New designs: the package at the core of investigations



Safe conditionings obtained through constantly improved processes

Current waste conditioning processes have demonstrated their effectiveness on an industrial scale. Major advances have nonetheless been achieved, with the development of processes having broader applications, and the ability to achieve further waste volume reductions yet.



Artistic presentation of a glass block. After investigation of a variety of solid crystalline compounds, borosilicate glass was selected for the confinement of the thirty-odd radionuclides found in the fission product solutions yielded by spent fuel reprocessing.

> Waste conditioning operations consist in treating that waste to bring it to an ultimate form that is suitable for its future management. The final waste package ensures the conditioning of waste in a stable, strong, monolithic form, with mechanical properties meeting the requirements of handling, transport, storage, and disposal (see Box C, *What stands between waste and the environment*?).

> A number of conditioning processes and matrices are being investigated by CEA, and used on an industrial scale in France, but equally around the world. These include *cementation* for sludges, evaporator concentrates, ashes from incineration, ion-exchange resins...; *bituminization*, in particular for the **encapsulation** of sludges and evaporator concentrates from the treatment of **liquid effluents**; and *vitrification*, for the **confinement** of **fission product** (FP) solutions.

Cementation, a long-established and widespread process

Cementation is the oldest, and most widespread, conditioning process in France, and in other countries, for low- and intermediate-level waste (LILW) generated by nuclear power plants, **spent fuel repro**- **cessing** facilities, and research centers. Cements, indeed, combine a number of favorable factors: availability, low cost, simplicity of use, high mechanical strength, and, in most cases, stability over time. Furthermore, the high internal pH of cement systems allows to precipitate and thus confine many **radionuclides**.

Cementation is used to immobilize bulky solid waste items, and to encapsulate liquid and wet solid waste, such as evaporator concentrates, chemical co-**precipitation** sludges produced by liquid effluent treatment stations, ion-exchange resins used for pool water filtration, powdered waste yielded by the reprocessing of spent fuel from the **UNGG pathway** (see Box 1 in *Radioactive waste management research: an ongoing process of advances*)...

Cementation is generally carried out close to the waste producing site, either by *in-container mixing*, or through *continuous processes*. In the former process, cement, waste and additives are separately fed into the **container**, mixed in order to obtain a homogeneous mixture, then left to stand until set. The container is then capped, sealed, tested, and, if required, decontaminated, before it is transported to a storage facility. With the second method, the cement, waste, and additives are dosed separately, and fed

into a mixer. The mixture thus obtained is then poured into the container. Once full, the container is sealed, decontaminated, and transported to the storage facility (see Figure 1).

Research work on encapsulation matrices is carried out, for the most part, in collaboration with waste producers, with a view to increase waste incorporation rate, and improve confinement performance of the cementitious materials, bearing in mind two factors: the significant increase in waste volume, after conditioning (as a rule, this is multiplied by a factor close to 2), and possible interactions between waste constituents and cement phases, which may perturb cement hydration, and thus impair the durability of the materials obtained.

To remedy such drawbacks, new cement **formulations** of the sulfoaluminate type, exhibiting better compatibility with certain types of waste than conventional calcium silicate cements, have been investigated. Sulfoaluminate cements are obtained in two steps, first calcination, at 1,300–1,350 °C, of limestone, bauxite, gypsum, and possibly recoverable industrial waste (fly ash, blast-furnace slag...), then intergrinding with gypsum. Through hydration, they mostly lead to formation of ettringite (3CaO.Al₂O₃.3CaSO₄.32H₂O) and/or hydrated calcium monosulfoaluminate (3CaO.Al₂O₃.CaSO₄.12H₂O), and in smaller amounts, hydrated calcium silicates and aluminates.

Waste from the treatment of liquid effluents generally exhibits a complex chemistry, and may contain high concentrations of borates $(B(OH)_{\overline{4}} \text{ ions})$ and sulfates $(SO_4^2 \text{ ions})$. Encapsulation of such waste in common calcium silicate cement results in major difficulties. Indeed, the borates inhibit binder setting, and sulfates may cause swelling and cracking of the hardened material, owing to delayed formation of ettringite. It is thus often necessary to carry out pretreatment of the waste so as to restrict its further interaction with the cement. The complexity of the process is increased, as well as the volume of waste requiring conditioning owing to the addition of pretreatment reactants.

Non-expansive sulfoaluminate cements could offer an interesting alternative since they form at early age significant amounts of ettringite and/or calcium monosulfoaluminate, these having the ability to incor-



Cemented waste package.



Figure 1.

Schematic of the ion-exchange resin cementation unit. After settling, the resins are transferred to the batch mixer, and mixed with cement. The encapsulated waste is then discharged into the container through a gate located under the mixer.

porate waste sulfates and borates into their structure. Laboratory trials, carried out with a sulfoaluminate cement as partial substitute for common cement and a synthetic waste, consisting in chemical co-precipitation sludge mixed with an evaporator concentrate, with respective sulfate and boron concentrations of 16 g/L and 5.5 g/L, have led to significant advances. The borates do not inhibit binder setting, the waste may be cemented with no prior treatment, and waste incorporation rate can be multiplied by a factor 1.8 (i.e. 56 wt%). Further, 90% of the sulfates and boron from the waste remain trapped in the solid fraction, as evidenced by a leaching test carried out at ambient temperature on ground encapsulated waste, thus demonstrating their satisfactory participation in binder hydration reactions. Use of this type of cement as a substitute for common cements does not require alteration of existing cementation facilities. This path will thus be further explored, to improve formulation of the materials, extend their application domain, and evaluate longterm behavior of the encapsulated waste. These cements could, in particular, offer interesting prospects for the conditioning of heavy metals.

Bituminization, a proven process

Bituminization, widely used in France, for in-line conditioning of chemical co-precipitation sludges, yielded by effluent insolubilization treatment, or of evaporator concentrates, resulting from heat treatment of effluents, is a proven process, benefiting from considerable feedback. This consists in mixing, in hot conditions, waste, in the form of sludge, with bitumen. The mixture obtained is dehydrated, and poured into a container, in which it is cooled. Bitumen encapsulation ensures at the same time dehydration of the waste, its homogeneous dispersion, and immobilization of radionuclides within the matrix. This process was developed in France as early as the 1960s. Industrial deployment of the process in line was

New designs: the package at the core of investigations



Bituminized sludge packages contain chemical co-precipitation sludges incorporated in bitumen, and placed in a metal drum.



achieved at Marcoule in 1966, in the effluent treatment station, and in 1989 at La Hague, in the STE3 facility (see Figure 2).

The bitumen matrix is manufactured through distillation of crude oil. Two types of bitumen, essentially, are employed: pure, or direct-distillation, bitumens, yielded by refining crude oil, and oxidized, or blown, bitumens, obtained through oxidation, in an air-blowing process, of heavy oils, or petroleum refining residues. To date, tens of thousands of bitumen drums have been produced, mainly at Marcoule and La Hague, for the bituminization of chemical co-precipitation sludges, using a direct-distillation bitumen.

In the context of an effluent management policy seeking to minimize the number, and diversity, of waste

> packages produced, bituminization, nowadays, is restricted to sodium effluents (i.e., containing sodium [Na]), and miscellaneous laboratory effluents, incompatible with vitrification or cementation.

Other specific applications of bituminization, drawing on the industrial experience gained, are being deployed. Thus, operation, between 1966 and 1989, of the first effluent treatment station at La Hague, the STE2 facility, generated some 9,300 m3 of sludges, stored in silos, while awaiting conditioning. Bearing in mind the chemical similarities between sludges treated in the STE2 and STE3 facilities, the positive feedback from operation of STE3, and the proven confinement qualities of bitumen, bituminization is currently being considered for the conditioning of this legacy waste (see Nuclear waste management and processing: between legacy and anticipation).

A major R&D effort has thus been initiated, to demonstrate mastery of process safety, and optimize the amounts of activity that may be incorporated in packages, to reduce the number of packages to be produced. The first goal resulted in development of a robust procedure, making it possible to preclude any risk of an incident during package fabrication and cooling. Indeed, the presence of oxidizer and reducer compounds may lead, during drum cooling, to initiation of exothermic reactions within the encapsulated waste. The main compounds that are reactants at encapsulation temperatures, cobalt salts and nitrates, have been identified, and reaction kinetics ascertained. Concurrently, drum cooling was modeled, using penalizing values as input data, to achieve an "envelope" simulation of the maximum temperature reached inside the drum. These investigations have made it possible to guarantee, in all cases, a safe domain of operation for the process.

Due to the effects of alpha, beta, and gamma radiation, bitumen undergoes transformations, taking the form of alterations in the hydrocarbon molecules, concomitant with release of radiolysis gases, mainly hydrogen, which may cause swelling of the encapsulant. The quantity of hydrogen formed depends on the activity held by the encapsulant. Empirical investigations have highlighted a significant fall in hydrogen production in the presence of cobalt salts.

eva/La Hague



Figure 2

Schematic of Areva's bituminization encapsulation unit at La Hague. This facility comprises a preparation and sludge feed unit, a surfactant tank, a bitumen production and supply installation, a Werner extruder (shown here), a pouring station, a drum filling station, and fume and distillate treatment equipment.

After a stage of ascertaining the mechanism, and empirically determining the trapping capacity of cobalt salts, the optimization of, and control of conditions for, fabrication of encapsulants **doped** with cobalt salts was successfully achieved, on various scales (laboratory, semi-industrial, industrial). This verified trapping process could thus be taken on board, for the determination of maximum allowable activity for bituminized sludge packages. This has allowed a reduction by a factor of about 3 of the number of drums to be produced.

Taken together, these findings form the support for the experimental campaign of bituminization of STE2 sludges, initiated at the end of 2005 by **Areva**.

Vitrification, a mature process

From the late 1950s, it was found indispensable to condition high-level waste (**HLW**) yielded by spent fuel reprocessing operations. Indeed, such FP solutions, while they only account for a limited volume, bear 96% of the **radioactivity**. Quite swiftly, the glass matrix won acceptance for the conditioning of such waste, both in France and in other countries (United States, Russia, the United Kingdom, Germany, Japan...). Indeed, glass confines radionuclides, not through encapsulation, but by the setting up of chemical bonds with the oxide constituents forming the glass's **amorphous** structure.

An initial industrial vitrification line was thus put on stream at Marcoule in 1978 (AVM: Atelier de vitrification de Marcoule – Marcoule Vitrification plant), for the conditioning of FP solutions yielded by the reprocessing of spent fuel from UNGG reactors. Six other lines are in operation at present, in the R7 and T7 facilities at La Hague, starting in 1989 and 1992 respectively, for the vitrication of FPs from **pressurized-water reactors**. These facilities allow vitrification of FP solutions yielded by current spent fuel reprocessing activities, and stand as a world industrial reference.

The continuous vitrification process, currently operated on an industrial basis, comprises two distinct steps. A *step of calcination* of liquid FP solutions, at around 400 °C, is followed by a *melting step* at around 1,100 °C, in an induction-heated metal crucible, in which the vitrification additive (glass frit) and calcined FP solution (calcine) are mixed, prior to pouring into a metal container. The **glass** thus fabricated holds between 12 wt% and 18 wt% HLW waste (see Figure 3).

Over 15 years, operation on an industrial basis of these facilities has brought a unique operational feedback, demonstrating the maturity of the French vitrification process. Materials and processes, however, needs must go on being adjusted to meet requirements, such as optimization of the number of glass packages, with regard to the changing characteristics of the fuel being reprocessed (high burnup rates, accompanied by rising alpha-emitter content in glasses), confinement of FP solutions yielded by the reprocessing of legacy fuel, and treatment of intermediate-level waste (ILW-LL)... (see Industrial solutions for long-lived, high- and intermediate-level waste). To meet such requirements, experiments with doped glasses are being carried out, and new glass formulations, optimized for the nature of the waste to be $\frac{d}{d}$



Figure 3. Current two-step vitrification process.

conditioned, together with new vitrification technologies, allowing access to higher melting temperatures, are being developed.

Direct-induction cold-crucible furnace technology

Compared with the current process, the cold-crucible melting technology brings major benefits. First of all, cooling the melting furnace allows a thin layer of solidified glass to form, protecting the crucible and precluding corrosion from the molten glass. Second, heating by direct induction within the molten glass bath allows higher fabrication temperatures to be achieved, thus enabling design of novel, higher-performance waste-confinement matrices (see Box).

Thus, a glass ceramic matrix, which may be fabricated at 1,250 °C, having the ability to hold up to 13 wt%,



Pouring glass fabricated by the current two-step vitrification process into a container.



Direct-induction cold-crucible vitrification

Direct-induction melting consists in placing the glass that is to be heated inside an alternating electromagnetic field, generated by an inductor. This field causes induced currents, which dissipate energy by the Joule effect inside the material, resulting in its melting. With direct induction, the point is to heat the material to be melted, directly, without heating the crucible. The latter, on the contrary, is cooled by water circulation, and sectorized, to ensure relative transparency to the electromagnetic field.





The technological test platform set up in 2001 by CEA and Areva, to demonstrate the feasibility of the advanced cold crucible technology (CFA: *creuset froid avancé*). **()** cold crucible (diameter: 1.1 m) **(2)** pouring glass station **(3)** gas treatment equipment.



Cold crucible, developed for Areva industrial applications, at La Hague.

of molybdenum oxide (MoO₃), and 4 wt% phosphorus oxide (P₂O₅), was developed for the conditioning of UMo solutions, yielded by the reprocessing of UNGG fuel, consisting in uranium metal, alloyed with molybdenum. Likewise, glass formulations of the **rare-earth** borosilicate type, lending themselves to fabrication at 1,250 °C, and exhibiting the ability to contain up to 22 wt% of waste, are being investigated for high-burnup fuel. Concurrently, demonstrations of glass formulation feasibility, and of mastery of the cold-crucible melting process have been carried out, for the treatment of HLW effluents in disposal at the Hanford (United States), Guangyuan (China), Aldermaston (United Kingdom), and Saluggia (Italy) sites. A tripartite working group, bringing together Cogema, SGN and CEA, has specified the main technological design options for a pilot cold crucible suited to a high-activity-level nuclear environment. A replica of the model to be installed at La Hague, by 2010, on a vitrification line, to replace existing technology, will soon be functioning in the R&D halls at Marcoule.

The next stage in the evolution of the vitrification process consists in eliminating the calcination step, with the direct feeding of the FP solution to the glass bath. This will contribute to minimization of secondary waste, and greater ease of maintenance inside a shielded cell. To ensure evaporation, drying, calcination and melting directly within the furnace, its diameter must be increased. A new large-diameter cold-crucible furnace design is being developed, and a test platform, including an Advanced Cold Crucible (CFA: Creuset froid avancé) of 1.1 m diameter was put in service in 2001. The increased furnace surface means a higher capacity, and thus the ability to treat, in conditions compatible with industrial operation, ILW-LL waste, taking up a larger volume as it does than FP solutions.

Incineration-vitrification processes

The advantages of vitrification may be extended to other types of waste, in particular incinerable waste, by combining burning the waste and the subsequent vitrification of the ashes into one single process. Incineration–vitrification processes consist, on the one hand, in burning the incinerable materials over a molten glass bath, and, on the other, in incorporating the oxidized ashes into the glass bath, thus ensuring durable confinement of the radionuclides initially held in the material.

The SHIVA process (Système hybride d'incinération–vitrification avancé – Advanced Hybrid Incineration–Vitrification System), consisting in a cold crucible fitted with a plasma torch above the glass bath, has been developed (see Figure 4). The oxygen plasma allows better materials incineration, in particular the oxidation of metal constituents that may occur in the waste, this subsequently enhancing the reactivity of the ashes with the vitrification additive. Combination of the two heating modes (plasma torch, and cold crucible) thus allows separate management of the incineration and vitrification functions, within one reactor, and brings flexibility of operation.

This process could be used for the incineration–vitrification of **technological waste**, such as heavily loaded ion-exchange resins, plastics and cellulose, or concentrated solutions bearing salts compatible with a glass formulation.

Conditionings for the entire range of waste types

Since the 1960s, processes have been developed for waste conditioning in a number of matrices, and have been successfully deployed on an industrial scale. Over the past decade, investigations on improvements to existing conditioning modes have resulted in the development of processes to cater for all types of waste, while optimizing the volume of waste produced.

Thus, new cement formulations, allowing management of waste incompatible with current cements, have been specified. The adaptation of the bituminization process for the conditioning of legacy sludges, at La Hague, makes it possible to contemplate the retrieval, on an industrial basis, of these sludges. A cold-crucible melting furnace for high-level solutions is to be installed, around 2010, on a vitrification line of the R7 facility, at La Hague. This process allows the fabrication of new glass formulations, whether it be for the retrieval of legacy waste, or for future waste. A compact incineration–vitrification process has been developed, and a pilot is in place at Marcoule.

These investigations are carried out in constant collaboration with waste producers, **Areva** in particular, to provide support for the changes in plants, and in the nature of the waste to be conditioned.

> > Catherine Fillet, Céline Cau Dit Coumes, Christophe Joussot-Dubien and Roger Boën Nuclear Energy Division CEA Valrhô-Marcoule Center



Figure 4.

In the SHIVA process, association of a cold crucible and a plasma torch positioned above the glass bath allows separate management of the incineration and vitrification functions in one reactor.



The SHIVA advanced hybrid incineration-vitrification system was developed for the treatment of an even wider range of waste.

A What is radioactive waste?

ccording to the International Atomic A Energy Agency (IAEA), radioactive waste may be defined as "any material for which no use is foreseen and that contains radionuclides at concentrations greater than the values deemed admissible by the competent authority in materials suitable for use not subject to control." French law in turn introduces a further distinction, valid for nuclear waste as for any other waste, between waste and final, or "ultimate," waste (déchet ultime). Article L. 541-1 of the French Environmental Code thus specifies that "may be deemed as waste any residue from a process of production, transformation or use, any substance, material, product, or, more generally, any movable property left derelict or that its owner intends to leave derelict." further defining as ultimate "waste, be it the outcome of waste treatment or not, that is not amenable to further treatment under prevailing technological and economic conditions, in particular by extraction of the recoverable, usable part, or mitigation of its polluting or hazardous character."

Internationally, experts from IAEA and the Nuclear Energy Agency (NEA) – an OECD organization – as those in the European Commission find that long-lived waste produced in countries operating a nuclear power program is stored securely nowadays, whilst acknowledging a final solution is required, for the long-term management of such waste. They consider burial in deep geological structures appears, presently, to be the safest way to achieve final disposal of this type of waste.

What constitutes radioactive waste? What are the volumes currently involved?

Radioactive waste is classified into a number of categories, according to its level of radioactivity, and the radioactive **period**, or **half-life**, of the radionuclides it contains. It is termed **long-lived waste** when that period is greater than 30 years, **short-lived waste** otherwise. The French classification system involves the following categories:

 very-low-level waste (VLLW); this contains very small amounts of radionuclides, of the order of 10–100 Bq/g (becquerels per gram), which precludes considering it as conventional waste;

short-lived low and intermediate level
waste (LILW-SL); radioactivity levels for
such waste lie as a rule in a range from

a few hundred to one million Bq/g, of which less than 10,000 Bq/g is from longlived radionuclides. Its radioactivity becomes comparable to natural radioactivity in less than three hundred years. Production of such waste stands at some 15,000 m³ per year in France;

 - long-lived low-level waste (LLW-LL); this category includes radium-bearing waste from the extraction of rare earths from radioactive ore, and graphite waste from first-generation reactors;

- long-lived intermediate-level waste (ILW-LL), this being highly disparate, whether in terms of origin or nature, with an overall stock standing, in France, at 45,000 m³ at the end of 2004. This mainly comes from spent fuel assemblies (cladding hulls and end-caps). or from operation and maintenance of installations; this includes, in particular, waste conditioned during spent fuel reprocessing operations (as from 2002, this type of waste is compacted, amounting to some 200 m³ annually), technological waste from the operation or routine maintenance of production or fuel-processing plants, from nuclear reactors or from research centers (some 230 m³ annually), along with sludges from effluent treatment (less than 100 m³ annually). Most such waste generates little heat, however some waste of this type is liable to release gases;

- high-level waste (HLW), containing fission products and minor actinides partitioned during spent fuel reprocessing (see Box B), and incorporated at high temperature into a glass matrix. Some 120 m³ of "nuclear glass" is thus cast every year. This type of waste bears the major part of radioactivity (over 95%), consequently it is the seat of considerable heat release, this remaining significant on a scale of several centuries.

Overall, radioactive waste conditioned in France amounts to less than 1 kg per year, per capita. That kilogram consists, for over 90%, of LILW-SL type waste, bearing but 5% of total radioactivity; 9% of ILW-LL waste, less than 1% HLW, and virtually no LLW-LL waste.

What of the waste of tomorrow?

From 1991, ANDRA compiled, on a yearly basis, a geographical inventory of waste present on French territory. In 2001, ANDRA was asked by government to augment this "National Inventory," with the threefold aim of characterizing extant stocks (state of conditioning, processing traceability), predicting future waste production trends to 2020, and informing the public (see An inventory projecting into the *future*). ANDRA published this reference National Inventory at the end of 2004. To meet requirements for research in compliance with the directions set out in the French Act of 30 December 1991 (see Radioactive waste management research: an ongoing process of advances), ANDRA, in collaboration with waste producers, has drawn up a Dimensioning Inventory Model (MID: Modèle d'inventaire de dimensionnement), for the purposes of arriving at estimates of the volume of waste packages to be taken on board in research along direction 2 (disposal). This model, including as it does predictions as to overall radioactive waste arisings from the current reactor fleet, over their entire lifespan, seeks to group waste types into families, homogeneous in terms of characteristics, and to formulate the most plausible hypotheses, with respect to conditioning modes, to derive the volumes to be taken on board for the purposes of the investigation. Finally, MID sets out to provide detailed stocktaking, intended to cover waste in the broadest possible fashion. MID (not to be confused with the National Inventory, which has the remit to provide a detailed account of actual waste currently present on French territory) thus makes it possible to bring down the variety of package families to a limited number of representative objects, and to specify the requisite margins of error, to ensure the design and assessment of disposal safety will be as robust as feasible, with respect to possible future variations in data.

To ensure consistency between investigations carried out in accordance with direction 2 and those along direction 3 (conditioning and long-term storage), CEA adopted MID as input data. MID subsumes waste packages into standard package types, then computes the number and volume of HLW and ILW-LL packages, according to a number of scenarios, all based on the assumption that current nuclear power plants will be operated for 40 years, their output plateauing at 400 TWhe per year.

Table 1 shows the numbers and volumes for each standard package type, for the scenario assuming a continuation of current strategy, with respect to spent fuel reprocessing: reprocessing of 79,200 UOX fuel assemblies and storage of 5,400 MOX

MID standard package types Symbols Producers Categories Number Volume (m³) CO — C2 HLW 42,470 Vitrified waste packages Cogema* 7,410 B1 EDF ILW-LL 2,560 470 Activated metal waste packages B2 CEA, Cogema* ILW-LL 105,010 36,060 Bituminized sludge packages 27.260 CEA, Cogema* ILW-LL 32.940 Cemented technological waste packages B3 R/ ILW-LL 2,730 Cemented hull and end-cap packages Cogema* 1.520 R5 ILW-LL 39,900 7,300 Compacted structural and technological waste packages Cogema* B6 ILW-LL 10,810 4,580 Containerized loose structural and technological Cogema* waste packages Total B 192,740 78,400 Total overall 235,210 85,810

* renamed Areva NC in 2006

A (next)

Table 1.

Amounts (number, and volume) of waste packages, as predicted in France for 40 years' operation of the current fleet of reactors, according to ANDRA's Dimensioning Inventory Model (MID).

assemblies discharged from the current PWR fleet, when operated over 40 years.

What forms does it come in?

Five types of generic packages (also found in MID) may be considered:

• cementitious waste packages: ILW-LL waste packages employing hydraulic-binder based materials as a conditioning matrix, or as an immobilizing grout, or yet as a container constituent;

• bituminized sludge packages: LLW and ILW-LL waste packages, in which bitumen is used as confinement matrix for low- and intermediate-level residues from treatment of a variety of liquid effluents (fuel processing, research centers, etc.);

• standard compacted waste packages (CSD-C: colis standard de déchets compactés): ILW-LL packages obtained through compaction conditioning of structural waste from fuel assemblies, and technological waste from the La Hague workshops;

• standard vitrified waste packages (CSD-V: colis standard de déchets vitrifiés): HLW packages, obtained mainly through vitrification of highly active solutions from spent fuel reprocessing;

• spent fuel packages: packages consisting in nuclear fuel assemblies discharged from reactors; these are not considered to be waste in France.

The only long-lived waste packages to be generated in any significant amounts by current electricity production (see Box B) are vitrified waste packages and standard compacted waste packages, the other types of packages having, for the most part, already been produced, and bearing but a small part of total radioactivity.

What is happening to this waste at present? What is to be done in the long term?

The goal of long-term radioactive waste management is to protect humankind and its environment from the effects of the materials comprised in this waste, most importantly from radiological hazards. Any release or dissemination of radioactive

	Short-lived Half-life < 30 years for the main elements	Long-lived Half-life > 30 years				
Very-low-level waste (VLLW)	Morvilliers dedicated dispos Capacity: 6	Morvilliers dedicated disposal facility (open since 2003) Capacity: 650,000 m ³				
Low-level waste (LLW)	Aube Center (open since 1992)	Dedicated disposal facility under investigation for radium-bearing waste (volume: 100,000 m ³) and graphite waste (volume: 14,000 m ³)				
Intermediate-level waste (ILW)	Capacity: 1 million m ³	MID volume estimate: 78,000 m ³				
High-level waste (HLW)	MID volume esti	mate: 7,400 m ³				

Table 2.

Long-term management modes, as currently operated, or planned, in France, by radioactive waste category. The orange area highlights those categories targeted by investigations covered by the Act of 30 December 1991.

(1) According to the Dimensioning Inventory Model (MID)

materials must thus be precluded, through the lasting isolation of such waste from the environment. This management is guided by the following principles: to produce as little waste as practicable; limit its hazardous character as far as feasible; take into account the specific characters of each category of waste; and opt for measures that will minimize the burden (monitoring, maintenance) for future generations.

As for all nuclear activities subject to control by the French Nuclear Safety Authority (Autorité de sûreté nucléaire), fundamental safety regulations (RFSs: règles fondamentales de sûreté) have been drawn up with respect to radioactive waste management: sorting, volume reduction, package confinement potential, manufacturing method, radionuclide concentration. RFS III-2.f, in particular, specifies the conditions to be met for the design of, and demonstration of safety for an underground repository, and thus provides a basic guide for disposal investigations. Industrial solutions (see Industrial solutions for all low*level waste*) are currently available for nigh on 85% (by volume) of waste, i.e. VLLW and LILW-SL waste. A solution for LLW-LL waste is the subject of ongoing investigation by ANDRA, at the behest of waste producers. ILW-LL and HLW waste, containing radionuclides having very long half-lives (in some cases, greater than several hundred thousand years) are currently held in storage installations coming under the control of the Nuclear Safety Authority. What is to become of this waste in the long term, beyond this storage phase, is what the Act of 30 December 1991 addresses (see Table 2).

For all of these waste types, the French Nuclear Safety Authority is drawing up a National Radioactive Waste Management Plan, specifying, for each type, a management pathway.

B Waste from the nuclear power cycle

Nost high-level (high-activity) radioactive waste (HLW) originates, in France, in the irradiation, inside nuclear power reactors, of fuel made up from enriched uranium oxide (UOX) pellets, or also, in part, from mixed uranium and plutonium oxide (MOX). Some 1,200 tonnes of spent fuel is discharged annually from the fleet of 58 pressurized-water reactors (PWRs) operated by EDF, supplying over 400 TWh per year, i.e. more than three quarters of French national power consumption.

The fuel's composition alters, during its irradiation inside the reactor. Shortly after discharge, fuel elements contain, on average,⁽¹⁾ some 95% residual uranium, 1% plutonium and other transuranic elements – up to 0.1% – and 4% of products yielded by fission. The latter exhibit very significant radioactivity levels – to the extent this necessitates management safety measures requiring major industrial resources – of some 10¹⁷ Bq per tonne of initial uranium (tiU) (see Figure 1).

The *uranium* found in spent fuel exhibits a makeup that is obviously different from that of the initial fuel. The greater the irradiation, the higher the consumption of fissile nuclei, and consequently the greater the extent by which the *uranium* will have been **depleted** of the fissile isotope 235 (²³⁵U). Irradiation conditions usually prevailing in reactors in the French fleet, with an average fuel residence time inside the reactor of some 4 years, for a





The main elements found in spent nuclear fuel.

burnup rate close to 50 **GWd/t**, result in bringing down final ²³⁵U content to a value quite close to that of **natural uranium** (less than 1%), entailing an energy potential very close to the latter's. Indeed, even though this uranium remains slightly richer in the fissile isotope than natural uranium, for which ²³⁵U content stands at 0.7%, the presence should also be noted, in smaller, though significant, amounts, of other isotopes having adverse effects in neutronic or radiological terms (²³²U, ²³⁶U), that had not figured in the initial fuel (see Table 1). The *plutonium* present in spent fuel is yielded by successive neutron capture and decay processes. Part of the Pu is dissipated through fission: thus about one third of the energy generated is yielded by "in situ recycling" of this element. These processes further bring about the formation of heavy nuclei, involving, whether directly themselves, or through their daughter products, long radioactive halflives. These are the elements of the actinide family, this including, essentially, plutonium (from ²³⁸Pu to ²⁴²Pu, the oddnumbered isotopes generated in part undergoing fission themselves during irradiation), but equally neptunium (Np), americium (Am), and curium (Cm), known as minor actinides (MAs), owing to the

(1) These figures should be taken as indicative values. They allow orders of magnitude to be pinpointed for enriched-uranium oxide fuel, taken from the main current French nuclear power pathway; they do depend, however, on a number of parameters, such as initial fuel composition and irradiation conditions, particularly irradiation time.

element	isotope	half-life (years)	UOX 33 GWd/tiU (E ²³⁵ U: 3.5%)		UOX 45 GWd/tiU (E ²³⁵ U: 3.7%)		UOX 60 GWd/tiU (E ²³⁵ U: 4.5%)		MOX 45 GWd/tihm (Ei Pu: 8.65%)	
			isotope content (%)	quantity (g/tiU)	isotope content (%)	quantity (g/tiU)	isotope content (%)	quantity (g/tiU)	isotope content (%)	quantity (g/tihm)
U	234	246,000	0.02	222	0.02	206	0.02	229	0.02	112
	235	7.04·10 ⁸	1.05	10,300	0.74	6,870	0.62	5,870	0.13	1,070
	236	2.34·10 ⁷	0.43	4,224	0.54	4,950	0.66	6,240	0.05	255
	238	4.47·10 ⁹	98.4	941,000	98.7	929,000	98.7	911,000	99.8	886,000
Pu	238	87.7	1.8	166	2.9	334	4.5	590	3.9	2,390
	239	24,100	58.3	5,680	52.1	5,900	48.9	6,360	37.7	23,100
	240	6,560	22.7	2,214	24,3	2,760	24.5	3,180	32	19,600
	241	14.4	12.2	1,187	12.9	1,460	12.6	1,640	14.5	8,920
	242	3.75·10 ⁵	5.0	490	7.8	884	9.5	1,230	11.9	7,300

Table 1.

Major actinide inventory for spent UOX and MOX fuel after 3 years' cooling, for a variety of enrichment and burnup rates. Burnup rate and quantity are expressed per tonne of initial uranium (tiU) for UOX, per tonne of initial heavy metal (tihm) for MOX.

B (next)

lesser abundance of these elements, compared with that of U and Pu, the latter being termed major actinides.

Activation processes affecting nuclei of nonradioactive elements mainly involve structural materials, i.e. the materials of the tubes, grids, plates and end-fittings that ensure the mechanical strength of nuclear fuel. These materials lead, in particular, to formation of carbon 14 (¹⁴C), with a halflife of 5,730 years, in amounts that are however very low, much less than one gram per tonne of initial uranium (g/tiU) in usual conditions.

It is the products yielded by fission of the initial uranium 235, but equally of the Pu generated (isotopes 239 and 241), known as fission products (FPs), that are the essential source of the radioactivity of spent fuel, shortly after discharge. Over 300 radionuclides - two thirds of which however will be dissipated through radioactive decay in a few years, after irradiation - have been identified. These radionuclides are distributed over some 40 elements in the periodic table, from germanium (32Ge) to dysprosium (66Dy), with a presence of tritium from fission, i.e. from the fission into three fragments (ternary fission) of ²³⁵U. They are thus characterized by great diversity: diverse radioactive properties, involving as they do some highly radioactive nuclides having very



After discharge, spent fuel is stored in cooling pools, to allow its radioactivity to come down significantly. Shown here is a storage pool at Areva's spent fuel reprocessing plant at La Hague.

family	UOX 33 GWd/tiU (E ²³⁵ U: 3.5%)	UOX 45 GWd/tiU (E ²³⁵ U: 3.7%)	UOX 60 GWd/tiU (E ²³⁵ U: 4.5%)	MOX 45 GWd/tihm (Ei Pu: 8.65%)
	quantity (kg/tiU)	quantity (kg/tiU)	quantity (kg/tiU)	quantity (kg/tihm)
rare gases (Kr, Xe)	5.6	7.7	10.3	7
alkali metals (Cs, Rb)	3	4	5.2	4.5
alkaline-earth metals (Sr, Ba)	2.4	3.3	4.5	2.6
Y and lanthanides	10.2	13.8	18.3	12.4
zirconium	3.6	4.8	6.3	3.3
chalcogens (Se, Te)	0.5	0.7	1	0.8
molybdenum	3.3	4.5	6	4.1
halogens (I, Br)	0.2	0.3	0.4	0.4
technetium	0.8	1.1	1.4	1.1
Ru, Rh, Pd	3.9	5.7	7.7	8.3
miscellaneous: Ag, Cd, Sn, Sb	0.1	0.2	0.3	0.6

Table 2.

Breakdown by chemical family of fission products in spent UOX and MOX fuel, after 3 years' cooling, for a variety of enrichment and burnup rates.

short lifespans, and conversely others having radioactive half-lives counted in millions of years; and diverse chemical properties, as is apparent from the analvsis, for the "reference" fuels used in PWRs in the French fleet, of the breakdown of FPs generated, by families in the periodic table (see Table 2). These FPs, along with the actinides generated, are, for the most part, present in the form of oxides included in the initial uranium oxide, which remains by far the majority constituent. Among some notable exceptions may be noted iodine (I), present in the form of cesium iodide, rare gases, such as krypton (Kr) and xenon (Xe), or certain noble metals, including ruthenium (Ru), rhodium (Rh), and palladium (Pd), which may form metallic inclusions within the oxide matrix

Pu is **recycled** nowadays in the form of MOX fuel, used in part of the fleet (some 20 reactors currently). Residual U may in turn be re-**enriched** (and recycled as a substitute for mined uranium). Recycling intensity depends on market prices for natural uranium, the recent upturn in which should result in raising the current recycling rate (about one third being recycled at present).

Such U and Pu recycling is the foundation for the **reprocessing** strategy currently implemented in France, for the major part of spent fuel (some two thirds currently). For the 500 kg or so of U initially contained in every fuel element, and after partitioning of 475 kg of residual U and about 5 kg Pu, this "ultimate" waste amounts to less than 20 kg of FPs, and less than 500 grams MAs. This waste management pathway (otherwise know as the closed cycle), consisting as it does in reprocessing spent fuel now, to partition recoverable materials and ultimate waste, differs from strategies whereby spent fuel is conserved as-is, whether this be due to a wait-and-see policy (pending a decision on a long-term management mode), or to a so-called **open cycle** policy, whereby spent fuel is considered to be waste, and designated for conditioning into containers, and disposal as-is.

In the nuclear power cycle, as it is implemented in France, waste is subdivided into two categories, according to its origin. Waste directly obtained from spent fuel is further subdivided into minor actinides and fission products, on the one hand, and structural waste, comprising hulls (segments of the cladding tubes that had held the fuel for PWRs) and end-caps (fittings forming the end-pieces of the fuel assemblies for these same PWRs), on the other hand. The process used for spent fuel reprocessing, to extract U and Pu, also generates technological waste (operational waste, such as spare parts, protection gloves...) and liquid effluents.

C What stands between waste and the environment?

aw, solid or liquid radioactive waste **N**undergoes, after characterization (determination of its chemical and radiological makeup, and of its physical-chemical properties), conditioning, a term covering all the operations consisting in bringing this waste (or spent fuel assemblies) to a form suitable for its transport, storage, and disposal (see Box D). The aim is to put radioactive waste into a solid, physically and chemically stable form, and ensure effective, lasting confinement of the radionuclides it contains. For that purpose, two complementary operations are carried out. As a rule, waste is immobilized by a material whether by encapsulation or homogeneous incorporation (liquid or powdered waste, sludges), or encasing (solid waste) - within a matrix, the nature of, and performance specification for which depend on waste type (cement for sludges, evaporation concentrates and incineration ashes; bitumen for encapsulation of sludges or evaporation concentrates from liquid effluent treatment; or a vitreous matrix, intimately binding the nuclides to the glass network, for fission product or minor actinide solutions). This matrix contributes to the confinement function. The waste thus conditioned is placed in an impervious contai-



Cross-section of an experimental storage borehole for a spent fuel container (the lower part of the assembly may be seen, top right), in the Galatée gallery of CECER (Centre d'expertise sur le conditionnement et l'entreposage des matières radioactives: Radioactive Materials Conditioning and Storage Expertise Center), at CEA's Marcoule Center, showing the nested canisters.

ner (cylindrical or rectangular), consisting in one or more canisters. The whole – container and content – is termed a package. Equally, waste may be compacted and mechanically immobilized within a canister, the whole forming a package.

When in the state they come in as supplied by industrial production, they are known as **primary packages**, the primary container being the cement or metal container into which the conditioned waste is ultimately placed, to allow handling. The container may act as initial confinement barrier, allotment of functions between matrix and container being determined according to the nature of the waste involved. Thus, the whole obtained by the grouping together, within one container, of a number of primary

c (next)

ILW-LL packages may ensure confinement of the radioactivity of this type of waste. If a **long-term storage** stage is found to be necessary, beyond the stage of industrial storage on the premises of the producers, primary waste packages must be amenable to retrieval, as and when required: durable primary containers must then be available, in such conditions, for all types of waste.

In such a case, for spent fuel assemblies which might at some time be earmarked for such long-term storage, or even for disposal, it is not feasible to demonstrate, on a timescale of centuries, the integrity of the cladding holding the fuel, forming the initial confinement barrier during the in-reactor use stage. Securing these assemblies in individual, impervious cartridges is thus being considered, this stainless-steel cartridge being compatible with the various possible future management stages: treatment, return to storage, or disposal. Placing these cartridges inside impervious containers ensures a second confinement barrier, as is the case for highlevel waste packages.

In storage or disposal conditions, the waste packages will be subjected to a variety of aggressive agents, both internal and external. First, radionuclide radioactive decay persists inside the package (self-irradiation process). Emission of radiation is concomitant with heat generation. For example, in confinement glasses holding high-activity (high-level) waste, the main sources of irradiation originate in the alpha decay processes from minor actinides, beta decay from fission products, and gamma transitions. Alpha decay, characterized by production of a recoil nucleus, and emission of a particle, which, at the end of its path, yields a helium atom, causes the major part of atom displacements. In particular, recoil nuclei, shedding considerable energy as they do over a short distance. result in atom displacement cascades, thus breaking large numbers of chemical bonds. This is thus the main cause of potential long-term damage. In such conditions, matrices must exhibit thermal stability, and irradiation-damage resistance.

Stored waste packages will also be subjected to the effects of water (leaching). Container canisters may exhibit a degree of resistance to corrosion processes (the overpacks contemplated for glasses may thus delay by some 4,000 years the arrival of water), and the confinement matrices must be proven to exhibit high chemical stability. Between the containers and the ultimate barrier provided, in a radioactive waste deep disposal facility, by the geological environment itself, there may further be interposed, apart, possibly, from an overpack, other barriers, so-called **engineered barriers**, for backfill and sealing purposes. While these would be pointless as backfill in clay formations, they would have the capability, in other environments (granite), of further retarding any flow of radionuclides to the **geosphere**, notwithstanding degradation of the previously mentioned barriers.



Technological demonstrators of ILW-LL packages for bituminized sludges.

From storage to disposal

The object of nuclear waste storage and disposal is to ensure the longterm confinement of radioactivity, in other words to contain radionuclides within a definite space, segre-

gated from humankind and the environment, as long as required, so that the possible return to the **biosphere** of minute amounts of radionuclides can have no unacceptable health or environmental impact.

According to the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management, signed on 5 September 1997, "storage" means "the holding of spent fuel or of radioactive waste in a facility that provides for its containment, with the intention of retrieval." This is thus, by definition, an interim stage, amounting to a delaying, or wait-and-see solution, even though this may be for a very long time (from a few decades to several hundred years), whereas disposal may be final.

Used from the outset of the nuclear power age, industrial storage keeps spent fuel awaiting reprocessing, and conditioned high-level waste (HLW), or long-lived intermediate-level waste (ILW-LL) in conditions of safety, pending a long-term management mode for such waste. Retrieval of stored packages is anticipated, after a period of limited duration (i.e. after a matter of



CEA design study for a common container for the long-term storage and disposal of long-lived, intermediate-level waste.

years, or tens of years).

Long-term storage (LTS) may be contemplated, in particular, in the event of the deferred deployment of a disposal facility, or of reactors to carry out



ANDRA design for the disposal of standard vitrified waste packages in horizontal galleries, showing in particular the packages' various canisters, and some characteristics linked to potential reversibility of the disposal facility.

recycling-transmutation, or simply to turn to advantage the natural decay of radioactivity (and hence the falling off of heat release from high-level waste), before putting the waste into geologi-

> cal disposal. By "long term" is meant a timespan of up to 300 years. Long-term storage may take place in a surface or subsurface facility. In the former case, the site may be protected, for instance, by a reinforcedconcrete structure. In the latter case, it will be located at a depth of some tens of meters, and protected by a natural environment (for instance, if buried in a hillside) and its host rock.

Whichever management strategy is chosen, it will be imperative to protect the biosphere from the residual ultimate waste. The nature of the radioelements the latter contains means a solution is required that has the ability to ensure their confinement over several tens of thousand years, in the case of long-lived waste, or even longer. On such timescales, social stability is a major uncertainty that has to be

taken on board. Which is why disposal in deep geological strata (typically, 500 m down) is seen as a reference solution, insofar as it inherently makes for deployment of a more passive technical solution, with the ability to stand, with no increased risk, an absence of surveillance, thus mitigating a possible loss of memory on the part of society. The geological environment of such a disposal facility thus forms a further, essential barrier, which does not exist in the storage case.

A disposal facility may be designed to be **reversible** over a given period. The concept of reversibility means the design must guarantee the ability, for a variety of reasons, to access the packages, or even to take them out of the facility, over a certain timespan, or to opt for the final closure of the disposal facility. Such reversibility may be envisaged as a succession of stages, each affording a decreasing "level of reversibility." To simplify, each stage consists in carrying out one further technical operation bringing the facility closer to final closure, making retrieval more difficult than at the previous stage, according to wellspecified criteria.

What is transmutation?

Transmutation is the transformation of one nucleus into another, through a reaction induced by particles with which it is bombarded. As applied to the treatment of nuclear waste, this consists in using that type of reaction to transform long-lived radioactive isotopes into isotopes having a markedly shorter life, or even into stable isotopes, in order to reduce the long-term radiotoxic inventory. In theory, the projectiles used may be photons, protons, or neutrons.

In the first case, the aim is to obtain, by bremsstrahlung,⁽¹⁾ through bombardment of a target by a beam of electrons, provided by an accelerator, photons able to bring about reactions of the (γ, xn) type. Under the effects of the incoming gamma radiation, x neutrons are expelled from the nucleus. When applied to substances that are too rich in neutrons, and hence unstable, such as certain fission products (strontium 90, cesium 137...), such reactions yield, as a rule, stable substances. However, owing to the very low efficiency achieved, and the very high electron current intensity required, this path is not deemed to be viable.

In the second case, the proton-nucleus interaction induces a complex reaction, known as **spallation**, resulting in fragmentation of the nucleus, and the release

of a number of particles, including highenergy neutrons. Transmutation by way of *direct* interaction between protons is uneconomic, since this would involve, in order to overcome the Coulomb barrier,^[2] very-high-energy protons (1-2 GeV). requiring a generating energy greater than had been obtained from the process that resulted in producing the waste. On the other hand, indirect transmutation, using very-high-energy neutrons (of which around 30 may be yielded, depending on target nature and incoming proton energy), makes it possible to achieve very significantly improved performance. This is the path forming the basis for the design of so-called hybrid reactors, coupling a subcritical core and a high-intensity proton accelerator (see Box F, What is an ADS?

The third particle that may be used is thus the neutron. Owing to its lack of electric charge, this is by far the particle best suited to meet the desired criteria. It is "naturally" available in large quantities inside nuclear reactors, where it is used to trigger fission reactions, thus yielding energy, while constantly inducing, concurrently, transmutations, most of them unsought. The best recycling path for waste would thus be to reinject it in the very installation, more or less, that had produced it...

(1) From the German for "braking radiation." High-energy photon radiation, yielded by accelerated (or decelerated) particles (electrons) following a circular path, at the same time emitting braking photons tangentially, those with the highest energies being emitted preferentially along the electron beam axis.

(2) A force of repulsion, which resists the drawing together of same-sign electric charges.

When a neutron collides with a nucleus, it may bounce off the nucleus, or penetrate it. In the latter case, the nucleus, by absorbing the neutron, gains excess energy, which it then releases in various ways:

• by expelling particles (a neutron, e.g.), while possibly releasing radiation;

• by solely emitting radiation; this is known as a *capture reaction*, since the neutron remains captive inside the nucleus;

• by breaking up into two nuclei, of more or less equal size, while releasing concurrently two or three neutrons; this is known as a *fission reaction*, in which considerable amounts of energy are released.

Transmutation of a radionuclide may be achieved either through neutron capture or by fission. Minor actinides, as elements having large nuclei (heavy nuclei), may undergo both fission and capture reactions. By fission, they transform into radionuclides that, in a majority of cases, are short-lived, or even into stable nuclei. The nuclei yielded by fission (known as fission products), being smaller, are only the seat of capture reactions, undergoing, on average, 4 radioactive decays, with a half-life not longer than a few years, as a rule, before they reach a stable form. Through capture, the same heavy nuclei transform into other radionuclides, often long-lived, which transform in turn through natural decay, but equally through capture and fission.

🗉 (next)

The probability, for a neutron, of causing a capture or a fission reaction is evaluated on the basis, respectively, of its capture **cross-section** and fission cross-section. Such cross-sections depend on the nature of the nucleus (they vary considerably from one nucleus to the next, and, even more markedly, from one isotope to the next for the same nucleus) and neutron energy.

For a neutron having an energy lower than 1 eV (in the range of slow, or thermal, neutrons), the capture cross-section prevails; capture is about 100 times more probable than fission. This remains the case for energies in the 1 eV-1 MeV range (i.e., that of epithermal neutrons, where captures or fissions occur at definite energy levels). Beyond 1 MeV (fast neutron range), fissions become more probable than captures.

Two reactor pathways may be considered, according to the neutron energy range for which the majority of fission reactions occur: thermal-neutron reactors, and fast-neutron reactors. The ther-



Figure.

Simplified representation of the evolution chain of americium 241 in a thermal-neutron reactor [shown in blue: radionuclides disappearing through fission]. Through capture, ²⁴¹Am transforms into ^{242m}Am, this disappearing predominantly through fission, and into ²⁴²Am, which mainly decays (with a half-life of 16 hours) through beta decay into ²⁴²Cm. ²⁴²Cm transforms through lapha decay into ²³⁸Pu, and through capture into ²⁴³Cm, which itself disappears predominantly through fission. ²³⁸Pu transforms through capture into ²⁴³Pu, which disappears predominantly through fission.

mal neutron pathway is the technology used by France for its power generation equipment, with close to 60 pressurizedwater reactors. In a thermal-neutron reactor, neutrons yielded by fission are slowed down (moderated) through collisions against light nuclei, making up materials known as moderators. Due to the moderator (common water, in the case of pressurized-water reactors), neutron velocity falls off, down to a few kilometers per second, a value at which neutrons find themselves in thermal equilibrium with the ambient environment. Since fission cross-sections for 235U and 239Pu, for fission induced by thermal neutrons, are very large, a concentration of a few per cent of these fissile nuclei is sufficient to sustain the cascade of fissions. The flux, in a thermal-neutron reactor, is of the order of 1018 neutrons per square meter, per second.

In a fast-neutron reactor, such as Phénix, neutrons yielded by fission immediately induce, without first being slowed down, further fissions. There is no moderator in this case. Since, for this energy range, cross-sections are small, a fuel rich in fissile radionuclides must be used (up to 20% uranium 235 or plutonium 239), if the neutron multiplication factor is to be equal to 1. The flux in a fast-neutron reactor is ten times larger (of the order of 1019 neutrons per square meter, per second) than for a thermal-neutron reactor.

F What is an ADS?

An ADS (accelerator-driven system) is a hybrid system, comprising a nuclear reactor operating in subcritical mode, i.e. a reactor unable by itself to sustain a fission chain reaction, "driven" by an external source, having the ability to supply it with the required comple-

ment of **neutrons**.^[1]

Inside the core of a nuclear reactor, indeed, it is the fission energy from **heavy nuclei**, such as **uranium** 235 or **plutonium** 239, that is released. Uranium 235 yields, when undergoing fission, on average 2.5 neutrons, which can in turn induce a further fission, if they collide with a uranium 235 nucleus. It may thus

is initiated, a chain reaction may develop, resulting, through a succession of fissions, in a rise in the neutron population. However, of the 2.5 neutrons yielded by the initial fission, some are captured, thus not giving rise to further fissions. The number of fissions generated from one initial fission is characterized by the effective multiplication factor k_{eff}, equal to the ratio of the number of fission neutrons generated, over the number of neutrons disappearing. It is on the value of this coefficient that the evolution of the neutron population depends: if k_{eff} is markedly higher than 1, the population increases rapidly; if it is slightly higher than 1, neutron multiplication sets in, but remains under control: this is the state desired at reactor startup; if k_{eff} is equal to 1, the population remains stable; this is the state

(1) On this topic, see Clefs CEA, No. 37, p. 14

for a reactor in normal operating conditions; and, if k_{eff} is lower than 1, the neutron population dwindles, and becomes extinct, unless – as is the case for a hybrid system – an external source provides a neutron supply.



be seen that, once the initial fission Principle schematic of an ADS.

From the effective multiplication factor, a reactor's **reactivity** is defined by the ratio $(k_{eff} - 1)/k_{eff}$. The condition for stability is then expressed by zero reactivity. To stabilize a neutron population, it is sufficient to act on the proportion of materials exhibiting a large **neutron capture** cross-section (neutron absorber materials) inside the reactor.

In an ADS, the source of extra neutrons is fed with protons, generated with an energy of about 100 keV, then injected into an accelerator (linear accelerator or cyclotron), which brings them to an energy of around 1 GeV, and directs them to a heavy-metal target (lead, lead-bismuth, tungsten or tantalum). When irradiated by the proton beam, this target yields, through spallation reactions, an intense, high-energy (1–20 MeV) neutron flux, one single incoming neutron having the ability to generate up to 30 neutrons. The latter then go on to interact with the fuel of the subcritical neutron **multiplier medium**, yielding further neutrons (fission neutrons) (see Figure).

Most hybrid system projects use as a core (of annular configuration, as a rule) fast-

neutron environments, since these make it possible to achieve neutron balances most favorable to transmutation, an operation that allows waste to be "burned," but which may equally be used to yield further fissile nuclei. Such a system may also be used for energy generation, even though part of this energy must be set aside to power the proton accelerator, a part that is all the higher, the more

subcritical the system is. Such a system is safe in principle from most reactivity accidents, its multiplication factor being lower than 1, contrary to that of a reactor operated in critical mode: the chain reaction would come to a halt, if it was not sustained by this supply of external neutrons.

A major component in a hybrid reactor, the window, positioned at the end of the beam line, isolates the accelerator from the target, and makes it possible to keep the accelerator in a vacuum. Traversed as it is by the proton beam, it is a sensitive part of the system: its lifespan depends on thermal and mechanical stresses, and corrosion. Projects are mooted, however, of windowless ADSs. In the latter case, it is the confinement constraints, and those of radioactive spallation product extraction, that must be taken on board.

The industrial context

The characteristics of the major part of the radioactive waste generated in France are determined by those of the French nuclear power generation fleet, and of the spent fuel reprocessing plants, built in compliance with the principle of reprocessing such fuel, to partition such materials as remain recoverable for energy purposes (uranium and plutonium), and waste (fission products and minor actinides), not amenable to recycling in the current state of the art.

58 enriched-uranium pressurized-water reactors (PWRs) have been put on stream by French national utility EDF, from 1977 (Fessenheim) to 1999 (Civaux), forming a second generation of reactors, following the first generation, which mainly comprised 8 UNGG (natural uranium, graphite, gas) reactors, now all closed down, and, in the case of the older reactors, in the course of decommissioning. Some 20 of these PWRs carry out the industrial recycling of plutonium, included in MOX fuel, supplied since 1995 by the Melox plant, at Marcoule (Gard *département*, Southern France).

EDF is contemplating the gradual replacement of the current PWRs by third-generation reactors, belonging to the selfsame pressurized-water reactor pathway, of the EPR (European Pressurized-Water Reactor) type, designed by Areva NP (formerly Framatome–ANP), a division of the Areva Group. The very first EPR is being built in Finland, the first to be built in France being sited at Flamanville (Manche *département*, Western France).

The major part of spent fuel from the French fleet currently undergoes reprocessing at the UP2-800^[1] plant, which has been operated at La Hague (Manche *département*), since 1994, by Areva NC (formerly Cogema,) another member of the Areva Group (the UP3 plant, put on stream in 1990–92, for its part, carries out reprocessing of fuel from other countries). The waste vitrification workshops at these plants, the outcome of development work initiated at Marcoule, give their name (R7T7) to the "nuclear" glass used for the confinement of long-lived, high-level waste.

A fourth generation of reactors could emerge from 2040 (along with new reprocessing plants), a prototype being built by 2020. These could be **fast-neutron** reactors (i.e. fast reactors [**FRs**]), either sodium-cooled (SFRs) or gas-cooled (**GFRs**). Following the closing down of the Superphénix reactor, in 1998, only one FR is operated in France, the Phénix reactor, due to be closed down in 2009.

(1) A reengineering of the UP2-400 plant, which, after the UP1 plant, at Marcoule, had been intended to reprocess spent fuel from the UNGG pathway.