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FAST BREEDER REACTOR FUEL REPROCESSING IN FRANCE

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Simultaneous with the effort on fast breeder reactors launched several years ago in France, equivalent investigations have been conducted on the fuel cycle, and in particular on reprocessing, which is an indispensable operation for this reactor.

(1) The Rapsodie experimental reactor was associated with the La Hague reprocessing plant ATI (1 kg/day), which has reprocessed about one ton of fuel.

(2) The fuel from the Phénix demonstration reactor is reprocessed partly at the La Hague UP2 plant and partly at the Marcoule pilot facility, undergoing transformation to reprocess all the fuel (TOR project, 5 t/an).

(3) The fuel from the Creys Malville prototype power plant will be reprocessed in a specific plant, which is in the design stage. The preliminary project, named MAR 600 (50 t/y), will mobilize a growing share of the CEA's R and D resources, as the engineering needs of the UP3 "light water" plant begins to decline.

. As can be seen from Table I which details the fuels reprocessed, nearly 20 tonnes of heavy metals irradiated in fast breeder reactors have been processed in France, 17 of which came from Phénix.

. The plutonium recovered during this reprocessing allowed the power plant cycle to be closed. This power plant now contains approximately 140 fuel assemblies made up with recycled plutonium, that is, more than 75% of the fuel assemblies in the Phénix core.

FRENCH EXPERIENCE OF FAST BREEDER REACTOR FUEL REPROCESSING

	FUEL	INITIAL Pu/U+Pu	YEARS	PLANT	GWd/t (1)	COOLING TIME (2)	kg U+Pu
RAPSODIE	1st core	25%	69 - 79	AT_1	40 - 45	6 - 12	250
RAPSODIE	Fortissimo	30%	69 - 79	AT_1	50 - 120	5 - 24	660
RAPSODIE	Fortissimo	"	75	SAP	55 - 76	6 - 10	50
KNK I (Enriched U)			75 - 76	SAP	3.4 - 6.8	12 - 20	1,650
PHENIX	Enriched U		77 - 78	SAP	38 - 45	10 - 30	2,300
PHENIX	Pu core I	18%	78 - 79	AT_1	8 - 44	18	180
PHENIX	Pu core I	"	79	SAP	37	10 - 30	155
PHENIX	Pu core I	"	79 - 83	UP_2	28 - 34	38 - 50	7,900
PHENIX	Pu core II	25%	79 - 83	SAP	36 - 83	14 - 42	6,360
PHENIX	Pu core I	18%	83	SAP	35 - 100	29 - 36	225
TOTAL							19,730

(1) Maximum burn-up per tonne of core oxide.

(2) In months

TABLE I

A. ACCUMULATED EXPERIENCE

1. ATl Workshop at La Hague

The ATl workshop at La Hague, specially designed to reprocess RAPSODIE fuel with a capacity of 1 kg per day, went on stream in 1969, and since July 1979, on which date it was shut down, it has reprocessed more than one tonne of mixed oxides irradiated to a burn-up as high as 120,000 MW d/t and sometimes very slightly "cooled" (five months, and 1.5 months for a small number of assemblies), thus closing the RAPSODIE cycle. Part of this fuel (about 150 kg) came from the core of PHENIX.

Operation of this workshop has proved that reprocessing of this type of very high specific activity fuel was feasible, and it has allowed a lot of data relating to the process chemistry to be collected. However, owing to its small size and to the nature of its equipment, the benefits it brought were rather limited as regards industrial reprocessing of these fuels.

2. La Hague UP2 Plant

COGEMA's UP2 plant carried out four reprocessing runs of Phénix fuel. After chopping and dissolution in the presence of gadolinium in the HAO plant (LWR head end), the solution was mixed to a solution produced by the dissolution of gas-graphite fuels in order to approximately restore the properties of "LWR" fuel. All these four runs concerned nearly 8 t of core I of the reactor and no particular problems were encountered.

3. Marcoule Pilot Facility

The Marcoule pilot facility (SAP) went on stream in 1963. In 1973, it was adapted to the reprocessing of oxide fuels. From this time, it has reprocessed nearly 11 tonnes of fuels, most of them from fast breeder reactors.

After reprocessing Rapsodie blankets, oxides from EL4, from the core of Rapsodie-Fortissimo, fuel from the German reactor KNK (1^{st} core) and from the uranium-enriched Phénix core, the pilot facility was exclusively dedicated to the Phénix plutonium core (core II) (1), which is most representative of this type of reactor. From 1979 to 1983, five reprocessing runs concerning an aggregate amount of 6.6 tonnes of U+Pu allowed a very large amount of data to be collected. It should be mentioned that two small "special" batches were reprocessed during the last run:

- one assembly whose maximum burn-up represents a record: more than 100,000 MWd/t,
- one assembly whose plutonium was being reprocessed for the third time:

(1) The Phénix core is composed of 2 fuel zones: zone 1 with an 18% Pu content and zone 2 whose Pu is 25% (Pu/U+Pu ratio in the mixed oxide).

. it had been extracted from a gas-graphite fuel in 1977 at the La Hague plant and had been introduced in Phénix core I,

. it had been extracted again from this core in 1979, also at the La Hague plant.

At each reprocessing run, the Pu balances became better controlled and the deviations progressively decreased. The aggregate deviation of the five runs is 0.48%.

The main results of the Phénix core II run are as follows:

- iodine is distributed, after batch dissolution, between the dissolution solution (0.4% at the end of boiling), the recombination bottom tank (0.5 to 8%), the soda column bottom tank (90 to 99.5%) and the soda-washed gasses.

- the iron content of the solution as a function of dissolution time and cladding type has been monitored over 204 dissolutions (6 tonnes of fuels).

- the cladding wastes contamination has been measured for the expansion chambers and the hulls. Ruthenium is the main contaminant: its radioactivity is 75 $Cikg^{-1}$, which represents 97% of the fission products' activity. The activation products account for 17% of the aggregate $\beta\gamma$ activity. Hull contamination by plutonium amounts to 150 $mg.kg^{-1}$, but decontamination is rather easy. The high level of contamination of the solid wastes were the main cause of the problems encountered during operation: irradiation, contamination, residual thermal energy, damage to the ventilation equipment of the cells where the dissolution baskets are handled.

- measurements carried out on dissolution residues turned out to be rather difficult. It was nevertheless possible to estimate that ^{106}RuRh is approximately equally distributed between solution and dissolution residues. Clarification, which is a low-yield process showed that the residues followed the fission products, in spite of the complexity of the path (7 buffer storages connected by air-lift or by vapour ejector).

- numerous observations have been carried out concerning the hydraulic behaviour of the pulsed columns (variation of the clogging limit in time, behaviour of the solids).

- the $\beta\gamma$ decontaminations have been measured at each cycle. The activity level of the first cycle feed solution (30 to 240 Cil^{-1}) does not have much influence on the FD obtained.

- after the first co-extraction cycle, RuRh becomes the main contaminant.

- uranium βγ activity is in conformity with the standards (less than 500 $\mu Ci.kg^{-1}$ U) at the output of the second cycle (partition cycle).

- the plutonium βγ contamination is only 0.8 to 4.5 $\mu Ci.g^{-1}$ (standard 1 $\mu Ci.g^{-1}$) at the output of the second cycle and it can be treated in the UP1 plant by oxalic precipitation without any third cycle being required.

- partition in settler/mixers by electrolysis or with hydroxylamine nitrate with some addition of U IV proved fully satisfactory. Average uranium contamination by plutonium was 0.25 ppm (0.05 ppm during steady-state operation), at the output of the second cycle.

- the third uranium cycle easily provided for less than 0.04 ppm Pu in uranium (that is, 10,000 dpm/g of U).

- plutonium contamination by uranium was 2,000 ppm at the output of the second cycle (required values are 15,000 ppm before oxalic precipitation).

- between 50 and 80% of the neptunium is extracted at the first cycle and it follows either uranium for 85% in the U IV partitions or plutonium for 90 to 100% in the hydroxylamine nitrate partitions, if the operating conditions are correctly adjusted (O/A, acidity levels).

In addition to these operation results, the workshop has experimented during certain runs new techniques produced by the R and D effort:

- solvent treatment by hydrazine carbonate instead of sodium carbonate,
- iodine entrapment on solid absorbant,
- measurements of the hull temperature inside the bins,
- hull decontamination tests.

B. THE PROJECTS

1. The TOR Project (5 t/y)

The pilot facility which has been shut down since the summer of 1983 is now undergoing thorough refitting, which constitutes the TOR Project, whose purpose is threefold:

- raising its capacity to 5 t of heavy metals per year,
- increasing its reliability, i.e. its load factor,
- matching the plants to the new reprocessing requirements and, in particular, offering new R and D potential.

The facility has three units, TOR 1, TOR 2 and TOR 3.

TOR 1, which is located in a new building north of SAP, consists of some 20 $\alpha\beta\gamma$ cells containing head-end equipment. This new workshop is equipped with extensive remote maintenance facilities, including:

- a common utilities unit involving spent fuel receipt, fuel storage and storage of solid wastes,
- a mechanical unit designed to accommodate choppers and, subsequently, advanced oxide/clad separation techniques,
- a dissolution unit in which it will be possible to install two new models of dissolvers connected to the gas treatment circuit,
- a clarification unit also offering several variants,
- a waste packaging unit for hulls, dissolution insolubles, iodine, etc.

Thus, two parallel equipment lines will be available for the initial reprocessing operations: a main line using essentially proven technology for production, and a set of experimental units that can be positioned on a by-pass to the main process line.

TOR 2 and 3 use part of the premises and equipment of the former pilot facility. TOR 2 mainly houses storage facilities for end products (U and Pu) in the form of purified nitrate. TOR 3 will contain a new first pulsed-column extraction cycle with a possibility of first cycle partition, as well as a new fission product solution concentrator.

TOR is scheduled to come on stream in 1985. In addition to the Phénix core, TOR will reprocess fuel from the West German KNK reactor and a small fraction of the core of the Creys Malville power plant.

The SAP, so renewed, will provide France with a first-grade tool for developing the process and the components in active medium at the time when the construction of the plant is to be decided. In this way, this plant could continuously benefit from the qualitative and quantitative experience accumulated in this pilot facility.

2. Project of an Industrial-Scale Unit

The CEA's R and D Department dedicates an ever-growing part of its resources to the MAR 600 project. Most of the steps of the process have been experimented in laboratory or in prototype plants, in order to optimize the process flow and promote the techniques best suited to the specific problems of the future plant. The pilot facility, supplemented by TOR, will play a

major role in demonstrating the processes. In particular, it will allow tests to be conducted on the continuous working dissolver prototype, on hull fusion, iodine entrapment on solids, backflow extraction cycles, etc. Thus, when a decision will have to be made on the future of this project, a whole set of results that will help in selecting the solutions most likely to succeed will be available.

C. SPECIFIC PROBLEMS RELATING TO "FBR" FUEL REPROCESSING

There is no technical rupture between the reprocessing of FBR fuels and the operations carried out to reprocess the fuels produced by other types of nuclear plants. The process used is the PUREX process based on the separation and purification of uranium and plutonium by successively chopping the fuel pins, putting the oxide in nitric solution, performing extraction cycle by TBP and converting the Pu nitrate into oxide by the oxalic channel.

The implementation of this process benefits from the industrial experience accumulated in the processing of large quantities of UNGG (natural uranium-graphite-gas) fuels and more than 900 tonnes (U+Pu) of fuels produced by the light water reactors in the HAO workshop of the La Hague plant. The requirements applying to the end products are the same.

The problems of "fast" oxide reprocessing are different from those of "light water" oxide reprocessing in three ways:

- configuration and composition of fuel assemblies,
- plant capacity, which is smaller,
- deadlines, which are longer.

1. Configuration and Composition of Fuel Assemblies

The specific features of fast breeder fuels are well known:

- presence of sodium,
- the existence of a thick case enclosing the fuel pins (the "hexagonal tube"),
- stainless steel clad and presence of a helical spacer wire around each pin, whose diameter is small,
- high Pu concentration (10 to 20%), which increases criticality problems and makes dissolution more difficult,
- high concentration of fission products, part of which is insoluble in nitric acid,
- high specific power due to high radioactivity.

2. Smaller Plant Capacity

Whereas the targeted capacities for "light water" plants are in the range of 1,000 t/y, those currently considered for the "fast" fuel reprocessing are about ten times smaller (taking into account the number of reactors). This factor of 10 may play an important role in certain technical decisions for head end operations. (This is not the case for operations on plutonium alone, which require capacities of the same order of magnitude.)

3. Longer Deadlines

A gap of more than 10 years can be estimated between the industrial operation of "fast" fuel reprocessing plants and that of "light water" fuel reprocessing. This interval will allow for the implementation of techniques that are still at R and D level today, mainly regarding highly "robotized" remote maintenance units, computerized equipment control and waste treatment mehtods (iodine, solvent, hulls) and development of the salt-free process. All these trends should lead to significant savings in investment and operating costs

D. CURRENT R AND D EFFORT FOR BRINGING THE PROCESS TO INDUSTRIAL SCALE

1. R and D Resources

The CEA's R and D resources for the development of processes are mainly:

- in Fontenay-aux-Roses: the hot laboratories which include the shielded lines where the dissolution, clarification and extraction operations are tested as Phénix is supplying ever more irradiated fuels. Analogue tests are conducted whenever new cladding materials appear.

  - The alpha laboraties, where pulsed columns have been installed for chemistry studies.

  - The material corrosion study and analysis method development laboratories.

  - Chemical engineering halls.

- in Marcoule: The industrial prototype facilities, built and operated at true scale, with their ancillary buildings and in accordance with the industrial technology, but without radioactivity.

  - The pilot workshop which experiments the processes and equipment at semi-industrial scale during extensive periods where irradiated fuels representing the cycle are treated.

2. Orientation of the Programmes

The R and D programme which has been initiated concerns techniques which can be adapted to suit plants of varying capacity. This programme is intended to prove the feasibility of the fuel cycle, whatever the broad outlines chosen for developing the fast neutron technology within the French and European framework in the 90's.

2.1 Wrapper and End-Pieces Cutting: Shearing of the whole fuel assembly as it is done for the LWR assemblies has been ruled out because of the heavy strength which would be needed and because of the problems which would have been raised by the presence of large steel pieces in the continuous dissolver.

On the contrary, chopping of pins one after the other matched well the throughput of the plant and the dissolver concept.

It is well known, however, that the swelling of the fuel pins forbid their axial extraction from the hexagonal wrapper without cutting it beforehand.

Two ways have been tested and are still in use for the dismantling of Phénix fuel assemblies:

a) grooving the wrapper with a high-speed circular saw and splitting it mechanically,

b) milling on an edge and mechanical opening.

The first method spreads active metal particles whereas a high level of accuracy is requested in the second case.

Laser cutting has also been tested for the cutting of the Super Phénix assembly hexagonal wrapper. This method also presents the drawback of spraying active particles and a high risk of damaging the pins beneath the wrapper.

Finally, two methods have been developed which produce very small active particles and do not require either a great accuracy in positionning or a great mechanical strength.

The first method, used for transverse cutting of the assembly in vertical position, is high-frequency electrical heating. A hexagonal ring is positionned around the assembly and a HF current is produced in the loop. Heating of the wrapper combined with the weight of the lower part of the assembly will produce a quick break of the wrapper. This method can be used for the first step of dismantling, that is, removal of the assembly upper end-piece whereas the assembly is vertical on the dismantling bench.

The second cutting method is called "cracking by foreign metal embrittlement". It is well known that foreign metals can produce embrittlement of stainless steel with crack formation. A TIG-type arc welding set is used for heating the sheath and simultaneously melting a foreign metal wire, copper for example. The strain due to the heating and subsequent cooling will drive forward a very clean crack along the path of the welding device. This method will be used to cut the wrapper in parallel or transverse direction to the axis of the assembly.

It has been successfully tested on three Phénix assemblies.

2.2 Pin Chopping: Pins could be extracted one at a time from the bundle by a specially designed device.

A machine has been built for the removal of the spacer wire.

The technique used for spacer wire removal is called "rotating arc". Six TIG-type welding devices, are placed on a circular frame, centered around a hole through which the pin will be moved. An electric arc is produced between one welding electrode and the pin, then transfered to the next electrode, then to the next and so on. Thus the arc seems to be rotating around the pin. Sooner or later, the wire will be under the arc and will melt. Thanks to this device, the wire is cut into 20 cm pieces, which is convenient for melting and conditioning.

The pin, without its wire, will be stored before chopping. A rotary chopper will cut the pin in small sections of about 30 mm long. This chopper is now under development, most of the effort concerning the remote replacement of the blade.

Alternatively, a pin chopper and feeding device capable of cutting the pins with their spacer wire is also being developed.

2.3 Dissolution: During this step, chemistry and equipment problems have been encountered. Many studies had to be conducted in a shielded line to guarantee correct dissolution of the irradiated oxides and the temperature and acidity conditions have been defined. It is now estimated that the quantity of dissolution insolubles is approximately 13 kg t^{-1}, 1% of which is plutonium, the rest of it being composed of ruthenium, zirconium, molybdenum, etc. Studies are currently being conducted to characterize the volatile fission products, the dissolution residues, the dissolution kinetics, cladding corrosion and hulls.

The ring-shaped continuous helicoidal dissolver (CHD) is envisaged. The pin sections are driven up an helicoidal ramp by vibrations. The fixed central core is filled with a neutron poison. All aspects of this equipment have been studied: zirconium feasibility, criticality, qualification of the neutron-absorbing material, inspection method for the double clad, monitoring of operation in active environment, (hull hold-up, clogging detection, etc.), fitting of the drive cylinder with a motor capable of operating in a nuclear environment. A full-scale prototype of the CHD which will be installed in the TOR facility has been built in 304 L . It is now undergoing testing. This facility represents MAR 600 at one third capacity.

Furthermore, the devices appended to this equipment, such as the measurement pot, the iodine desorption column and the nitreous vapour recombination columns have been studied.

2.4 Clarification: The current lack of experience concerning the quantities of dissolution residues and their plutonium content depending on the fuels and burn-ups, as well as the lack of certainty concerning the behaviour of the fines in the CHD, have led us to include a "digestion" step. This is a new attack of the residues by concentrated acid in a subcritical tank. This requires two clarification systems which operate under different flow and performance conditions; one of these systems will produce a solid-free liquor, whereas the other will be intended to recover correctly washed solids. CEA draws from its experience in centrifuges, mainly gained during the UP3 project, for defining the machines of the future fast neutron fuel reprocessing plants.

2.5 Off-gases Reprocessing: Recombination of nitreous vapors is not a new problem, it is mainly studied for optimization purposes. On the other hand, trapping of the fission gases remains to be developed. The PbI_2 production technique implemented in the UP3 plant has a major drawback, this product must be conditioned and stored. On the other hand, the effluent containing soda and loaded with active materials increases the salt loads of the new effluent reprocessing stations where head-end concentration is to be carried out. Another solution would consist in trapping iodine on solid supports impregnated with silver. All these studies are currently being conducted. The trapping of krypton and carbon 14 has been deferred.

2.6 Solvent Extraction: The size of a plant such as MAR 600 allows the design problems concerning the balance tank to be considerably alleviated. The extraction cycles mainly try to use the salt-free process. For instance, hydroxylamine nitrate partition and solvent treatment by hydrazine carbonate and other destructible salts have been developed.

The chemistry of neptunium is now fairly well known so its partition can be controlled. It is oriented toward plutonium in order to be recycled in the reactors without any specific treatment.

The second plutonium cycle has been selected to ensure concentration and a backflow will provide the necessary flexibility. No plutonium concentrator is planned. The reextraction agent which has been considered is hydroxylamine nitrate.

The second uranium cycle, also provided with a backflow, should allow the specified requirements to be met.

This scheme will be studied in the laboratory, in small-size pulsed columns, and it will be ascertained by the operation of the TOR facility.

As for the equipment, the low flows should make it possible to ensure nuclear safety with cylindrical pulsed columns. They are currently being studied at CEA, with disk-crown packing, so as to define the dimensioning rules for the shells, the packing and the auxiliary equipment as well as the instrumentation and the cycle control.

The very stringent regulations concerning waste discharge, and the will to minimize the alpha discharges of all forms have led us to develop some "alpha trapping" operations which allow the plutonium to be reextracted from the solvent before its basic treatment. The use of hydrazine carbonate, in addition, allows the alpha and gamma contaminants to be sent back to vitrification after treatment and acidification of the basic effluent.

Finally, diluent washing operations in pulsed columns have been developed to avoid bringing TBP to the concentrators or storage areas by entrainment and solubility.

2.7 PuO_2 Preparation and Packaging: The capacities required for oxalic precipitation and oxalate calcination are of the same order of magnitude for "light water" and "fast" fuels. New techniques may be proposed for MAR 600. This remark applies to the following units:

- rotary bowl centrifugal precipitator, which allows capacities in the order of 5 $kg.h^{-1}$ per hour,
- flat filter capable of processing 5 $kg.h^{-1}$ of oxalate,
- double screw calcinator.
- PuO_2 packaging, transport and storage in recoverable 20 to 30 kg containers; these packages will be filled and emptied by pneumatic facilities, which have been full-scale tested on plutonium at Cadarache.
- Reprocessing of PuO_2 residues or "out-of-tolerance" batches thanks to oxidative redissolution of PuO_2.

2.8 Vitrification: The application of this technique to FBR fuels requires that the glass formulas be adapted, in accordance with the spectrum of fission products, to the incorporation of dissolution residues and effluents produced by the basic solution, and the soda containing alpha radioactive effluents of a plant has to be taken into consideration.

In addition, glasses of better grade can be obtained by implementing the high-temperature furnace (direct induction in autocrucible)developed on a prototype scale at CEA.

An effort has been undertaken to restore a continuous vitrification study line (PIVER II) which could be used as an active pilot facility for the fission products and the TOR dissolution residues.

2.9 Packaging of the Hulls: This problem, different from that of UP3, owing to the kind of material (stainless steel instead of zircaloy), allowed a new technology to be developed: melting in induction autocrucible allows ingots to be produced. An inactive prototype is currently operating and its introduction into a nuclear environment has begun. The gas-processing auxiliary units, the hull and spacer wire (provided it is separated) feed systems and ingot packaging still have to be defined more precisely.

It is hoped that the alpha emitters can be correctly confined and a certain level of decontamination in the "flux" (a kind of lubricant) sent back to vitrification can be achieved. The tests regarding alpha, beta and gamma emitters are still to be conducted.

This technique will be implemented in the TOR facility.

2.10 Support Studies: Computation programs for the characteristics of the fuels irradiated in Phénix and Super Phénix are being developed. New nuclear sensors, such as fibre optics spectro-photometry have been perfected. Utilization standards for such materials as zirconium are being elaborated. Other studies dealing with corrosion, analyses, etc., are supporting this general R and D programme.

2.11 Remote Control and Maintenance: The smaller space requirements of the equipment can only lead to a reduction of the cost of a reprocessing plant. Therefore, line duplication must be minimized and a compact layout must be selected. All this requires that reliable remote maintenance equipment be developed, that it be more mobile and more dexterous than the conventional mechanical master/slaves and that new opportunities offered by the development of electronics and data processing be exploited.

Thus, studies are being conducted or projects exist for partially remote-controlled telemanipulators fitted with mobile cameras and computerized controls, and special tools adapted to these telemanipulators.

CONCLUSION

The R and D effort undertaken in France for the reprocessing of the FBR fuels has been going on for many years. It takes place in a coherent programme of development for this technology, and it extends the large-scale work carried out for the industrial reprocessing of fuels produced by light water reactors in La Hague. As a feedback, this industrial reprocessing will benefit from the studies carried out on FBR fuel reprocessing.

Long-term experiments carried out to close the Rapsodie and then the Phénix fuel cycle have demonstrated that reprocessing by the PUREX process is feasible and tests are still conducted in order to develop the specific technology which will be used in the future plants. The TOR pilot facility, currently being built in Marcoule, will allow this technology to be tested on the fuel representative of Phénix from 1985.