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FRENCH DEVELOPMENT PROGRAM ON FUEL CYCLE

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ABSTRACT

The need to close the fuel cycle of fast reactors makes the development of the cycle installations (fuel fabrication, irradiated assembly conditioning before reprocessing, reprocessing and waste management) especially independent with the development of the reactor.

French experience with the integrated cycle over a period of about 25 years, the tonnage of fuels fabricated (more than 100 t of mixed oxides) for the Rapsodie, Phénix and SuperPhénix reactors, and the tonnage of reprocessed fuel (nearly 30 t of plutonium fuel) demonstrate the control of the cycle operations.

The capacities of the cycle installations in existence and under construction are largely adequate for present needs, even including a new European EFR reactor. They include the Cadarache fuel fabrication complex, the La Hague UP2-800 reprocessing plant, and the Marcoule pilot facility.

Short- and medium-term R and D programs are connected with fuel developments, with the primary objective of very high burnups.

For the longer term and for a specific plant to reprocess fast reactor fuels, the programs could concern new fabrication and reprocessing systems and the study of the consequences of the reduction in fuel out-of-core time.

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1 INTRODUCTION

The operation of fast breeder reactors (FBR) entails the implementation of a complete fuel cycle. Their essential virtue is the upgrading of the U 238 in plutonium-burning reactors, and reprocessing is necessarily associated with them.

Thus with the progressive development of the FBR system in France, the corresponding resources in the fuel cycle have been set up in a coherent manner.

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- The Rapsodie 40 MWt experimental reactor commissioned in 1967 at Cadarache is associated with:
  - a fabrication line for 0·5 t (oxide)/year of the Cadarache plutonium fuel fabrication complex (CFCa),
  - the La Hague AT1 reprocessing facility commissioned in 1969, shut down in 1979 and currently being decommissioned,
- The Phénix power plant at Marcoule (250 MWe) commissioned in 1973 is associated with:
  - a fabrication line for 4 t (oxide)/year at the CFCa (Complexe de Fabrication de Cadarache),
  - the adaptation of the Marcoule pilot facility (APM) to the reprocessing of fast oxide fuels in 1973, followed by its renovation and capacity increase undertaken in 1983 (TOR project, Tête Oxyde Rapide, fast oxide head end) to 5 t (ML)/year, more than what is needed to close the Phénix cycle: the renovated APM was recommissioned in January 1988. Part of the Phénix fuel (core 1) was nevertheless reprocessed from 1979 to 1984 in the UP2 plant at La Hague, by dissolution in the HAO facility (Haute Activité Oxyde, high activity oxide) in the presence of gadolinium and mixing with a solution of GCR fuels (Bugey).
- The Creys Malville power plant (SPX 1) is associated with a fabrication line for 20 t (oxide)/year of the CFCa. The capacity of the APM is insufficient to reprocess all the spent fuels from SPX 1. After having considered this operation as part of more ambitious projects (PURR Project, then MAR 600 Project) fitting into the context of a relatively large-scale development of fast reactors, the reference solution today is treatment by dilution in the UP2-800 plant.

In short, it is clear that France can fabricate the fuel and reprocess it, but this does not mean that everything is definitively fixed for decades to come. Much work remains to be done, although overall industrial feasibility has been demonstrated.

For the long term, R and D on the fuel cycle will have to be consistent with the development projects of the fast reactor system. In the absence of clear scenarios, we shall provide a list of R and D topics generally covering the subsequent steps of the fuel cycle - not including residence in the reactor:

- fuel assembly fabrication,
- conditioning of irradiated assemblies before reprocessing,
- reprocessing and waste management.

2 FUEL ASSEMBLY FABRICATION

The fabrication of uranium and plutonium mixed oxide fuel by the mechanical mixing process is fully developed at the present time. The results obtained meet the present objectives of the reactors as well as reprocessing.

Since 1964, the CFCa has produced a total or more than 100 t of mixed oxides for the Rapsodie, Phénix and SuperPhénix reactors (Table 1). With minor changes, this installation could be capable of fabricating the fuel for one or two 1500 MWe reactors.

Table 1

Production of fabrication facilities (CFCa)
Status on 31 May 1991

reactor	mixed oxide (t oxide)	number of fuel assemblies
Rapsodie	1·6	570
Phénix	34·0	778
SuperPhénix 1	80·0	765

Medium-term developments are associated with improving the fuel in order to obtain very high burnups.

- On the fuel side, there is a clear tendency to increasing the plutonium concentration in the mixed oxide, due to the change in the isotopic composition of the plutonium used and the choice available of the heterogeneous core concept.

- On the cladding material side, to extend beyond the already excellent possibilities of 15.15 Ti steel, it is planned to use ferritic steels.

For the longer term, development guidelines could be concerned with the following.

- Fabrication of so-called advanced fuels such as nitrides, carbonitrides and carbides, and possibly IFR (integral fast reactor) type metallic fuels, in order to increase burnups. R and D is currently under way on a very modest scale on nitride fuels. The choice of this type of fuel was dictated for reasons of better integration in the fuel cycle, especially better compatibility with present reprocessing plants.

- The oxide fuel fabrication process, using methods that do not require the separate supply of uranium and plutonium oxides by the reprocessing plant in order to modify the final preparations of the basic materials:

  - direct codenitration, with or without chemical reagent, by microwave heating,
  - the Nitrox process,
  - sol/gel processes.

- Other developments may result from changes in the specifications of the basic materials used by the fuel fabricators, either for economic reasons, since these materials are produced in 'simplified' reprocessing plants with a small number of purification cycles, but leading to lower decontamination factors, or for the transmutation of the actinides by recycling them in the fuel itself, or in the form of targets.

3 CONDITIONING OF IRRADIATED FUELS BEFORE REPROCESSING AND THEIR TRANSPORT

The duration of the fuel cycle is an important parameter in evaluating the cost of the cycle, and the optimum still remains to be found. The short cycle reduces the time for obtaining the quantity of plutonium necessary to make a new core, but the level of residual power of the

assemblies is a severe constraint for their packaging, transport and, to some degree, their reprocessing. By contrast, the short cycle reduces the storage requirements and the quantity of basic material immobilized. For the EFR (European Fast Reactor) Project, a number of competing alternatives are still in the field today. For 'short' out-of-core times, the European engineering firms propose packaging the assemblies in sodium, from reactor discharge to the reprocessing plant. Other alternatives, which are consistent with longer out-of-core cycle times, make use of transfer under gas, possibly followed by interim storage of the assemblies in a pond.

In the longer term, as part of studies relative to the 'short' out-of-core cycle, an assembly disposal system could be considered, in which the assemblies would be discharged under gas at a high residual power level, a maximum of about 25 kW. This scenario implies control of the thermal and physicochemical mechanisms encountered throughout the disposal system. It is also necessary to check whether the residual sodium can be eliminated effectively and rapidly.

An R and D program has already been initiated at the CEA to support this alternative. The variations in the parameters include the corresponding ranges, both at SuperPhénix 1 and at the EFR Project, and accordingly make this program useful in the short and medium term.

In contrast to the above, if the time elapsed between reactor discharge and arrival at the reprocessing plant were to involve long-term interim storage of the assemblies under water, it would be necessary to make sure that the main components of the assemblies (hexagonal tube, spacer wire and rods) do not, by corrosion, undergo deterioration liable to cause contamination of the storage pond water, and hinder the transfer of the assemblies to the reprocessing unit. To achieve this, a large number of tests on fuels from the Fortissimo and Phénix reactors is under way, and should help to explore the areas pertinent both to the SuperPhénix 1 reactor, and those which may be proposed with respect to the fast reactors.

4 REPROCESSING AND WASTE MANAGEMENT

4.1 Present situation and review

The feasibility of fast mixed oxide fuel reprocessing has been sufficiently demonstrated, both in the CEA's pilot installations, without dilution, and in COGEMA's UP2-400 plant, with dilution by gas/graphite fuel (Table 2).

- The AT1 facility at La Hague (1 t/day) has reprocessed more than 1 t of fuel, chiefly from Rapsodie, irradiated to 120,000 MWday/t (max core ox), and even for a small part up to 200,000 MWday/t.
- The Marcoule pilot facility (APM) has reprocessed 13 t (ML) of Phénix plutonium fuel since 1975, including 7·2 t (ML) since its recommissioning in January 1988 after renovation.
- The UP2 plant at La Hague has reprocessed about 10 t of Phénix core 1 fuels from 1979 to 1984 by dilution with GCR fuel.

The R and D programs supporting this activity were set up at the CEA more than 25 years ago, even before those connected with the processing of LWR fuels.

In recent years, all research projects on FBR fuel reprocessing have been conducted to support projects for specific plants:

- up until 1982, the PURR Project (130 t/year) initiated in 1977,
- from 1982 to 1985, the MAR 600 Project (50 t/year).

The MAR 600 plant should cover the needs of SuperPhénix 1 and three other 1500 MWe reactors.

Table 2

Production of reprocessing facilities
Status on 31 May 1991

facility	reactor	PuO_2 (U Pu)O_2 (%)	years	tonnage (ML)
AT1 (La Hague)	Rapsodie	25 to 30	69 to 79	0·91
	Phénix	18	78 to 79	0·18
APM (Marcoule)	Rapsodie	30	75	0·05
	KNK 1	enriched uranium	75 to 76	1·65
	Phénix	enriched uranium	77 to 78	2·30
	Phénix core 1	18	79 and 83	}
	Phénix core 2	25	79 to 83	}6·74
	Phénix cores 1 and 2	18 and 25	88 to 90	7·20
HAO, (UP2 La Hague)	Phénix core 1	18	79 to 84	10·07
	total			29·10

4.2 Short and medium-term prospects

For the time being, it does not appear realistic to expect the construction of a significant number of FBRs before the years 2010 to 2020. The principal factor will be to pick the time when it will be necessary to have a totally specific installation, and to decide on the size of this installation, in relation to its cost, the cost of transporting the irradiated assemblies, and the cost of waste management, as well as the rate of penetration of FBRs connected with the availability of plutonium.

However, the Phénix and SuperPhénix 1 reactors exist today, and the European EFR reactor project could culminate in an industrial prototype around the year 2000.

The fuels of these reactors will be reprocessed by relying on the UP2-800 plant operating by dilution, with the possibility of using the available capacity of APM to reprocess part of these fuels.

In fact, FBR and PWR fuels are similar (oxide). The core of the process (extraction cycles) can handle both types of fuel, allowing treatment by dilution. Three special factors need to be determined:

- dismantling of the FBRs before shearing,
- the high plutonium content (dissolution) extraction cycles,
- the high burnup (vitrification, extraction cycles).

The short- and medium-term R and D programs have been adjusted in the following directions:

- continue to match developments in fuels (burnup, clad material) by examining their behavior and studying reprocessing by dilution in UP2-800,
- optimal use of the APM by specializing the runs in order to acquire specific knowledge, especially with two main objectives:
  - reprocessing of fuels with very high burnup (over 120,000 MWday/t),
  - reprocessing of fuels with short cooling times.

We shall examine below the specific 'fast' programs connected in particular with the operations employing fuel assemblies which are morphologically different from those of light water reactors.

4.2.1 Assembly dismantling

The presence of the very thick hexagonal tube (TH) makes shearing of the whole assembly unfeasible, as practised for LWR fuels. The assemblies, rid of sodium, must therefore be dismantled to obtain separate assemblies. This operation, performed at the Fuel Assembly Dismantling Laboratory (LDAC, Laboratoire de Démantèlement des Assemblages Combustibles) at Cadarache for Rapsodie fuels, is currently performed at the examination cell (CE) of the Phénix power plant for its fuels, by mechanical milling on the angle of the TH containing a fake fuel rod to avoid the risk of fuel rod damage.

For SuperPhénix, it is not yet known whether this disassembly will be performed at the power plant or in a specialized installation, as for example the Irradiated Assembly Monitoring Laboratory (LSAI, Laboratoire de Surveillance des Assemblages Irradiés) at Marcoule.

After having examined a number of alternatives (milling, laser cutting), the cracking process was selected for the MAR 600 Project, in which a weld is executed with filler metal in the melt zone of a foreign metal (copper) causing a stress crack.

After inactive development at Grenoble, four irradiated Phénix assemblies were dismantled by this method in 1983 and 1984 at the Cadarache LECA. The advantages of this process are the absence of any production of solid debris and the projection of fines, and the possibility of avoiding damage to the rods placed at the opening position without requiring very accurate positioning.

The Marcoule LSAI is equipped with such a system for the transverse cutting of end pieces and the longitudinal opening of SuperPhénix assemblies.

After opening the TH, the rods are removed intact by a combination of thrust and percussion on their support.

These techniques could require a number of adjustments for large-scale industrial use.

4.2.2 Rod shearing

For this operation, the question arises whether or not to remove first the spacer wire. The answer is related to the operation of the shear and that of the dissolver. The reprocessing of Rapsodie fuels at AT1, and those of Phénix at the APM and at UP2, was carried out without removing the wire (rotary shear rod by rod at AT1 and APM, and canister shear by guillotine-type blade at UP2 and batch dissolvers. Only the shearing of non-intact rods at APM require

this operation. However, the presence of sections of chopped wires in a continuous dissolver could raise a few problems (risks of holdup, clogging).

Rod shearing into lengths of a few centimeters to facilitate the dissolution of the fuel does not raise any particular difficulty, either with the small shears fed rod by rod, or with the larger shears operating on a row of rods, on a bundle or on canisters filled with rods.

4.2.3 Fuel dissolution

Two aspects need to be considered:

- total dissolution (or the most complete possible dissolution) of the uranium or plutonium in boiling nitric acid,
- behavior of the cladding material in the dissolution medium.

Uranium oxide UO_2 and plutonium oxide PuO_2 form solid solutions which are soluble in nitric acid up to $PuO_2/UO_2 + PuO_2$ ratios of about 35%. Above this level, it behaves like PuO_2, which is virtually insoluble. Hence the importance of keeping away from this value (we decided to remain below a limit of 25%) and ensuring fabrication quality that guarantees the homogeneity of the mixed oxide, excluding the presence of islands or aggregates of PuO_2 or mixed oxides, in which the Pu content could be higher than the limit content of 35%.

The Phénix fuels (core 1 at 18% Pu, core 2 at 25%) raised no problem in this respect, but this does not apply to certain foreign fuels of earlier fabrication (KNK 2 containing 30% Pu). Extreme vigilance is therefore necessary, and if the plutonium concentration is increased, it is essential to check the solubility of the fresh oxide, by means of solubility tests, for which irradiation plays a rather favorable role.

The basic knowledge acquired so far on the kinetics and characterization of the different products obtained (and especially of the dissolution insolubles) will have to be updated as the burnups are increased.

The behavior of the cladding material during dissolution may have repercussions on the down process steps.

The passage into solution of elements such as iron, which are found in the fission product solutions, could raise problems of corrosion and of the dimensioning of the concentration, storage and vitrification facilities.

The disintegration of the clads could lead to pipe clogging and could overload the clarification units.

The cladding materials used so far (316 L, 316 Ti, 15-15 Ti) display good behavior which should continue to be scrutinized as the burnup is increased.

If the use of ferritic steels, which exhibit poor behavior in dissolution media, proves indispensable, this would certainly entail the need to separate the clad before dissolution. Two alternatives appear to be available:

- a mechanical process by crushing/screening, designed some years ago on a small scale,
- a thermal process by clad fusion, developed in the Soviet Union, which offers the advantage of conditioning the metallic wastes simultaneously.

For the PURR and MAR 600 Projects, it was planned to use a continuous dissolver of the annular helicoidal type operating as a vibrating corridor, offering the advantage of high capacities while being geometrically safe. A prototype on the scale of the APM was built, and a location reserved in a cell of this facility.

The design of a dissolver should be resumed as part of the new specific fast fuel plant projects in accordance with the capacity and the choice of the cladding material. In fact, if prior separation of the clad proves necessary, it could be possible to use more compact dissolvers fed exclusively with oxides, such as the powder dissolver.

In the case of the risk of incomplete dissolution of the plutonium, it is possible, as COGEMA is considering for UP2-800, to provide for a supplementary dissolver using silver ions, a process used for non-irradiated plutonium oxide, but remaining to be developed for irradiated products.

4.2.4 Clarification

The separation of insoluble or reprecipitated solid products in order to obtain the clearest possible solution before the extraction operations is not specific to fast fuels. A thermal problem only arises for slightly cooled fuels if the feed of solution to be clarified to the centrifuge is stopped (the centrifuge also cools the cake) for the periodic removal of the centrifuged solids.

4.2.5 Extraction cycles

These operations are also not specific to fast fuels. However, given the relatively high Pu/U ratio (a function of the dilution provided by the built-in axial blanket), it could be preferable to use destructible reagents (hydroxylamine) or electrolysis for uranium/plutonium partition, instead of uranium IV.

All these considerations on light water fuels designed to optimize and simplify the reprocessing operations (reduction of the number of extraction cycles) or to separate the actinides can obviously be transposed to fast fuels.

4.2.6 Waste

There are few specific problems in this area, since the waste generated by MOX reprocessing appears to constitute an envelope, due to the isotopic composition of the Pu.

One specific problem is related to the type of clad. The process being developed to reprocess the hulls is fusion in an autocrucible, which is more favorable to the stainless hulls of fast fuels because the waste is also slightly decontaminated.

4.3 Long-term prospects

The long-term programs obviously depend on the development scenarios of fast reactors and the alternatives selected for the fuel.

- For the longer term, if the new carbide, nitride, carbonitride or metallic fuels are used, it would be necessary to intensify the research conducted today on a very modest scale on nitrides, to determine both the optimal conditions for dissolution and offgas treatment (problem of carbon 14 for nitrides) and to facilitate the extraction operations.

- If the cladding materials used are incompatible with the dissolution media, it would be necessary to resume studies on fuel clad separation and powder dissolution.

- In the case of very short out-of-core residence times, it would be necessary to examine all the consequences of increasing the activities and specific power, especially in the clarification step.

- It may also be possible to use other processes than the PUREX process. From 1957 to 1976, we investigated a so-called dry method process based on the separation of U, Pu and FP in the form of their fluorides. It must also be pointed out that the United States is working on a pyrometallurgical process using exchanges between salts and molten metals, as well as pyrogenic electrolysis, as part of the IFR Project, for metallic fuels.

These processes, which will probably be better adapted to new fuels, merit at least thoughtful consideration.

5 CONCLUSIONS

French experience in the fuel cycle of fast breeder reactors integrates the activities conducted for more than 25 years by following development in reactors, from Rapsodie to SuperPhénix 1, demonstrating the validity of the basic processes adopted.

The present cycle installations, both for fuel fabrication (CFCa) and for reprocessing (APM and UP2-800) are amply dimensioned for reactors in operation today (Phénix and SuperPhénix 1) or reactors that could emerge before the end of the century (EFR prototype reactor, for example).

The possibility of reprocessing very significant quantities of fast fuels by dilution in light water fuel is of capital importance. This could make it possible, in France, to elude the paradox of fast fuel reprocessing cost, which could be relatively cheap provided a plant were available, hence a certain number of reactors - if the plant is specific.

Short- and medium-term R and D for fast fuel reprocessing is practically limited to observing fuel developments to obtain increasingly high burnups and to reprocessing by dilution in the UP2-800 plant. Major resources are devoted at the CEA to improving the plants at La Hague, for optimal waste management and the development of new packaging methods, and to boost the capacity and reliability of the facilities for adaptation to the new fuels. They will largely be housed in the new Atalante facility at Marcoule, of which the first section is scheduled for active startup in September 1991.

Only in the case of intensified development of fast reactors could R and D for a specific or even multiple reprocessing plant be reactivated..

Since the activities of the cycle and the reactor are strongly inter-dependent, and the alternatives selected for one interfering with the others, it is necessary for all the partners in the cycle (including the reactor) to work together closely in order to optimize the overall system, to obtain a minimum kWh cost, and not to deal with each part separately.