PRESENT STATUS OF REPROCESSING WASTE CONDITIONING IN FRANCE

Maurice Delange
Director
Cogema

ABSTRACT

For its French and foreign clients, Cogema can now perform in-line conditioning and packaging of nearly all reprocessing wastes, in its UP1, UP2 and UP3 plants at Marcoule and La Hague, in specified and approved forms for final disposal meeting the regulations of the Safety Authorities of the different countries concerned.

INTRODUCTION

It is well known that France, due to its small conventional primary energy resources (hydropower, coal, oil and gas) decided very early to rely on nuclear energy to generate electricity. The 55 reactors in operation in France on 1 January 1991 include 49 PWR reactors, 4 GCR reactors, and 2 FBR reactors. They enable Electricité de France (EDF) to account for over 75% of total French demand by nuclear electricity generation, the highest rate in the world at the present time.

To accompany this vast nuclear power generation program throughout its development, the French Atomic Energy Commission (CEA), followed by Cogema (Compagnie Générale des Matières Nucléaires), set up the necessary spent fuel reprocessing installations relating to the back end of the nuclear fuel cycle.

France picked the immediate reprocessing option from the outset, and has since then continued in this path without interruption. In the application of this policy, Cogema operates three reprocessing plants today:

- at Marcoule, the UP1 plant commissioned for plutonium production in 1958, and then converted to the reprocessing of electricity generating fuels since 1977,
- at La Hague:
  - the UP2 plant commissioned in 1966 for GCR fuels and adapted in 1976 and 1977 to LWR and FBR fuels,
  - the UP3 plant commissioned in 1989/1990 for LWR fuels.

These plants have been designed to meet all the needs of the French EDF utility, as well as the needs of a number of European and Japanese electrical utilities.

The tonnages reprocessed by these three plants as of 1 January 1991 are summarized in the table below.

TABLE I
Tonnages

	Marcoule	La Hague		
fuels	UP1 (1977)	UP2 (1966)	UP3 (1990)	Total
GCR (including foreign)	4300[1] (1210)	(260)		9200 (1470)[2]
LWR (including foreign)		3200 (2135)	200 (220)	3420 (2355)[3]
heavy water	20			20
FBR		10		10

[1]This table does not include plutonium-generating fuels reprocessed by UP1 since 1958.

[2]From the Spanish Vandellos power plant (Hifrensa).

[3]From Japan, Germany, Switzerland, Belgium, the Netherlands and Sweden.

The scale and variety of the experience thus built up in France make Cogema the uncontested world leader in reprocessing.

Let us now see what spent fuel reprocessing represents and what it serves to accomplish.

- Recovery of the energy materials uranium and plutonium contained in the spent fuel, and their recycle to light water reactors or fast breeder reactors.
- Separation of the different unusable radioactive elements or materials produced by the nuclear reactions in the reactors, in accordance with their respective characteristics, and their packaging in suitable form for final disposal in optimal safety conditions.

We shall now discuss the second aspect, by first identifying the wastes generated by reprocessing, and then describing the conditioning techniques applied in France, followed by the balances and results recorded so far, and finally the future guidelines set in this area.

WASTES GENERATED BY REPROCESSING

The three classes of unusable radioisotopes produced in the reactors and present in spent fuel assemblies are the following.

- Fission products, short- or medium-lived ß emitter radioisotopes. They are present in the spent fuel pellets. They account for about 3% of the initial fuel weight.
- Transuranium elements other than plutonium (Np, Am, Cm), long-lived a emitter radioisotopes. They are present in the spent fuel pellets, and they account for 0.07% of the initial fuel weight.
- Activation products, short- or very short-lived ß emitter radioisotopes. They are present in the metal hulls and end fittings of the fuel assemblies.

Due to the operation of the reprocessing plant, these radioisotopes are found distributed between wastes (concentrated solutions and solids) and effluents (dilute solutions or gases). Before being discharged into the environment, these liquid and gaseous effluents are treated in-line to trap most of the radioisotopes contained. The processes used are the following.

- Gaseous effluents are treated in packed columns, sprayed with basic solutions. Iodine 129 is trapped. However, due to the release authorizations in force in France for the time being, the trapping of krypton 85 is not carried out.
- The liquid effluents, to which the gaseous effluent washing solutions are added, are treated:
  - at Marcoule by a process including evaporation followed by overall neutralization of the concentrate, producing medium-level radioactive sludges that are settled,
  - at La Hague, by a process of selective coprecipitation of the different radioisotopes, which also produces radioactive sludges that are settled.

Thus the treatment of gaseous and liquid effluents leads to the production of radioactive sludges which represent an additional waste.

On the whole, accordingly, the following wastes are produced by reprocessing and effluent treatment operations.

- Concentrated solutions containing most (> 99.7%) of the fission products and transuranium elements.
- Radioactive sludges from effluent treatment and containing small amounts (< 0.1%) of FP, TRU, U and Pu.
- Fuel assembly hulls and end pieces containing activation products and traces of FP (< 0.2%), TRU, U and Pu.
- Miscellaneous 'technological' solid wastes (used equipment and tools, protective clothing) containing traces of various categories of radioisotopes.

It is customary in these wastes to distinguish between:

- those coming from the cells housing the process equipment and which may contain a significant amount of long-lived emitters (zone 4),
- those from the maintenance and servicing zones which contain little or no long-lived emitters (zones 2 and 3).

WASTE CONDITIONING PROCESSES

For each of the wastes described above, conditioning operations consist in first fixing the sorted radioisotopes in a suitable matrix, and then packing them in appropriate containers.

In France, three types of matrix have been adopted and approved by the safety authorities after many years of research, tests and characterization, conducted by the CEA, and after the industrial experience gained in Cogema's plants:

- glasses for very high-level concentrates,
- bitumens for effluent treatment sludges,
- cements for assembly hulls and end fittings and technological wastes.

The processes employed in Cogema's plants producing the packaged wastes approved by the safety authorities are described below.

Vitrification of Very High-level Concentrated Solutions

The process used in France has been described on several occasions. It was designed and developed by the CEA at Marcoule from 1957 on. It has been in industrial operation since 1978.

In this process, the concentrates are converted to vitrified products in two steps:

- evaporation of the concentrates and continuous calcination in a resistance-heated rotary furnace, producing, after nitrate decomposition, a fission product oxide powder,
- fabrication of a glass by reaction of these oxides with a prefabricated frit (sodium borosilicate) in an electric induction furnace at 1100 °C.

The glass produced is poured in batches (at the rate of two castings for one canister) in refractory steel canisters

with an effective capacity of about 150 liters, representing 400 kg of glass.

After the glass is solidified and cooled, the canister is fitted with an electronically-welded lid and then subjected to external inspection, decontaminated if necessary, and transferred to vertical interim storage shafts provided with air circulation to cool the canisters.

For each vitrification line, installed on series on the gas outlet from the calcination furnace, are a cyclon which recycles the fines to the head of the furnace, a scrubber and a condenser.

The capacity of a continuous vitrification line as described above is one glass canister per day. Two facilities are in industrial operation today in France:

- at Marcoule: the AVM facility of the UP1 plant commissioned in June 1978, with one vitrification line and interim storage capacity for 2200 canisters,
- at La Hague: the R7 facility of the UP2 plant commissioned in June 1989; with three vitrification lines in parallel and an interim storage capacity of 4500 canisters.

At La Hague, a second vitrification facility, the T7 facility of the UP3 plant, is also undergoing completion. Like R7, it has three vitrification lines and a preliminary interim storage capacity of 3600 canisters, which can be expanded as needs arise.

Note also that the AVM process was selected by the BNFL for its vitrification facility at Sellafield. It has two vitrification lines and is preparing for active commissioning.

Bituminizing of Effluent Treatment Sludges

The process used in France was designed by the French Atomic Energy Commission (CEA). It has been operating industrially at Marcoule since 1966.

It is based on the following principle: the settled sludges are introduced at the same time as bitumen and an emulsifier into an extruder heated by steam to around 12 °C and equipped with four helicoidal screws to perform evaporation, mixing of the sludge with the bitumen, and to advance the bitumen/waste mix along the extruder tube.

The bitumen coated waste (BCW) is poured at the exit in two stages into a 210 liter stainless steel drum. After the BCW is cooled and solidified, the drum is fitted with a crimped lid and transferred to an interim storage cell located near the bituminizing facility.

The evaporation products are sent to a condenser.

Two bituminizing facilities are currently in operation today at the Cogema sites:

- at Marcoule: a facility started in 1966 equipped with a bituminization line with an interim storage capacity of 50,000 drums divided into thirteen cells,
- at La Hague: one facility started in 1989 (equipped with two bituminizing lines in parallel) capable of producing 4000 drums/year and with an interim storage capacity of 20,000 drums.

Cement Embedding of Hulls and End Fittings of LWR Fuel Assemblies

When reprocessing began on LWR fuels at La Hague in 1976, France did not yet have an approved packaging process for hulls and end fittings. These wastes were accordingly stored temporarily in bulk under water in a concrete silo, and then in sealed drums stored in ponds.

Meanwhile, the CEA developed a cement embedding process. Based on this process, Cogema built the first packaging facility in the UP3 plant and secured approval for the specifications of the corresponding containers.

The process operates according to the following principle.

In the shearing unit, the end fittings of spent fuel assemblies are first separated and then rinsed in two stages in a batch washer. Pieces of clad or hulls, after dissolution of the nuclear material, are washed in countercurrent flow in a continuous helicoidal washer.

The hulls and end fittings are then combined and introduced into a perforated basket placed in a 1600 liter stainless steel drum. The drum is then transferred to a fissile material counting containment.

A cement grout of predetermined quality is then injected between the basket and the wall of the drum in order to embed all the materials.

The system can perform the cement embedding of a drum in about 20 minutes.

After drying, the drum is fitted with a lid and transferred to a decontamination pit for transfer to interim storage on the site.

An embedding installation of this type is in industrial operation since August 1990 at the head end of the UP3 plant. The interim storage capacity at La Hague is 2400 drums for the time being, divided into three modules.

Cement Embedding of Technological Wastes

"Technological wastes" include:

- used equipment and materials (pumps, valves),
- work clothes and materials (gloves),
- laboratory equipment (glassware, sampling jugs),

- miscellaneous materials (cotton, vinyl, card/board, wood).

These objects are sorted and collected in each facility, in bins, drums or canisters transportable under an MERC (mobile equipment removal containment).

They are then conveyed to the packaging facility which produces two main classes of cemented technological wastes:

- materials consisting of wastes produced in the process cells of the plant, normally inaccessible to the operating personnel (zone 4),
- materials consisting of wastes produced in the plant service zones accessible to operating personnel (zones 2 and 3).

The main operations carried out at the La Hague plant are the following.

- Conditioning of zone 4 wastes
- Reception of the wastes in their collection packages (bins, drums, canisters).
- Weighing and measurement of the activity contained;
- Placement in a prefabricated asbestos-cement containment (CAC) with an overall volume of 1ß2 m^3.
- Positioning of the lid on the CAC.
- Injection of cement grout after drying and of a finishing product.
- Weighing, measurement of gamma dose rate and inspection for absence of surface contamination.
- Interim storage at site facility with a capacity of 16,400 CAC.
- Conditioning of zone 2 and 3 wastes
- Reception of wastes collected in 100 or 200 liter drums.
- Weighing and measurement of the activity contained.
- Compacting of the 100 liter drums and placement in 200 liter drums.
- Placement of the 200 liter drums in a prefabricated Cylindrical Fiber Concrete container (CFC) with an overall volume of 0.7 m^3.
- Placement of a lid and injection of fiber concrete of the same grade as that of the container.
- Drying, weighing, measurement of the gamma dose rate, and inspection for absence of surface contamination.
- Interim storage on the site facility with a capacity of 16,000 CFC.

CHARACTERISTICS AND SPECIFICATIONS OF PACKAGED WASTES

Table II below gives the main characteristics of the packaged wastes as produced at the La Hague plant.

Following lengthy characterization studies conducted by the CEA, Cogema drafted technical specifications for each of these packaged wastes, including a number of guaranteed parameters, in agreement with the Fundamental Safety Rules published by the French Authorities. They were initially submitted for approval to the French Safety Authorities, and then, for wastes from abroad, to the different electrical utilities concerned, for the approval of their respective Safety Authorities.

A technical audit procedure for the processes applied for these specifications was set up on behalf of the French and foreign clients using the services of Bureau Veritas.

INDUSTRIAL RESULTS OBTAINED IN FRANCE AND FUTURE PROCESS DEVELOPMENTS

The industrial experience built up so far by the two facilities at Marcoule and La Hague is summarized in the Table III below.

After interim storage for a few years at the reprocessing sites, these different packaged wastes can be transported to final disposal installations at the surface or in deep repositories.

These transportation programs are scheduled:

- for wastes belonging to foreign utilities, from 1994, in application of contracts in force,
- for those belonging to the French EDF electrical utility, as of now for wastes intended for shallow-ground disposal, and from 2010 for wastes intended for deep disposal.

In this respect, it may be recalled that, in accordance with French policy, the following classes of wastes will be intended for deep disposal:

- category C: wastes with very high ß activity and containing long-lived radioisotopes (vitrified products),
- category B: high-, medium- and low-level wastes containing long-lived radioisotopes:
  - bituminized waste products,
  - hulls and end pieces,
  - zone 4 technological wastes.

As to the second category (category B wastes), the Cogema policy set for the future is to minimize the volumes for disposal. Accordingly, R and D is currently under way on new processes, both at the CEA and Cogema, including:

TABLE II
Conditioned Wastes
(La Hague Plant)

WASTE		MATRIX	PACKAGE		CONDITIONNED RESIDUE		MAXIMUM ACTIVITY (Ci)		MAXIMUM THERMAL RELEASE
NATURE	TYPE		TYPE (Nb/ tHM)	CAPACITY (l)	PAY LOAD (kg)	TOTAL WEIGHT (kg)	ALPHA	BETA	
FISSION PRODUCTS CONCENTRATES (1)	C	GLASS	stainless steel container (0,75)	150	400	480	3 800	800 000 (FP)	2 980 W
SLUDGES (2)	B	BITUMEN	stainless steel DRUM (2,5)	200	280	300	4	500 (FP)	2 W
HULLS AND END FITTINGS (3)	B	CEMENT	stainless steel container (0,5)	1 300	3 600	4 000	5	3 000 (FP) 12 000 (AP)	130 W
TECHNOLOGICAL WASTES ZONE 4 (4)	B		CBFC 2 (1)	1 200	2 400	3 500	10	50	
TECHNOLOGICAL WASTES ZONES 2 ET 3 (5)	A		CBFC 1 (5,2)	650	1 100	1 800	0,1	20	

NOTA : CBFC = cylindrical fiber concrete container
FP = fission products
AP = activation products.

Specifications : (1) 300 AQ 016 (7/86)
(2) (in progress)
(3) 300 AQ 025 (1/90)
(4) 300 AQ 038 (1/90)
(5) 300 AQ 034 (1/90)

- in reprocessing, design of internal recycles avoiding or reducing the transmission of long-lived emitters to effluent treatment, i.e. ultimately to the bituminized waste products,
- reduction of the volume of hulls and end pieces by fusion or compaction,
- reduction of zone 4 technological wastes by intensive decontamination of the equipment and possible re-use,
- large-capacity incinerator for plutonium-rich wastes and ash recycling.

The range of processes in operation today in the two French plants will also be supplemented with new processes as soon as they are fully developed:

- cement embedding of ion exchange resins,
- cement embedding of magnesium clads of GCR fuels,
- large-capacity incinerator for ß/γ wastes.

CONCLUSION

In its two facilities at Marcoule and La Hague, Cogema, with the support of the CEA and the approval of the French Safety Authorities, has gradually developed a diversified set of packaging processes covering the different types of waste generated by reprocessing operations.

Packaging facilities employing the following processes are currently operating in-line today, near or in its UP1, UP2 and UP3 plants in operation:

- vitrification for very high-level fission product concentrates,
- bituminizing for effluent treatment sludges,
- cement embedding for all other solid wastes (hulls and end pieces, miscellaneous technological wastes).

All these packaged wastes are produced within the strict obligation of guaranteed technical specifications, accepted by Cogema's different French and foreign clients, and approved by their respective Safety Authorities.

In application of its contractual agreements with its customers, Cogema thus enjoys the position of being able to return the packaged wastes to their countries of origin as soon as the different final disposal installations of these countries have been commissioned.

TABLE III
Industrial Experience

Process	Unit	Marcoule	La Hague
Vitrification			
FP concentrate volume	m^3	1435	518
activity	10^6 Ci	291	177
glass canisters	qty	1830	520
glass weight	t	640	260
Bitumen coating			
bitumen drums	qty	59,865	2096
Cement coating			
hulls and fittings, drums	qty	-	45
100 liter compacted drums	qty	720,000	7900
200 liter cemented drums	qty	150,000	-
fiber concrete containers (CBF)	qty	-	1364
asbestos-cement containers (CAC)	qty	-	22