

An ongoing process of advances

Research work, seeking the best solutions for the long-term management of long-lived, high-level radioactive waste is part and parcel of a process of staged advances, based, from the outset of the use of nuclear power, in France, on two principles: optimum use of energy-rich materials, and the safe conditioning of ultimate waste.

Production of nuclear-generated electric power, in France, is supported by a mature, high-performance industrial resource, constructed, and constantly improved, from 1975 on, comprising reactors and cycle plants (see Box 1), industrial materials and waste **storage facilities**, and **surface disposal facilities**. This production goes hand in hand, through a policy fostering ongoing advances, with optimization of the

management of the **radioactive waste** it generates (see Box A, *What is radioactive waste?* p. 16), consisting in partitioning and recycling materials recoverable for energy purposes, while reducing, **conditioning**, and consigning to disposal ultimate waste (see Chapter I, *The groundwork of current solutions*, p. 13).

Short-lived, low-level waste (about 15,000 m³/year) is managed by **ANDRA**, and kept in surface disposal at the Aube Center (and previously at the Manche Center, which has ceased taking in **packages** since 1994). Nowadays, after completing spent **fuel reprocessing** operations at La Hague (Manche *département*, Western France), all waste is conditioned. Over 95% of total **radioactivity** is concentrated in **long-lived, high-level** waste, which is incorporated into a glass **matrix** (see Box B, *Waste from the nuclear power cycle*, p. 20). Such waste is stored in purpose-built facilities.

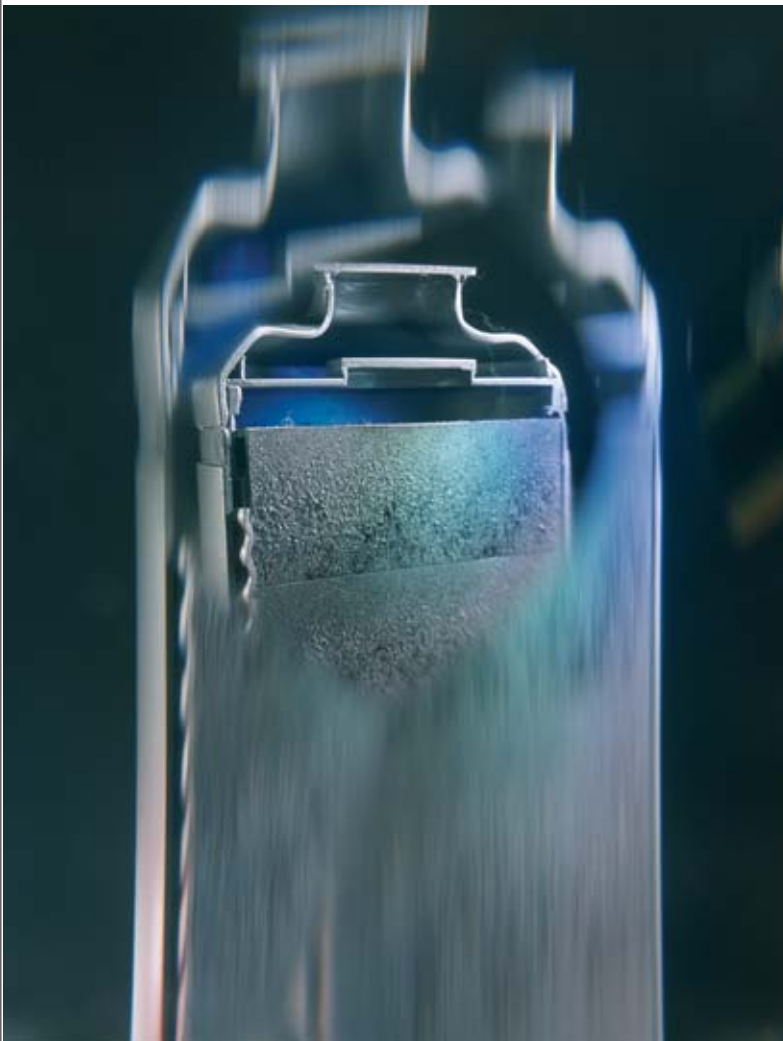
Reprocessing operations, carried out on an industrial basis in France, make it possible, on the one hand, to achieve a reduction in waste **radiotoxic inventory**, compared to the no-reprocessing option (**open cycle**), by extracting, and recycling, **plutonium**, an energy-rich material, and, on the other, by conditioning long-lived waste in safe, durable fashion, in a very small volume.

What has been the outcome of the research effort since 1991?

Under the aegis of the Act of 30 December 1991 (see Box 2), the French government charged CEA with conducting investigations in accordance with direction 1 (partitioning and **transmutation**; see Box E, *What is transmutation?* p. 90), and direction 3 (conditioning; see Box C, *What stands between waste and the environment?* p. 28; and storage; see Box D, *From storage to disposal*, p. 50), while entrusting ANDRA with direction 2 (**geological disposal**). These investigations have been carried out in highly sustained fashion from 1992, drawing on major collaborations in France, as on the European and world scenes (see Box 3). Reports setting out the results achieved by this research effort were presented (on 30 June 2005, for the CEA part) to the Minister for Industry, and the Minister for Research (see Chapter II, *New designs: the package at the core of investigations*, p. 79). These reports were subsequently updated, at the end of 2005.

Direction 1: partitioning and transmutation

Investigations on partitioning and transmutation seek to achieve separation of the more radiotoxic long-lived elements present in waste (mainly **minor actinides**, which currently are **vitrified**, along with **fission products**), going on to transmute them, by recycling them in nuclear reactors, to transform them into non-radioactive elements, or elements having a shorter half-life (see Chapter III, *Advances for tomorrow and after*, p. 79).



Artistic composite view of sectioned standard compacted waste packages, as produced on an industrial basis by Areva NC (Cogema).

D. Sarrault/CEA



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.

Enhanced separation

A whole new chemistry of separation had to be developed (related, nonetheless, to aqueous-solution processes such as the **Purex** process), with the design, synthesis, and testing, on representative radioactive solutions, of some one hundred new, highly selective molecules. Investigations ultimately made it possible to select the three molecules used in the process chosen for enhanced separation. Its feasibility was demonstrated in the laboratory, in CEA's **Atalante** facility, at Marcoule, at first on actual solutions yielded by dissolution of a few hundred grams of spent fuel, with a highly satisfactory performance: the recovery of at least 99% of minor actinides. This is a major outcome for this research effort.

That feasibility was confirmed by the successful setting up of continuous processes, using a solution containing some 15 kg of spent fuel. For neptunium, the trial, carried out in April 2005, corroborated the findings obtained previously. For the minor actinides americium and curium, the confirmation came in November and December of the same year.

Transmutation

Once plutonium and **uranium** are recycled, partitioning of minor actinides, and their subsequent transmutation, would allow a further reduction in ultimate waste radiotoxic inventory (dividing, by a factor ranging from 10 to 100, according to the nuclei involved, the radiotoxicity at 500 years for new vitrified waste, compared to that of current waste). This new stage, as regards advances relating to ultimate waste, may not be achieved by means of the **thermal-neutron** reactors in the current fleet, but requires deployment of a novel, **fast-neutron** nuclear system.

Indeed, the best conditions for transmutation are obtained with a fast neutron spectrum. In 1998, after the decision to close down the Superphénix reactor was taken, the experimental demonstration program relating to transmutation, which had been set in train in



J.M. Taillet/Areva/CEA

that reactor, was redeployed. A substantial redesign and rejigging effort, for the scheduled irradiations, was successfully completed in 2002, to enable the introduction of irradiation experiments into the Phénix reactor. Post-irradiation inspections of compounds for transmutation were initiated with two new experiments, MATINA 1A (investigation of matrices) and METAPHIX (transmutation of minor actinides into metal fuel), which were taken out of this reactor in 2004, with the other irradiations following, up to 2009. On the basis of the numerous findings from irradiations already obtained, the technical feasibility of transmutation of americium and neptunium has been established, for oxide fuels in sodium-cooled fast reactors. The next stage for advances concerning transmutation of minor actinides is associated to a new generation of industrial instruments, emerging around 2040, fourth-generation fast reactors, along with new cycle plants, allowing the recycling and transmutation of all **heavy nuclei**, or dedicated burner reactors, whether critical or **subcritical**, along with their cycle (see Box F, **What is an ADS?** p. 103).

These new instruments will have to meet criteria relating to resource savings, waste minimization, control

The Atalante facility, at CEA's Marcoule Center (Gard *département*, Southern France), where the demonstration of feasibility was carried out, using actual solutions yielded by dissolution of spent fuel, for enhanced separation of actinides.



The Act of 1991

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In 1990, controversies on the issue of **radioactive waste** led the French government to decide a moratorium on the quest for a **geological disposal** site, and to draw up a Bill to set the framework for research work on the long-term management of **long-lived, high-level** radioactive waste. Passed on 30 December 1991, the Act, No. 91-1381, commonly known as the "Bataille Act," from the name of the author of the accompanying parliamentary report, was incorporated in 2000 into the French Environmental Code (article L. 542). Accompanied as it was by a 15-year moratorium on any decision as to the long-term future of such waste, the Act outlined a research program, to be carried out over that period, specifying that Parliament would be presented, in 2006, with a general evaluation report on the investigations thus provided for, to be conducted along three directions:

- to seek solutions enabling the partitioning and **transmutation** of long-lived radioactive elements present in this waste;
- to investigate the possibilities of **reversible**, or irreversible, disposal in deep geological formations, in particular through construction of underground laboratories;
- to investigate processes for the **conditioning**, and long-term **storage**, in **surface** or **subsurface** locations, of long-lived, high-level radioactive waste (an extension to cover the subsurface case was called for by the government, in 1998).

Steering investigations in accordance with directions 1 and 3 was entrusted to CEA by the government, and for direction 2 to **ANDRA**.

These investigations, accounting for budget appropriations of some 2.6 billion euros, from 1992 to 2005 (including some €1.6 billion for the directions steered by CEA), were carried out in highly sustained fashion from 1992, drawing on major collaborations in France, as on the European and world scenes (see Box 3).

They have been monitored, on a regular basis, by the Parliamentary Office for the Assessment of Scientific and Technological Options (**Office parlementaire d'évaluation des choix scientifiques et technologiques**) and the government, coordination being carried out through the Committee for the Monitoring of Research on the Cycle Back-End (**Comité de suivi des recherches sur l'aval du cycle**), set up by the French Ministry for Research. Research work has been constantly audited by the National Review Board (**Commission nationale d'évaluation**), provided for in the Act.

of proliferation risks, and cycle economic balance. The choice of a transmutation system, leading to the construction of a demonstrator and setting up of the associated recycling process, could occur by 2015.

The outcomes achieved in 2006 show that waste (vitrified waste, or long-lived, **intermediate-level** waste [**ILW-LL**]) already produced, or currently being produced, is to stand as ultimate waste, the prospects opened up by separation–transmutation concerning future waste, from 2040 on.

A variety of solutions for the long-term management of radioactive waste were thought up in the past, and subsequently abandoned. Left standing, nowadays, are the solution of well-controlled disposal, in **deep**, stable **geological formations**, and that of **long-term storage** of waste in surface or **subsurface** facilities, extrapolated from current industrial storage practice (see Chapter II).

Direction 2: geological disposal

Disposal in deep geological strata presents a number of advantages, namely: the existence of a geological **barrier**, isolating packages from the **biosphere**; high chemical stability of the water present, making for good control of waste package corrosion; and a lesser risk of human intrusion, lightening the duty of surveillance. Research carried out by ANDRA warrants the conclu-

sion that such a solution is feasible, and could be deployed on an industrial basis by 2025.

Direction 3: conditioning and storage

The R&D effort covers two aspects: conditioning, this being concerned with the fabrication, and understanding, of packages (development of radioactive materials conditioning processes, of **containers**, package characterization, and investigation of package long-term behavior); and storage, this being concerned with the specification, and qualification, of designs for long-term storage facilities, whether surface or subsurface.

Conditioning

Development work carried out in the area of waste treatment and conditioning sought, on the one hand, to achieve improvements (volume reduction...) in some of the current processes, taking on board, in particular, the evolution in waste composition, and, on the other hand, to ensure availability of qualified processes, that might be used for older, legacy waste.

Concurrently, in order to explore all potential future avenues, specific conditioning processes for elements amenable to partitioning, in accordance with direction 1, are being developed at the laboratory level, in the event of such elements proving not to be transmutable. All the work on development, and evaluation, of these processes, as regards technological, and subsequently industrial, feasibility, still remains to be done.

In this area, the outcomes achieved, and their industrial deployment, have resulted, overall, in dividing, by a factor of at least 6, the volume of solid waste, and by a factor 10 the activity of liquid effluents, since the La Hague reprocessing plant came on stream. Such remarkable, ongoing advances on the part of research organizations, and manufacturers, sustained over the 15 years covered by the Act of 30 December 1991 (see Figure, p. 7), now makes it possible to have safe, high-performance conditionings accommodated in industrial storage facilities, this being a medium-term solution, though not a long-term management solution.

Package characterization and control

Investigations carried out at CEA, in collaboration, in particular, with **Areva/Cogema** and ANDRA, have allowed development of highly comprehensive methods, and systems, for the characterization and control of packages. These methods have been qualified with actual packages, in particular in the CHICADE facility, at Cadarache.

Investigation of package long-term behavior

The purpose of conditioning being to ensure durable confinement, through all stages in package management, there is the requisite to ascertain the scientific and technical elements enabling prediction of long-term package behavior, and confirming that the associated functions will be ensured, particularly handling, retrieval, and confinement. Investigations carried out over the past few years have made it possible to set out the foundations of a veritable science of long-term behavior. The high durability of glasses (their dissolution will extend over several hundred thousand years,

in disposal conditions!), in particular, has been established.

For all package types, the ensemble of processes governing long-term behavior has been modeled, the models being compared with experimental findings, and qualified. They are used, in particular, by ANDRA, for overall investigations on operation and behavior over extended timespans of disposal facilities.

Containers

A major effort was concerned with the development and qualification of containers for disposal, or long-term storage. Thus, common containers for the purposes of long-term storage and disposal of long-lived, intermediate-level waste (bituminized waste, and compacted **hull** and **end-cap** waste) were developed, in collaboration with ANDRA.

Although the strategy adopted in France does not consider spent fuel to be waste, a “storage-compatible” disposal container for spent fuel has been the subject of studies, jointly carried out by CEA, **EDF**, and ANDRA.

Finally, containers for the long-term dry storage of spent fuel, guaranteeing retrieval at a later date, have been specified.

All the demonstrators for such containers have been fabricated, and may be viewed at the Radioactive Materials Conditioning and Storage Expertise Center (**CECER**: *Centre d'expertise sur le conditionnement et l'entreposage des matières radioactives*), set up at Marcoule.

Long-term storage

Lifetime extension, and possible renovation, of industrial storage facilities stand as one initial response to the requirement for long-term storage. Durability assessments have been carried out, for the more recent

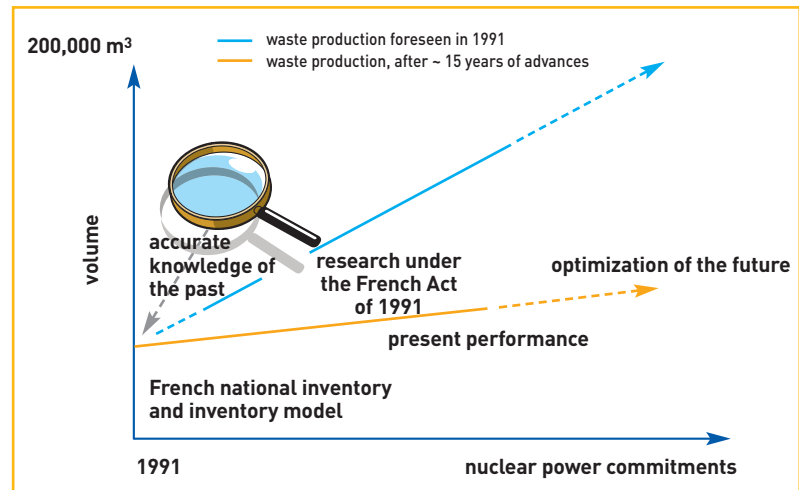


Figure. Evolution of the volume of radioactive waste production in France, as foreseen in the past, and predicted today.

storage facilities. It has been shown these could last at least one hundred years, provided the required authorizations are forthcoming.

Investigations have also been concerned with storage facilities designed from the outset for the long term, whether in surface or subsurface locations, to accommodate spent fuel and long-lived, high- or intermediate-level waste packages. Investigations were carried out on the basis of considering generic sites (i.e., not related to any specific geographic location). Indeed, for storage, as opposed to disposal, practically no radioelement confinement function is required from the geological environment.

Studies for two designs for long-term storage facilities, one surface and one subsurface, have been carried out, to accommodate spent fuel, vitrified waste, and ILW-LL waste (preliminary studies, safety assessments, and design definition studies by the end of 2005). A demon-

Partnerships in France and international collaborations

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In compliance with direction 1, as provided for by the Act of 1991, CEA has developed a major partnership with the French **CNRS**, taking the form of the setting up of coordinated research groups (GDRs: *groupements de recherche*), under the aegis of CNRS's Program on the Nuclear Power Cycle Back-End (**PACE**: *Programme sur l'aval du cycle électronucléaire*). These groups, bringing together many partners, including manufacturers from the nuclear power sector, include **PRACTIS**, subsequently **PARIS**, working on separation (with **EDF** and **ANDRA**); **GEDEON**, subsequently **GEDEPEON**, on **transmutation** and associated scenarios (also with EDF, **Framatome-ANP** [now Areva NP] and ANDRA); and **NOMADE**, on specific **conditionings** for partitioned elements, and transmutation targets (with Areva NC [Cogema] and EDF). In the context of investigations in accordance with directions 2 and 3, ANDRA and CEA have been working in close collaboration, to investigate **package** behavior in disposal conditions, and to guarantee the most effective compatibility between the strategies being investigated, of **long-term-enabled storage**, and disposal, for waste packages.

To ensure coordination of the work carried out along the various directions, and enable the required scientific and technical interchange, topical technical committees were set up, under the aegis of the strategic partnership agreement between ANDRA and CEA. For the same reason, industrialists, mainly EDF and Areva (NC and NP divisions), traditional partners for CEA, under the provisions of existing research agreements, also proved to be major players. They made it possible to bring to investigations the bene-

fit of their know-how, particularly with regard to primary waste package management, and operational feedback with respect to existing storage facilities.

On the international scene, collaborations have been set up, on the one hand, with Japan (**JAERI**, **JNC**), which is pursuing closely related goals, and, on the other, with Russian institutes, on partitioning, and transmutation experiments (in the BOR-60 **fast-neutron** reactor, in particular). Investigations on transmutation benefited from experimental collaborations around the HFR reactor, at Petten (Netherlands). The Institute for Transuranium Elements (ITU), Karlsruhe (Germany), contributed to investigations on **actinides**. More recent collaborations with the United States (in particular with research laboratories run by **DOE**) are concerned with partitioning and **fuel** irradiations, and, lately, with cycles, in the context, in particular, of development work for future nuclear systems.

Major collaborations were carried out under the aegis of the successive European Framework Programs for Research and Technological Development (**FPs**). The main topics addressed, in this respect, were aqueous- or dry-path partitioning (**EUROPART** Program, under the 6th Framework Program [FP6], devoted to investigation of minor actinide partitioning, following from the **PARTNEW** Program under FP5, and the **NEWPART** Program under FP4), transmutation (including the **EUROTRANS** Program under FP6, devoted to investigation of **ADSs**), and long-term waste package behavior.



Entrance to the demonstration gallery for a long-term subsurface storage facility for spent fuel and nuclear waste, at the Radioactive Materials Conditioning and Storage Expertise Center (CECER: *Centre d'expertise sur le conditionnement et l'entreposage des matières radioactives*), at Marcoule.



A. Gomin/CEA

tration gallery, representative of subsurface storage conditions, has been constructed at CECER, at Marcoule.

These studies warrant the conclusion that long-term storage is indeed feasible. This management solution, however, does require monitoring, maintenance, and the retrieval of packages, at the end of the storage period, burdens that will have to be shouldered by future generations.

Taking into account extant, recent industrial storage facilities, a requirement for storage facilities designed from the outset for long-term operation, for high-level and long-lived waste, would only emerge if the geological disposal solution were not given the go-ahead. In that case, selection of the storage site, and engineering studies that still require to be carried out, should lead to completion by 2015.

The international context

Storage is the prevailing industrial practice. To date, no country has implemented a long-term management policy for high-level nuclear waste. Sweden, Finland and the United States are going for direct disposal of spent fuel in deep geological formations. The United States, for their part, initiated, in 1999, deep disposal of intermediate-level waste (see *How are other countries managing their nuclear waste?* p. 75).

Rising energy demand, the necessary curbing of greenhouse gas emissions, the growing scarcity of natural oil resources – a form of fossil energy – are factors that could lead to increased reliance on nuclear power, over the course of the 21st century. This trend is already manifest, in particular, in a number of countries such as Finland, China, and, to a certain extent, the United States.

What are the stages to come after 2006?

CEA is taking part, and presenting its results, in the currently ongoing national debate being staged in France on radioactive waste management. The Visiatome visitor center, opened at Marcoule in 2005, contributes to information of the public on radioactive waste and nuclear power.

CEA published, at the end of 2005, the final reports on the research effort it has steered, contributing to providing the technical elements that will enable the decisions to be taken in 2006, regarding management modes for long-lived waste.

The time constants associated to evolution of the existing industrial resource are extremely long (typically,

30 years to design and build a new reactor, 25 years for a separation plant). However, as has been the case since the outset of nuclear power, as in all areas of technology, advances will go on supporting industrial developments, through a succession of stages over time that will enable industrial, and political, choices, for the major milestones to come.

These stages could be based, in research terms, on the following developments:

- with a view to support industrial deployment of facilities, the carrying through of investigations on retrieval and conditioning of older, legacy waste, and waste volume reduction, in particular for waste from decommissioning;
- development of highly comprehensive methods, and systems, for waste package characterization and control, allowing existing inventories to be more precisely detailed;
- the specification of long-term management pathways for all types of waste;
- studies for, and construction of, a geological disposal facility for ultimate waste;
- the specification of ultimate waste packages directed to disposal, with regard to disposal facility specifications, and investigation on long-term behavior of such packages in a disposal environment;
- demonstration of the industrial feasibility (by 2025) of actinide separation, and transmutation in a fast reactor;
- initial deployment of the future industrial system (reactor and cycle plants, by 2040), allowing, in the longer term, an evolution of ultimate waste.

This policy, of staged advances, is part and parcel of a broader drive for the sustainable development of nuclear energy, based on optimum use of energy-rich materials. The reprocessing of spent fuel stands as the cornerstone of this strategy.

France has resolutely advanced along this path, from the outset of the nuclear power age, with the development, and demonstration, of industrial technologies for the back-end of the cycle (reprocessing, recycling, high-performance conditioning of ultimate waste), securing the status of clear world leader in this area. Taking on board the decisions to be taken in 2006, CEA will continue, in its various fields of expertise, to carry out the required research effort, drawing on the support of players on the French scene, and further promoting international collaborations.

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A What is radioactive waste?



According to the **International Atomic 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 long-lived 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 **fusion 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 TWh 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 (m ³)
Vitrified waste packages	CO – C2	Cogema*	HLW	42,470	7,410
Activated metal waste packages	B1	EDF	ILW-LL	2,560	470
Bituminized sludge packages	B2	CEA, Cogema*	ILW-LL	105,010	36,060
Cemented technological waste packages	B3	CEA, Cogema*	ILW-LL	32,940	27,260
Cemented hull and end-cap packages	B4	Cogema*	ILW-LL	1,520	2,730
Compacted structural and technological waste packages	B5	Cogema*	ILW-LL	39,900	7,300
Containerized loose structural and technological waste packages	B6	Cogema*	ILW-LL	10,810	4,580
Total B				192,740	78,400
Total overall				235,210	85,810

* renamed Areva NC in 2006

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

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 high 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.

	Short-lived Half-life < 30 years for the main elements	Long-lived Half-life > 30 years
Very-low-level waste (VLLW)	Morvilliers dedicated disposal facility (open since 2003) Capacity: 650,000 m ³	
Low-level waste (LLW)	Aube Center (open since 1992) Capacity: 1 million m ³	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)		MID volume estimate: 78,000 m ³
High-level waste (HLW)	MID volume estimate: 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)

B Waste from the nuclear power cycle

Most 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

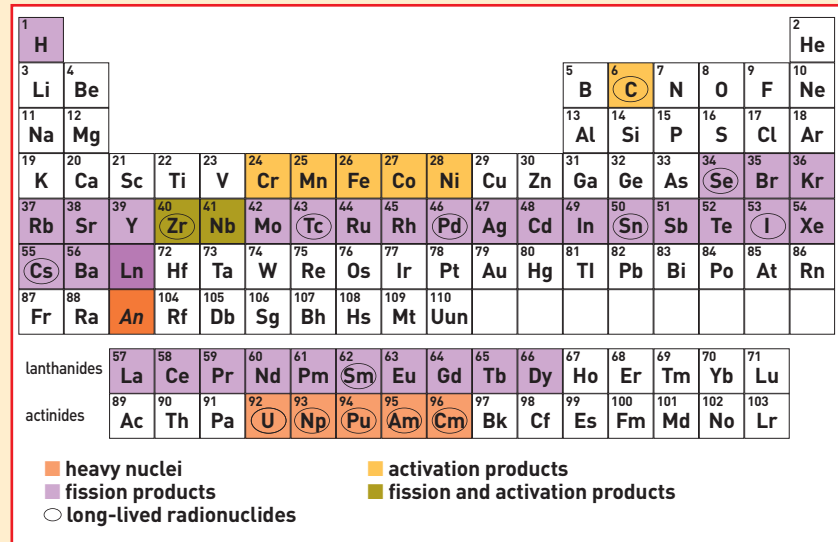


Figure 1. The main elements found in spent nuclear fuel.

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 half-lives**. These are the elements of the **actinide** family, this including, essentially, plutonium (from ²³⁸Pu to ²⁴²Pu, the odd-numbered 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 non-radioactive 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** (^{14}C), with a half-life 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 (^{32}Ge) to dysprosium (^{66}Dy), with a presence of **tritium from fission**, i.e. from the fission into three fragments (ternary fission) of ^{235}U . They are thus characterized by great diversity: diverse radioactive properties, involving as they do some highly radioactive nuclides having very



Magnum/Harry Gruyaert

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 analysis, 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 known 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?

Raw, solid or liquid **radioactive waste** 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 **fusion product** or **minor actinide** solutions). This matrix contributes to the confinement function. The waste thus conditioned is placed in an impervious **contai-**



A. Gomin/CEA

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 pri-

mary 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 **high-level** 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.





D From storage to disposal

The object of nuclear waste **storage** and **disposal** is to ensure the long-term **confinement** of **radioactivity**, in other words to contain **radionuclides** within a definite space, segregated 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).

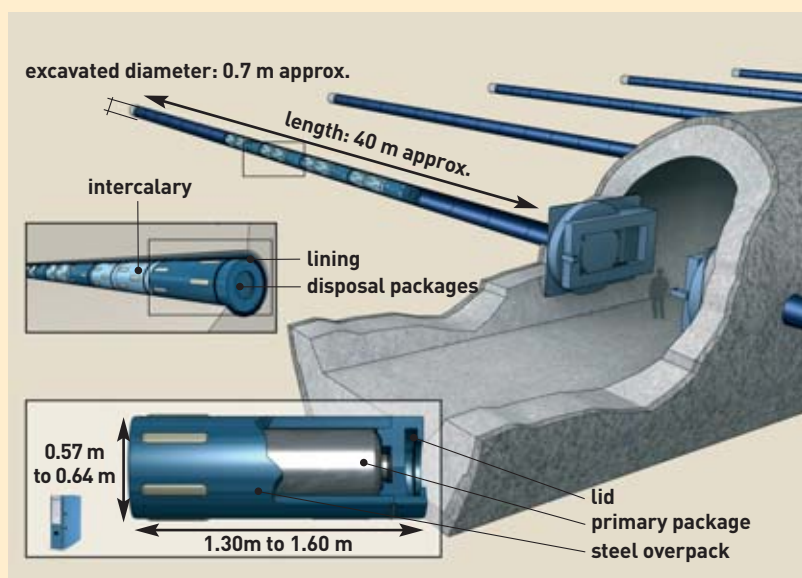
Long-term storage (LTS) may be contemplated, in particular, in the event of the deferred deployment of a disposal facility, or of reactors to carry out

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 **geological disposal**. By "long term" is meant a timespan of up to 300 years. Long-term storage may take place in a **surface** or **sub-surface** facility. In the former case, the site may be protected, for instance, by a reinforced-concrete 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 hill-side) 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 well-specified criteria.



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.

A. Gonty/CEA

ANDRA

E 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 high-energy 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...

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.

(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.

E (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-

mal neutron pathway is the technology used by France for its power generation equipment, with close to 60 pressurized-water 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 ^{235}U and ^{239}Pu , 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.

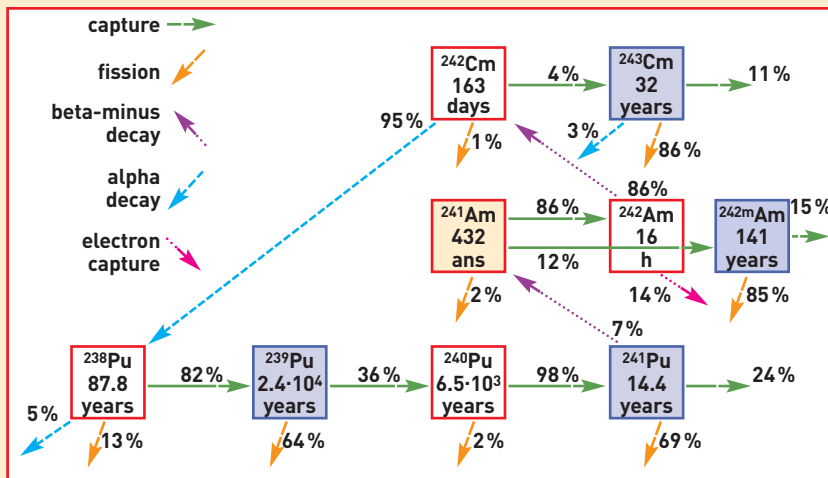


Figure.

Simplified representation of the evolution chain of americium 241 in a thermal-neutron reactor (shown in blue: radionuclides disappearing through fission). Through capture, ^{241}Am transforms into ^{242m}Am , this disappearing predominantly through fission, and into ^{242}Am , which mainly decays (with a half-life of 16 hours) through beta decay into ^{242}Cm . ^{242}Cm transforms through alpha decay into ^{238}Pu , and through capture into ^{243}Cm , which itself disappears predominantly through fission. ^{238}Pu transforms through capture into ^{239}Pu , which disappears predominantly through fission.

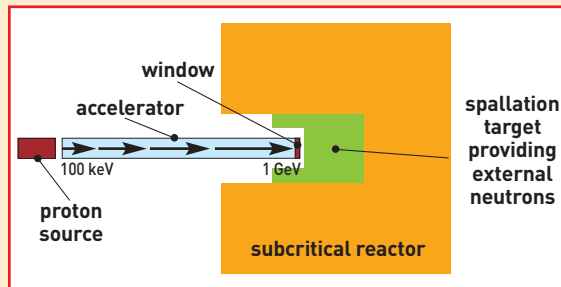
F What is an ADS?

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 complement 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 be seen that, once the initial fission

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

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.



Principle schematic of an ADS.

From the effective multiplication factor, a reactor's reactivity is defined by the ratio $(k_{\text{eff}} - 1)/k_{\text{eff}}$. 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 lat-

ter 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.

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

The industrial context

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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.