed.

Three characteristics of the fast breeder core fuel are at the basis of the arising major problems : the plutonium content; the burn up and cooling time; the presence of sodium on the fuel elements.

The high plutonium content of the fast breeder core fuels results in increased criticality risks, which will lead to geometry limitations and operational restrictions in reprocessing.

The burn up and cooling time of the fuel before processing are the important parameters determining the heat generation and the gaseous fission product content of the fuel. The processing of short cooled fuels ( 30 to 60 days) as proposed to day will require adaptation of existing techniques or new solutions for a number of problems such as : heat evacuation during transport storage and mechanical processing, gaseous fission product release and off gas treatment.

The presence of adhering sodium on the fuel elements will require the introduction of a sodium removal step before the fuel elements can be transferred to an aqueous cooling medium

All of the above mentioned major problems are examined and studied at the various laboratories and organisations concerned with these problems.

The fuel cycle cost for fast breeder reactors will depend to a great extent on the solutions developed for the various problems, and only when satisfactory solutions will exist, will it be possible to evaluate with some precision the cost of fast breeder fuel processing and the fuel cycle cost as a whole.

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ANNEXE

CHARACTERISTICS OF THE FAST BREEDER REACTOR

REFERENCE FUEL

- I. Characteristics of the reference fuel.
- II. Characteristics of the reference fast breeder reactor fuel assembly.
- III. Comparison AL fuel - Na1 fuel.
- IV. Decay heat of the reference fast breeder reactor fuel.
- V. Fission product content of the reference fast breeder reactor fuel.

Annexe I.

CHARACTERISTICS OF THE REFERENCE FUEL.

For the study of the reprocessing of fast breeder reactor fuel element, the core fuel of a 1,000 MWe power reactor has been taken as reference.

The comparison of different fast breeder power reactor projects and the progress in the development work actually undertaken permit to define the characteristics of the reference fuel as follows :

- Burn-up	85,000 MWD/t
- Specific power	146 kW/kg
- Nature of the fuel	pellets ( $UO_2$ - $PuO_2$ )
- Initial Pu content (Pu/U + Pu)	18,1 w/o
- Initial composition of the Pu	63/30/5/2 w/o
- Pu content after irradiation	18,9 w/o
- Composition of the Pu after irradiation	60,5/30,65/6,35/2,5 w/o
- Residence time	730 days
- Charge factor	0,8

Annexe II.

CHARACTERISTICS OF THE REFERENCE FAST BREEDER REACTOR FUEL ASSEMBLY.

The reference fast breeder reactor fuel assembly is the core assembly of the Na-1 1,000 MWe fast breeder reactor.

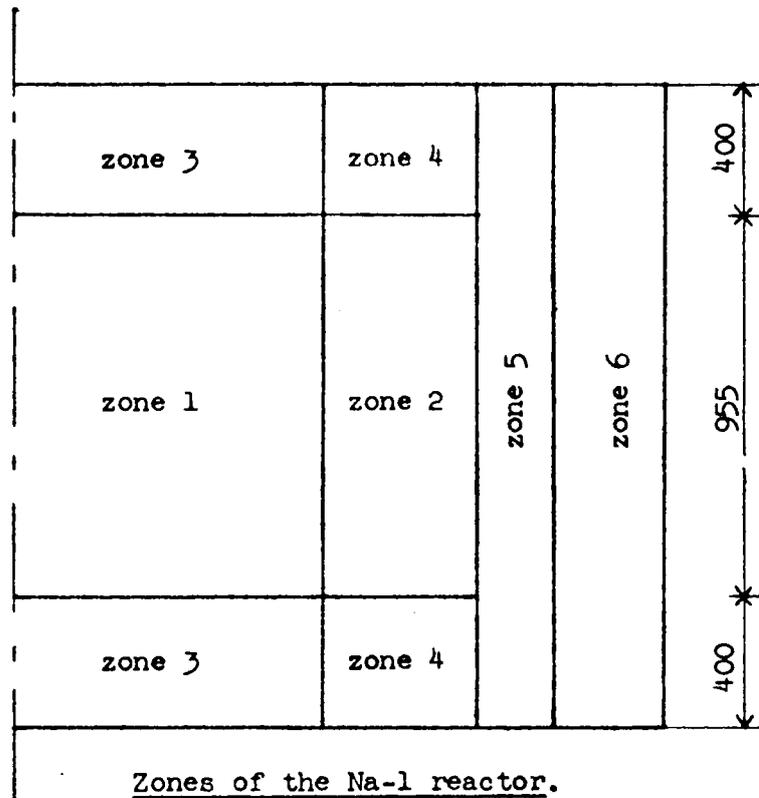
This assembly includes the core fuel and the axial blanket fuel. It consists of 331 fuel pins fixed at support bars. Spacer grids are foreseen every        mm.

The fuel pins are enveloped in a hexagonal closed wrapper.

- Number of fuel pins/assembly	331
- Weight of the assembly	315 kg
. weight of core fuel (zone 1)	85.5 kg
. weight of axial blanket fuel (zone 3)	73.8 kg
. weight of the canning/assembly	52.3 kg
. weight of the wrapper	103.5 kg
- Weight of one fuel pin (core and axial blanket)	639 g
. weight of core fuel	258.3 g
. weight of axial blanket fuel	223 g
. weight of the canning	158 g.

The Na-1 reactor is subdivided in six different zones :

- zone 1 : core
- zone 2 : core
- zone 3 : axial blanket
- zone 4 : axial blanket
- zone 5 : radial blanket
- zone 6 : radial blanket.



The specific power of the different zones is :

- zone 2 : 115 kW/kg (residence time : 730 d)
- zone 3 : 6.5 kW/kg (residence time : 730 d)
- zone 4 : 3.3 kW/kg (residence time : 730 d)
- zone 5 : 10.3 kW/kg (residence time : 600 d)
- zone 6 : 3.75 kW/kg (residence time : 1,800 d).

Annexe III.

COMPARISON OF THE REFERENCE FUEL CHARACTERISTICS FOR ATOMICS  
INTERNATIONAL F.B.R. OXIDE FUEL AND Na-1 F.B.R. OXIDE FUEL

	A.I.*	Na 1**
<u>Assembly</u>		
- length	5380 mm	790 mm
- wrapper	hexagonal	hexagonal
- material wrapper	AISI 304	
- thickness wrapper material	3,3 mm	4 mm
- hexagone size	135 mm	178,5 mm
<u>Fuel rod</u>		
- total length	3660 mm	2672 mm
- fuel hight core	1220 mm	955 mm
blanket	2 x 305 mm	2 x 400 mm
gas plenum <sup>L</sup>	1755 mm	800 mm
- diameter (O.D.)	6,35 mm	6,7 mm
- thickness of tube	0,38 mm	0,35 mm
- material	AISI 304	AISI 304
<u>Oxides</u>		
- quantity/assembly		
core	60,8 kg	85,5 kg
blanket	30,4 kg	73,7 kg
total	91,2 kg	159,1 kg
- quantity/rod		
core	280 gr	258,3 gr
blanket	140 gr	223,- gr
total	420 gr	481,3 gr
<u>Burn-up (core fuel)</u>	80.000 MWD/T	85.000 MWD/T
<u>Irradiation time (core and axial blanket)</u>	540 D	584 D
<u>Specific power (core fuel)</u>	148,15 kw/kg	146 kw/kg

\* A.I : Atomics International Reference Oxide Fuel, NAA-SR-MEMO-12604 (jan. 1968).

\*\* Na 1 : Sodium cooled - fast breeder reactor N° 1 (Germany)

Annexe IV.

DECAY HEAT OF THE REFERENCE FAST BREEDER REACTOR FUEL (Na-1)  
AS A FUNCTION OF THE COOLING TIME.

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The decay heat of the reference fuel has been estimated following the results of K. Shure (AA).

The mean decay heat for mixtures of fuel ( $C_{14}$  -  $C_{16}$  ...) (A) are calculated for mixtures of fuel quantities proportional to the discharged quantities from each zone :

- zone 1 : 1/3 per period (= 730 days)
- zone 2 : 1/3 per period "
- zone 3 : 1/3 per period "
- zone 4 : 1/3 per period "
- zone 5 : 1/3 per period "
- zone 6 : 1/6 per period. (= 1,800 days)

- 
- (A)  $C_1 + 3$  mixture of fuel contained in the core 1 pins,  
 $C_2 + 4$  mixture of fuel contained in the core 2 pins,  
 $C_{14}$  mixture of fuel from zones 1 to 4,  
 $C_{16}$  mixture of fuel from zones 1 to 6,  
 $C_5 + 6$  mixture of fuel from zones 5 and 6.

- (AA) Note BN n° 308.09/177/ni/065.  
K. Shure,  
Fission Product Decay Energy, Bettis Technical Review,  
WAPD - BT - December 24 1961, pages 1 - 17.

DECAY HEAT OF THE REFERENCE FAST BREEDER REACTOR FUEL (Na-1).

per kg (UO<sub>2</sub> - PuO<sub>2</sub>)

	Unit	Decay time - days							
		10	20	30	60	90	120	150	180
Reference fuel (core fuel zone 1)	W/kg	320	247	197	139	104	85	70	60
Axial blanket fuel (zone 3)	W/kg	14.2	10.9	8.8	6.2	4.6	3.8	3.1	2.7
Core fuel (zone 2)	W/kg	251.8	194.3	155.2	109.2	81.6	66.7	55.2	47.1
Axial blanket fuel (zone 4)	W/kg	7.2	5.6	4.5	3.1	2.3	1.9	1.6	1.4
Radial blanket fuel (zone 5)	W/kg	22.2	17.1	13.6	9.5	7	5.8	4.7	4
Radial blanket fuel (zone 6)	W/kg	8.6	6.7	5.4	3.9	3	2.5	2.1	1.9
Fuel mixture C <sub>14</sub>	W/kg	153.2	118.2	94.4	66.5	49.7	40.7	33.5	28.7
Fuel mixture C <sub>16</sub>	W/kg	79.9	61.6	49.2	34.7	25.9	21.2	17.5	15

DECAY HEAT OF THE REFERENCE FAST BREEDER REACTOR FUEL (Na-1).

per kg ( $UO_2 - PuO_2$ )

	Unit	Decay time - days o							
		10	20	30	60	90	120	150	180
Fuel mixture $C_5 + 6$	W/kg	15.14	11.7	9.34	6.59	4.92	4.08	3.35	2.91
Fuel mixture $C_1 + 3$	W/kg	178	137.5	110	77.4	57.7	47.2	39	33.3
Fuel mixture $C_2 + 4$	W/kg	139.5	107	85.6	60.2	45	36.8	30.5	26

DECAY HEAT OF THE REFERENCE FAST BREEDER REACTOR FUEL

PER ASSEMBLY AND FUEL PIN.

	Unit	Decay time - days							
		10	20	30	60	90	120	150	180
Assembly zone 1 + 3	kW/ass.	28,39	21.9	17.5	12,31	9.2	7.52	6.22	5,31
Assembly zone 2 + 4	kW/ass.	22,22	17.07	13,63	9.59	7,17	5.86	4,85	4,14
Assembly zone 5	kW/ass.	6,48	4.98	3.96	2.76	2,04	1,68	1,38	1,17
Assembly zone 6	kW/ass.	2.99	2.34	1.88	1.37	1.04	0.87	0.73	0.65
Fuel pin zone 1 + 3	W/pin	85.8	66.2	52.8	36.7	27.7	22.74	18,79	16.19
Fuel pin zone 1	W/zone	82.6	63.7	50.9	35.3	26.7	21.9	18.1	15.6
Fuel pin zone 3	W/zone	3,2	2.5	1.9	1.4	1.0	0.84	0.69	0.59
Fuel pin zone 2 + 4	W/pin	66.81	51.54	41.19	29.0	21.7	17.73	14.65	12,5
Fuel pin zone 2	W/zone	65.2	50.3	40.2	28.3	21.2	17.3	14.3	12.2
Fuel pin zone 4	W/zone	1,61	1,24	0.99	0.70	0.50	0,43	0.35	0.3
fuel pin zone 5	W/pin	71.2	54.7	43.5	30.3	22.4	18.5	15.2	12.8
fuel pin zone 6	W/pin	49.1	38.4	30.9	22.4	17.1	14.3	11.9	10.7

Annexe V

FISSION PRODUCT CONTENT OF THE IRRADIATED REFERENCE FUEL -

FAST BREEDER REACTOR Na 1

The fission product content of the reference fuel has been determined following the calculations of D. Gupta\*

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\* D. Gupta, F. Heller, R. Schröder  
The amounts of Fission Products Nuclides Produced in Pu<sup>239</sup> -  
Fuelled Fast Reactor and the Delayed Heat Generation after Shut-  
Down,  
KFK 722, November 1968.

Note BN n° 712.20/380/n/002, F. Servais.

FISISON PRODUCTS CONTENT OF THE IRRADIATED REFERENCE FUEL  
gr /kg of FRESH FUEL

ISOTOPE	30 d.		60 d.		90 d.		120 d.		150 d.		180 d.	
	gr/kg	Ci/kg	gr/kg	Ci/kg	gr/kg	Ci/kg	gr/kg	Ci/kg	gr/kg	Ci/kg	gr/kg	Ci/kg
H <sub>2</sub>		3.3		3.3		3.3		3.3		3.3		3.3
Rr 81	0.0640		0.0640		0.0640		0.0640		0.0640		0.0640	
Se 82	0.0080		0.0080		0.0080		0.0080		0.0080		0.0080	
Kr 85	0.1970	7.73.10 <sup>1</sup>	0.1962	7.691.10 <sup>1</sup>	0.1952	7.651.10 <sup>1</sup>	0.1942	7.612.10 <sup>1</sup>	0.1931	7.560.10 <sup>1</sup>	0.1918	7.518.10 <sup>1</sup>
Kr 83												
Kr 84	0.5867		0.5867		0.5867		0.5867		0.5867		0.5867	
Kr 86												
Rb 85	0.1970		0.197		0.197		0.197		0.197		0.197	
Rb 87	0.3330		0.383		0.383		0.383		0.383		0.383	
Sr 88	0.5698		0.5698		0.5698		0.5698		0.5698		0.5698	
Sr 89	0.0552	1.6.10 <sup>3</sup>	0.0376	1.065.10 <sup>3</sup>	0.0241	0.701.10 <sup>3</sup>	0.0161	0.468.10 <sup>3</sup>	0.0106	0.308.10 <sup>3</sup>	0.0070	0.203.10 <sup>3</sup>
Sr 90	0.8992	1.27.10 <sup>2</sup>	0.8974	1.274.10 <sup>2</sup>	0.8954	1.271.10 <sup>2</sup>	0.8936	1.268.10 <sup>2</sup>	0.8919	1.266.10 <sup>2</sup>	0.8899	1.263.10 <sup>2</sup>
Y 89	0.6380		0.6569		0.6708		0.6775		0.6828		0.6874	
Y 91	0.1041	2.62.10 <sup>3</sup>	0.0728	1.834.10 <sup>3</sup>	0.0509	1.282.10 <sup>3</sup>	0.0356	0.871.10 <sup>3</sup>	0.0247	0.622.10 <sup>3</sup>	0.0174	0.438.10 <sup>3</sup>
Zr 93	1.3460	0.003	1.3460	0.003	1.346	0.003	1.346	0.003	1.346	0.003	1.346	0.003
Zr 95	0.2432	5.1.10 <sup>3</sup>	0.1770	3.717.10 <sup>3</sup>	0.1285	2.698.10 <sup>3</sup>	0.0934	1.961.10 <sup>3</sup>	0.0679	1.425.10 <sup>3</sup>	0.0494	1.037.10 <sup>3</sup>
Zr 90												
Zr 91												
Zr 92	6.8192		6.8315		6.8731		6.8883		6.8989		6.9062	
Zr 94												
Zr 96												
Nb 95	0.1684	6.63.10 <sup>3</sup>	0.1400	5.516.10 <sup>3</sup>	0.1107	4.361.10 <sup>3</sup>	0.0890	3.506.10 <sup>3</sup>	0.0691	2.722.10 <sup>3</sup>	0.0509	2.005.10 <sup>3</sup>
Mo 95												
Mo 96												
Mo 97												
Mo 98	9.9907		10.0855		10.1720		10.2293		10.2727		10.3083	
Mo 100												

FISSION PRODUCTS CONTENT OF THE IRRADIATED REFERENCE FUEL

gr /kg of FRESH FUEL

ISOTOPE	30 d.		60 d.		90 d.		120 d.		150 d.		180 d.	
	gr/kg	C1/kg										
Tc 99	2.4244	0.041	2.4244	0.041	2.4244	0.041	2.4244	0.041	2.4244	0.041	2.4244	0.041
Ru 100	8.3366		8.3366		8.3366		8.3366		8.3366		8.3366	
Ru 101												
Ru 102												
Ru 104												
Ru 103	0.1502	$4.791 \cdot 10^3$	0.0894	$2.851 \cdot 10^3$	0.0532	$1.697 \cdot 10^3$	0.0317	$1.011 \cdot 10^3$	0.0188	$0.599 \cdot 10^3$	0.0111	$0.354 \cdot 10^3$
Ru 106	1.2502	$4.175 \cdot 10^3$	1.1967	$3.996 \cdot 10^3$	1.1266	$3.762 \cdot 10^3$	1.060	$3.540 \cdot 10^3$	0.9949	$3.322 \cdot 10^3$	0.9357	$3.125 \cdot 10^3$
Rh 103	2.4255		2.4225		2.5207		2.5436		2.5537		2.5588	
Pd 104	5.8278		5.8957		5.9708		6.034		6.0882		6.1414	
Pd 105												
Pd 106												
Pd 107												
Pd 108												
Pd 110												
Ag 109	0.6802		0.6802		0.6802		0.6802		0.6802		0.6802	
Ag 111	0.0089	$1.388 \cdot 10^3$										
Cd 110	0.2573		0.2573		0.2573		0.2573		0.2573		0.2573	
Cd 111												
Cd 112												
Cd 114												
Cd 113	0.0319	7.368	0.0318	7.345	0.0317	7.322	0.0317	7.322	0.0314	7.253	0.0313	7.230
Cd 115	0.0011	$2.882 \cdot 10^1$	0.0006	$1.572 \cdot 10^1$	0.0003	$0.786 \cdot 10^1$	0.0002	$0.524 \cdot 10^1$	0.0001	$0.262 \cdot 10^1$		

FISSION PRODUCTS CONTENT OF THE IRRADIATED REFERENCE FUEL

gr/kg of FRESH FUEL

ISOTOPE	30 d.		60 d.		90 d.		120 d.		150 d.		180 d.	
	gr/kg	Ci/kg										
In 115	0.0164		0.0168		0.0171		0.0173		0.0174		0.0176	
Sn 123	0.0272	$2.216 \cdot 10^2$	0.0232	$1.890 \cdot 10^2$	0.0197	$1.605 \cdot 10^2$	0.0167	$1.361 \cdot 10^2$	0.0142	$1.157 \cdot 10^2$	0.0121	$0.986 \cdot 10^2$
Sn 125	0.0055	$0.050 \cdot 10^2$	0.0007	$0.770 \cdot 10^2$	0.0001	$0.110 \cdot 10^2$						
Te 129	0.0345	$1.055 \cdot 10^3$	0.0183	$0.559 \cdot 10^3$	0.0096	$0.293 \cdot 10^3$	0.0051	$0.156 \cdot 10^3$	0.0026	$0.079 \cdot 10^3$	0.0015	$0.045 \cdot 10^3$
Te 126	1.6610		1.6610		1.6610		1.6610		1.6610		1.6610	
Te 128												
Te 130												
I 127	0.9739		0.9879		0.9955		0.9933		1.002		1.003	
I 129												
I 131	0.0032	$3.904 \cdot 10^2$		$2.964 \cdot 10^1$		2.196						
Xe 133	0.001	$1.720 \cdot 10^2$		4.365								
Xe 128	14.1006		14.1023		14.1029		14.1029		14.1029		14.1029	
Xe 130												
Xe 131												
Xe 132												
Xe 134												
Xe 135												
Xe 136												

FISSION PRODUCTS CONTENT OF THE IRRADIATED REFERENCE FUEL

gr/kg of FRESH FUEL

ISOTOPE	30 d.		60 d.		90 d.		120 d.		150 d.		180 d.	
	gr/kg	ci/kg										
Cs 133	8.29		8.29		8.29		8.29		8.29		8.29	
Cs 135												
Cs 137	1.35	1.179.10 <sup>2</sup>	1.35	1.176.10 <sup>2</sup>	1.35	1.174.10 <sup>2</sup>	1.35	1.172.10 <sup>2</sup>	1.345	1.170.10 <sup>2</sup>	1.34	1.167.10 <sup>2</sup>
Ba 134												
Ba 137	4.2141		4.2141		4.2141		4.2141		4.2141		4.2141	
Ba 138												
Ba 140	0.0211	1.521.10 <sup>3</sup>		2.956.10 <sup>2</sup>		5.840.10 <sup>1</sup>		1.150.10 <sup>1</sup>				
La 139	3.6356		3.6356		3.6356		3.6356		3.6356		3.6356	
Ce 140												
Ce 142	6.7344		6.7511		6.755		6.7554		6.7558		6.7558	
Ce 141	0.1386	3.992.10 <sup>3</sup>	0.0732	2.071.10 <sup>3</sup>	0.0387	1.095.10 <sup>3</sup>	0.0205	0.580.10 <sup>3</sup>	0.0108	0.305.10 <sup>3</sup>	0.0057	0.161.10 <sup>3</sup>
Ce 144	1.2195	3.865.10 <sup>3</sup>	1.1311	3.585.10 <sup>3</sup>	1.0491	3.325.10 <sup>3</sup>	0.9731	3.084.10 <sup>3</sup>	0.9027	2.861.10 <sup>3</sup>	0.8374	2.654.10 <sup>3</sup>
Pr 141	3.2408		3.2884		3.3202		3.3354		3.3482		3.3553	
Pr 143	0.0211	1.403.10 <sup>3</sup>	0.0045	0.299.10 <sup>3</sup>		6.450.10 <sup>1</sup>		1.33.10 <sup>1</sup>		2.860		
Nd 142												
Nd 143												
Nd 145												
Nd 146	10.0129		10.0828		10.1338		10.1720		10.1968		10.2245	
Nd 148												
Nd 150												
Nd 147	0.0052	4.165.10 <sup>2</sup>		6.408.10 <sup>1</sup>		9.832						

FISSION PRODUCTS CONTENT OF THE IRRADIATED REFERENCE FUEL

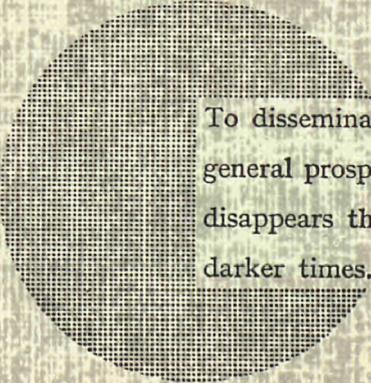
gr /kg of FRESH FUEL

ISOTOPE	30 d.		60 d.		90 d.		120 d.		150 d.		180 d.	
	gr/kg	ci/kg										
Pm 147	1.0333	9.485.10 <sup>2</sup>	1.0134	9.303.10 <sup>2</sup>	0.9904	9.091.10 <sup>2</sup>	0.9700	8.904.10 <sup>2</sup>	0.9482	8.704.10 <sup>2</sup>	0.9217	8.461.10 <sup>2</sup>
Sm 151	0.4034	1.089.10 <sup>1</sup>										
Sm 147	1.9309		1.9309		1.9309		1.9309		1.9309		1.9309	
Sm 148												
Sm 149												
Sm 150												
Sm 152												
Sm 154												
Eu 153	0.1427		0.1427		0.1427		0.1427		0.1427		0.1427	
Eu 154	0.0426	6.134	0.0425	6.120	0.0422	6.076	0.0421	6.062	0.0420	6.048	0.0418	6.019
Eu 155	0.1022	1.387.10 <sup>2</sup>	0.1008	1.270.10 <sup>2</sup>	0.0977	1.231.10 <sup>2</sup>	0.0942	1.186.10 <sup>2</sup>	0.0913	1.150.10 <sup>2</sup>	0.0878	1.106.10 <sup>2</sup>
Eu 156	0.0006	3.360.10 <sup>1</sup>		8.400								
Gd 155	0.1942		0.1942		0.1942		0.1942		0.1942		0.1942	
Gd 156												
Gd 157												
Gd 158												
Tb 159	0.0013		0.0013		0.0013		0.0013		0.0013		0.0013	
Total <u>Σ</u>	101.8	40,657.-		27,466.-		20,791.-		16,582.-		13,790.-		11,297.-

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Alfred Nobel

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