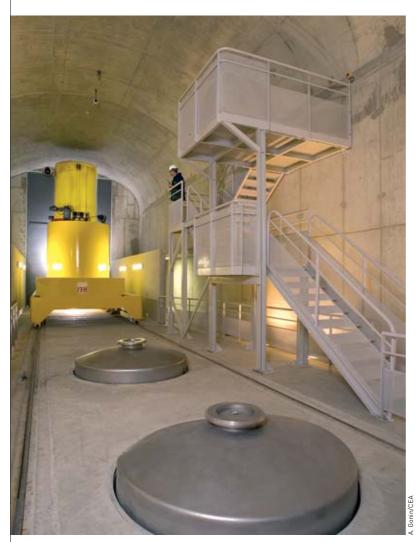


How may storage and the long term be made compatible?



Storage Test Activity Gallery (Galatée: Galerie activités de tests pour l'entreposage), constructed at CEA's Marcoule (Gard département, Southern France) Center, for investigation of the longterm storage of heatgenerating waste. Two well heads and a 170tonne transfer cask, for container handling, may be seen, in the foreground and at the back, respectively.

By definition an interim solution, is industrial storage compatible with the long term? Depending on the standpoint adopted, it may be restricted to a minimum, or, on the contrary, it may stand as an integral part of a solution for the very long term.

Storage, in all industries, makes for responsiveness and flexibility in the management of the products involved. If it be envisaged as a long-term operation, as regards **nuclear waste**, a number of queries arise instantly: What do we mean by "long term"? Why the call for a **long-term storage** (**LTS**) **facility**? What does such a timespan involve, with respect to structure design, and construction? What is to be placed in an LTS facility?

By definition (see Box D, *From storage to disposal*, p. 50), a storage facility is a temporary depository, since the items stored there have to be systematically retrie-

ved at the end of their storage time, which is not intended to stretch into infinity, contrary to what holds for a **geological disposal** facility, or repository. Compared to current facilities, designed for an anticipated lifetime of a few tens of years, the long-term case stands out, owing to the fact it concerns several generations. The lifespan selected for investigations has been set at 1–3 centuries. To meet this challenge, two approaches may be considered: either to extend the lifespan of industrial storage facilities as they currently stand, or build facilities directly designed for the long term.

The first approach was covered by the investigation of current industrial storage facility lifespans. Operational feedback, and the investigations carried out warrant that an extension of this lifespan be contemplated, up to one hundred years or so. Research work carried out by CEA has focused more particularly on the second option, i.e. on a facility designed for operation over 100–300 years.

Why go for LTS?

While LTS is not a final management mode, owing to the obligatory retrieval of **packages**, it may make for flexibility in waste management. Indeed, an LTS facility allows waste to be kept in safe conditions, pending availability of an **outlet**, or to enable optimization of outlet design. For example, if a disposal facility is being planned, waiting for a falloff in the thermal power released by the packages allows ground area to be limited.

How may the long term be taken on board?

300 years, i.e. three centuries: from 1700 to this day, France, for instance, has seen exponential growth, but equally wars, revolutions, economic crises... Taking on board the long term may thus not simply be based on technical performance, in terms of package and storage facility durability, it must also take into consideration societal uncertainties and upheavals: this is where the full originality of LTS lies. Scenarios relating to safety over the long term have thus been specified. CEA took on board the most forceful outside aggressions (in terms, e.g., of an airplane crash), but chose to emphasize a scenario related to the societal environment, involving loss of technical control.

A long-term storage facility, by its very nature, is an installation that must remain under the control of society. That society may be faced with troubles forcing it to curtail, or relinquish this role. The scenario considered includes the interruption of all maintenance activity at the facility, and surveillance and monitoring restricted to the presence of guards, over a period possibly extending to ten years or so. In such conditions, the facility must remain safe and, if required, provide society with sufficient lead time to allow it to deploy corrective actions, before serious consequences occur.

What are the items to be put in storage?

The aim for the research program is to investigate the full gamut of solutions, to enable all possible options, with regard to management strategy. The waste concerned by the French Act of 1991 comprises long-lived waste, including ILW-LL (long-lived, intermediate-level) waste, and HLW waste (high-level waste, or vitrified waste). The program equally takes on board spent fuel, which is not deemed to be waste in France, but may however be subjected to storage, pending reprocessing, or direct disposal, the latter option not being the one taken up by France, out of the various management pathways for spent fuel (see Figure 1).

Owing to the goal of retrieval, and – as will be explained further on – the consequences of a possible loss of technical control, it was decided to store packages, rather than loose waste. The first stage thus consists in specifying the inventory of such waste. The reference, for the purposes of investigation, is **ANDRA**'s Dimensioning Inventory Model (MID: *Modèle d'inventaire de dimensionnement*) (see *An inventory projecting into the future*, p. 14; and Box A), which allows consistency in investigations carried out in accordance with the various directions specified by the Act. Studies thus take into account **primary packages** (i.e. packages as supplied by industrial production) of ILW-LL and HLW waste, as of spent fuel.

On the basis of all of the elements provided in responses to the initial questions, the R&D effort carried out by CEA has made it possible to design long-term storage facilities, and indicate their performance. The latter must be suitable for an industrial storage facility, and, furthermore, allow safety over time to be taken on board. To meet all of these requirements, the investigations are based on three main elements. Control of aging, first of all: degradation processes over time must be ascertained, and modeled, for the various components of an LTS facility. This is the research aspect, as narrowly defined, of the investigations carried out by CEA. Then, qualification must be achieved, for the processes to be used during construction, and demonstration of performance. This development aspect mainly takes the form of setting up demonstrators and integrated test benches. Finally, facility and equipment design makes it possible, by integrating the findings from the previous two aspects, to quantify indicators such as safety, costs, and durability.

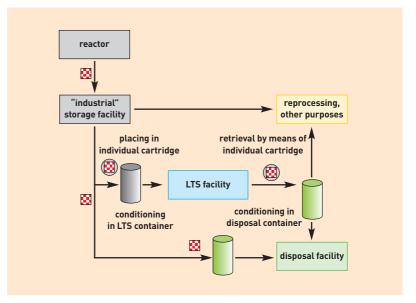


Figure 1.
The various spent fuel management pathways.

What are the design principles involved?

Due to the ensemble of performance requirements, and, in particular, with regard to the loss of technical control scenario, a number of principles were decided on: From the very initial design stages, aging is be taken on board, through use of components for which degradation over time can be evaluated, along with monitoring and maintenance issues, with the prospect of limiting the burden passed down to future generations. Robustness with respect to unforeseen circumstances relies, on the other hand, on a design and construction based on the simplest, and most passive,(1) components technically feasible. One final principle: the greatest possible facility inertia, allowing a large lead time to be available, between the time an operating anomaly is detected, and the time when possible disruption may occur, that would compromise facility safety, allowing corrective actions to be deployed, to enable a return to the design regime.

(1) By "passivity" is meant the fact that only components functioning in a natural manner are used, not involving moving parts.



Storage container demonstrator, with the ability to hold 7 UOX assemblies (see Figure 3).



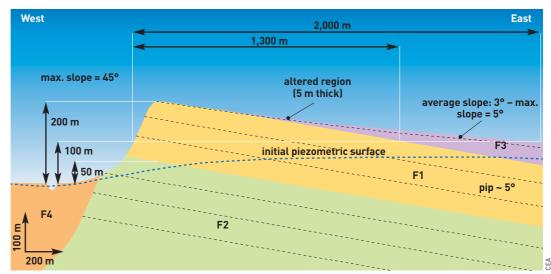


Figure 2. Cross-section of a generic hillside site (in hard rock) with the ability to accommodate a long-term storage facility.

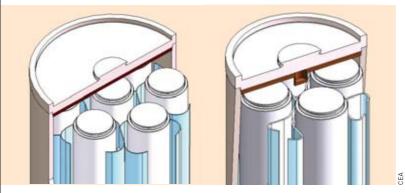


Figure 3.

Design for a canisters + container set, of 1.3-m diameter, able to accept 7 UOX assemblies, or 4 MOX assemblies, 5.2 m and 4.5 m in length, for a weight of 16 tonnes and 10 tonnes, respectively. A steel lid seals the 45-mm thick cast iron container, by means of a weld of great thickness, via a crown, also in steel.

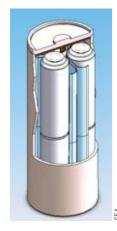


Figure 4.
High-level waste
package, comprising
6 standard vitrified waste
containers (CSD-Vs),
each forming the initial
confinement barrier,
placed in a container
fabricated to a design
derived from that
selected for spent fuel
(see Figure 3), of 1-m
diameter, for a length
of 2.9 m and a weight
of 10 tonnes.

Generic site studies

At the behest of the government, investigations considered two types of location for the LTS facility, **surface** and **subsurface**. No site having been earmarked on French soil, generic sites had to be defined, in particular for the subsurface configuration. For the latter case, a hillside location was considered, in hard rock (granite or limestone), above the aquifer, a situation that obtains in much of the country (see Figure 2).

Properties of primary packages and spent fuel

ILW-LL primary packages differ in terms of their nature (chemical composition of the waste, and of the encapsulation **matrices**), **container** type (concrete or steel materials), and dimensions. They release little, or even no heat. Some packages, on the other hand, may release small quantities of gas.

HLW packages comprise the **vitrified** waste packages yielded by reprocessing. The greater part of these consists in **CSD-V**s produced at La Hague. They are impervious, and release large amounts of heat, especially during the initial years after they are produced.

Spent fuel includes **UOX**, ERU (enriched reprocessed uranium oxide-based fuel), and **MOX**. Dimensions of these items are about the same, and they release large

amounts of heat. Indeed, the thermal power released by packages is one of the essential characteristics that must be taken on board in the design of an LTS facility, since it must be evacuated, to keep package temperature down.

For that reason, investigations are targeted, on the one hand, at non-heat-generating packages (ILW-LL packages), and, on the other, at heat-generating packages (HLW packages and spent fuel).

What are the selected design options?

Taking on board the design principles described above led to going for wholly static confinement of radioactive materials, this thus being entirely ensured by the stored package. This option entails securing spent fuel assemblies in individual, impervious **canisters**, since it is not possible to demonstrate fuel rod cladding integrity over timespans of centuries. This further involves placing such canisters and HLW packages, inside impervious containers, to ensure a second containment **barrier**, this being a safety requisite (see Box C). Finally, this entails placing a number of ILW-LL primary packages inside a container, the whole ensuring containment, while allowing venting of gases released by the primary packages, as mentioned earlier.

The main functions assigned to the storage facility infrastructure thus become those of accommodating packages, protecting them against outside aggression, evacuating thermal power and venting gases, and allowing package retrieval. CEA consequently designed canisters and containers, along with storage infrastructures, and demonstrated their functionality and durability.

Containers

The "canisters and container" design for spent fuel is shown in Figure 3. For reasons of thermal equivalence, the container is designed to accept seven UOX assemblies, or only four MOX assemblies. The individual cartridge is fabricated from impervious welded stainless steel. This involves well-mastered industrial technologies. CEA has developed a cast-iron container, which may be sealed by means of a weld of great thickness, via a steel crown. The advantage of cast iron essentially lies in the homogeneity achieved for the objects

(body and lid being welded solid), but equally in its low fabrication cost. To verify the issues had been fully investigated, even though this did not represent the reference solution, a spent fuel disposal container was developed, under the aegis of an ANDRA–CEA–EDF agreement.

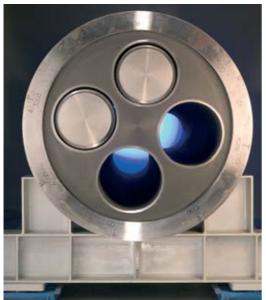
For HLW packages, the primary container forms the initial confinement barrier. The second barrier is provided by a container using a design derived from the spent fuel design, with a capacity to hold 6 CSD-Vs (3 packages stacked in 2 levels) (see Figure 4).

For ILW-LL waste, a ceramic primary container was developed for chemically aggressive waste, together with a concrete external container, holding one or more primary packages (see Figure 5). This container, common to LTS and disposal operations, was designed in collaboration with ANDRA. Its dimensions may vary, according to primary package geometry.

Storage facilities for spent fuel and HLW packages

In a surface facility, containers are held in dry storage vertically, in concrete bunkers covered by a roofing slab (see Figure 6). Cooling is ensured by natural convection (chimney effect), thus being fully passive. Atmospheric corrosion processes are retarded by keeping a low, constant atmospheric humidity, by means of the heat released by the containers, these being indeed cooled, in controlled fashion however, to preclude condensation forming on their walls. This allows the thickness of corroded wall to be estimated, after 300 years, at 350 mm maximum, compared to the 45 mm container wall thickness (i.e. 8%). The bunkers are of modular design, to be adaptable to the inventory and loading flows. The concrete used, and design dimensioning, make for stability of the structures over the anticipated timespans, provided classic monitoring and maintenance for this type of structure are adhered to.

For subsurface storage, the generic site selected is a hill, accessible along its side, allowing level access to the network of galleries and wells (see Figure 7) that will accommodate the packages (modular design). Each





Drop test, from a height of 6 m, carried out at CEA/Cesta, with a concrete container demonstrator (common to long-term storage and disposal operations), designed to hold 4 bitumen drum primary packages. The lid remained integral with the container body, and main damage was restricted to the impact area.



Figure 5.
Concrete container,
common to long-term
storage and disposal
operations, designed
to hold one or more primary
packages. For 4 bitumen
drums, this exhibits
a square section, 1.54 m per
side, for a height of 1.40 m
and a weight of 10 tonnes.
Thickness varies from
100 mm to 150 mm.





Figure 6.
Design for a surface storage facility, for dry storage of spent fuel and high-level waste packages. One bunker measures 110 m x 50 m.

Disposal containers, compatible with storage operations (compatibility being ensured by presence of a canister), for 4 fuel assembly canisters). Their weight reaches 45 tonnes.



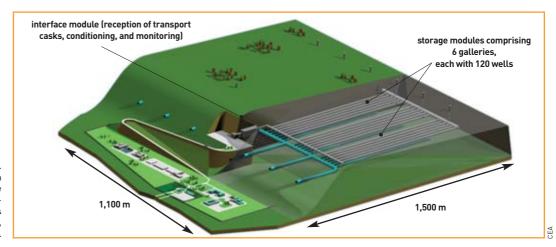


Figure 7.
Artist's impression
of a subsurface storage
facility for heatgenerating packages
(high-level waste,
or spent fuel).

well holds two containers, these being cooled by natural convection in this case also. Cool air intake is through the bottom of the well, from a lower gallery, the warm

air being exhausted by way of an upper gallery, and vent stacks (see Figure 8). The site ensures natural protection for the packages, against outside aggressions,

HERA, a full-scale LTS components and facility demonstration

Demonstration of performance, for a long-term storage facility as for containers, relies on actual constructions, and experiments, at a scale representative of the structures considered. CEA has therefore set up, at the Marcoule Center, a dedicated facility, HERA (Hall d'essais pour l'Entreposage des matières RAdioactives – Test Hall for the Storage of Radioactive Materials), comprising two experiment chambers, and a segment of subsurface gallery, known as Galatée (for GALerie Activités de Tests pour l'Entreposage – Storage Test Activity Gallery). The halls bring together the

containers fabricated for the demonstration program, the associated functionality and durability test benches, and the thermics and air engineering (aeraulics) experiments for the storage facilities. The gallery is a representation of a subsurface LTS facility, including its handling aspects, and accommodates a full-scale experiment on the behavior of an actual infrastructure subjected to thermal loading.

Containers

Fabrication of container demonstrators was carried out in two stages. In 2002, the first functional demonstrators were

container for spent fuel storage, polymer, enameled steel and ceramic prototypes for chemically aggressive ILW-LL waste, and two concrete containers for ILW-LL primary packages. The second stage covered fabrication of technological demonstrators, corresponding to the reference solutions selected: ceramic container, concrete container (common to storage and disposal operations, developed in collaboration with ANDRA), and cartridge and container for spent fuel storage.

fabricated: two canisters) and one

To verify the issues had been fully investigated, spent fuel disposal containers were fabricated, under a partnership with ANDRA and EDF.

All of these demonstrators, including sealed segments, were fabricated using industrial tools, and, after characterization and audits (control of dimensions, homogeneity...), the first qualification stage was passed. Functionality and durability are subsequently being demonstrated, either through direct experiments on the items involved (drop tests on ILW-LL containers, for instance, carried out at CEA/Cesta), or on representative-scale samples.



Bandy test bench used for the measurement of hydrogen diffusion through the concrete lid of an ILW-LL container. The sample is secured between two cells, respectively filled with hydrogen (or, for safety reasons, with helium, this exhibiting similar behavior) and nitrogen. Measurement over time of hydrogen concentration in the downstream compartment allows the sample's diffusion coefficient to be specified. This modular bench offers the specific characteristic of allowing use of samples representative of a container, in terms of size, concrete formulation, reinforcements, but equally of offering the ability to place them in the actual environment a container would be subjected to [measurement without the downstream cell].

Functionality and durability trials

In the HERA halls, some of the trials are carried out, concerning functionality and durability. For HLW waste and spent fuel, investigations are aimed more particularly at aspects relating to corrosion and thermal aging. The former point is being addressed at CEA/Saclay, who are carrying out R&D on dry corrosion regimes and atmospheric corrosion regimes, resulting in the drawing up of models,

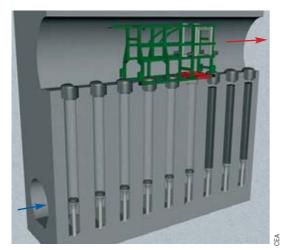


Figure 8.

Detail view of a group of 9 storage wells for high-level waste, with a depth of 14 m. Cool air intake is shown in blue, warmed air exhaust in red.

and, as with the surface design, corrosion of packages is limited, owing to their temperature.

ILW-LL packages

In surface storage, ILW-LL packages are placed in facilities of the type shown in Figure 9, with the possible addition of a radiological protection slab, for the more heavily irradiative waste. Venting of gases is ensured by natural means, through apertures, and by taking wind into account, which, however weak, ensures the facility's internal atmosphere is constantly refreshed.

In the subsurface case, packages are stored in pits (see Figures 10 and 11), accessible via circulation galleries. Since investigations are based on a generic site, it is not possible to demonstrate the effectiveness of a natural ventilation system in the wells and galleries. The design therefore provides for a forced ventilation system (mechanical fans), reaction time, from the moment of

the findings of which, regarding the spent fuel storage container, were mentioned earlier. With respect to thermal aging, a specific platform was set up. Its purpose is to monitor the evolution of the properties of metallic materials (stainless steel, carbon steel, and cast iron), subjected to a temperature environment over a period of ten years. The findings from tests carried out on representative samples and mockups, initiated in 2002, corroborate the predictive models.

For ILW-LL waste, the demonstration is concerned with the ceramic primary package, for which a durability test bench is being built (laboratory research tests on samples having warranted the finding that the tested solution is indeed feasible), and the concrete external container, for which two specific test benches are to enable qualification of gas diffusion through the lid (this being the design option selected), and primary package protection by the container, with regard to chemical aggressions (chlorides). Investigations on concrete behavior are being carried out at Saclay.

Thermics-aeraulics demonstration

Demonstration of long-term storage facility functioning may be said, in simplified terms, to come down to the thermics-aeraulics (functioning) demonstration, and demonstration of infrastructure durability. The latter point is addressed in Galatée

The thermics-aeraulics demonstration for the surface case (cooling across a bunker) is being carried out in Grenoble, on the Valida representative mockup. For the subsurface case, two points need to

be investigated: the well, and the galleries. Two demonstration experiments have thus been set up: Prométhée, this being representative of a full-scale well, for the measurement of transfer coefficients between the container and the well's atmosphere, and Sigal, a 1/20 scale model, to measure pressure losses at the intersections of a network of galleries.

Prométhée, which was initiated owing to the dearth of data in the literature, has allowed investigation of the thermics aspects relating to natural convection inside a storage well. The test bench comprises a heating body, representative of a container, an enclosed space corresponding to the well, and a set of measuring equipment (200 thermocouples and two flowmeters). This experiment has allowed the determination of convective transfers between the container and the well's atmosphere. These findings were integrated into the models used for in-well storage package behavior simulation.

Galatée

Demonstration of the durability of infrastructures subjected to thermal loading is carried out in a segment of subsurface gallery, representative of an LTS facility for spent fuel and HLW waste. As a matter of fact, this gallery has two main purposes, illustration and demonstration. The segment is a full-scale representation of the upper gallery that allows container handling, and the exhaust of warm air from the storage wells (see How may storage and the long term be made compatible?). A mockup of the handling device allows simulation of well

loading, and validation of design options. The second part of this segment is fitted with sensors submerged in the concrete, and a heating system, the aim being to achieve an ambient atmosphere at a temperature of about 80 °C, this being the maximum value obtaining in an actual LTS facility. The roof will thus be subjected to the stresses generated by that temperature. The sensors (temperature, humidity, displacement) will allow structure evolution to be measured over timescales of the order of one year. A test wall will allow physical analysis of the evolution of the concrete itself.



Trial platform for durability tests for metallic materials subjected to a temperature environment. The trials, initiated in 2002, have the aim of monitoring the evolution of properties over time.

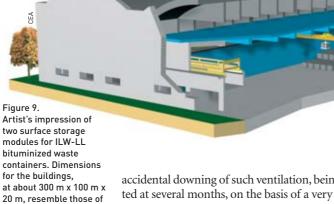
New designs: the package at the core of investigations



civil engineering structures. Each building

houses from 7 to 10

storage modules.



accidental downing of such ventilation, being estimated at several months, on the basis of a very generous gas source term estimate. In the event of an actual LTS facility being built, ventilation studies, carried out at the actual site, will allow possible consideration of natural ventilation, on the lines of what pertains in some caves. Here again, the design is modular.

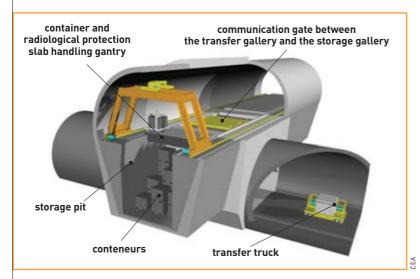


Figure 10. 3D view of the intersection of the gallery used for transfer of ILW-LL waste containers with a storage gallery, in a subsurface design (gallery dimensions: $12 \text{ m} \times 13.5 \text{ m}$).

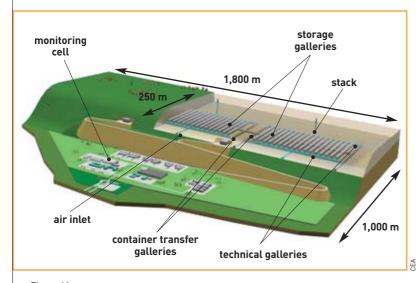


Figure 11.

Artist's impression of a subsurface storage facility for ILW-LL waste, comprising 3 blocks of 17 galleries (only two blocks are shown).

From research...

back, and, on the other, on knowledge of the processes and evolution kinetics involved, over timespans of centuries. Investigation of the aging of components in an LTS facility thus considered primary packages (HLW and ILW-LL waste, and spent fuel) in storage configuration (i.e., kept dry), the storage containers that have been developed, and the infrastructures. This laboratory investigation allowed models of behavior to be arrived at, and validated, for all of these components (evolution of mechanical properties under irradiation or thermal loading, corrosion...).

Assessed durability, for a component, is based, on the one hand, on operational feed-

... through development...

The goal of this segment of R&D is the qualification of the technological processes that will be used for the fabrication of containers, and long-term storage facility construction. The first stage is concerned with achieving mastery of these processes (fabrication, sealing...), through the setting up of representative demonstrators, the second with qualification of functionality, and the third with demonstration of durability. A technological demonstration platform has been set up at Marcoule, to accommodate much of this development work (see Box).

... to integration

Integration is concerned with the design as such, as described above, but equally with the quantification of indicators, particularly with regard to safety. CEA has drawn up safety direction dossiers (setting out the goals and providing an analysis of the facility with respect to internal and external risks), and techno–economic dossiers. The latter aspect obviously takes in the costs of facility construction, and containerization, but equally operating costs. To specify these, the studies included monitoring and maintenance operations, these also forming part of the demonstration of safety. All of these design studies were brought together in a design-definition study dossier, issued at the end of 2005.

A basis for decision

This research program, the outcome of an effort involving CEA departments, in collaboration with universities, laboratories, design bureaux and manufacturers, has made it possible to show that the long-term storage option, for spent fuel, HLW waste and ILW-LL waste, is a solution for which deployment is available for a decision. This demonstration, taking on board as it does the feedback from current industrial storage facilities, is based on an ensemble of predictive models of component behavior in an LTS facility, together with the qualification of the processes to be used for construction of the structures.

> Jean-Paul Silvy

Nuclear Energy Division CEA Valrhô-Marcoule Center

What is radioactive waste?

ccording 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 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)
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 Haque 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 lowlevel 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.

	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 m³) and graphite waste (volume: 14,000 m³)							
Intermediate-level waste (ILW)	Capacity: 1 million m ³	MID volume estimate: 78,000 m ³							
High-level waste (HLW)	MID volume esti	e 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)

Waste from the nuclear power cycle

ost 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

1 H																	² He
3 Li	⁴ Be											⁵ B	်င	⁷ N	8	⁹ F	Ne
Na	Mg	=									13 Al	14 Si	15 P	16 S	17 Cl	18 Ar	
19 K	²⁰ Ca	21 Sc	Ti	23 V	Cr	Mn	Fe	27 Co	Ni Ni	Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	Kr
Rb	38 Sr	39 Y	Zr	Nb	42 Mo	43 (Tc)	Ru	45 Rh	Pd)	Ag	48 Cd	49 In	50 Sn	51 Sb	⁵² Te	53	Xe
55 Cs	56 Ba	Ln	72 Hf	⁷³ Ta	74 W	75 Re	⁷⁶ Os	⁷⁷ Ir	78 Pt	79 Au	Hg	81 TI	Pb	83 B i	84 Po	At	Rn
87 Fr	Ra	An	104 R f	105 Db	106 Sg	107 Bh	¹⁰⁸ Hs	109 M t	110 Uun								
lanth	anides	57 La	⁵⁸ Ce	59 Pr	Nd	61 Pm	62 (Sm)	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	⁷⁰ Yb	71 Lu	
actin	actinides																
C	■ heavy nuclei ■ activation products ■ fission products ○ long-lived radionuclides																

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).

(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.

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

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 (E ²³⁵ U	GWd/tiU : 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)
	234	246,000	0.02	222	0.02	206	0.02	229	0.02	112
U	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
	238	87.7	1.8	166	2.9	334	4.5	590	3.9	2,390
Pu	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

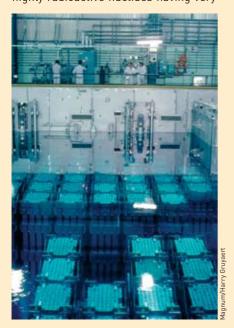
Table 1.

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.

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 (14C), 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 (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 Haque.

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

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.

What stands between waste and the environment?

aw, solid or liquid radioactive waste Tundergoes, after characterization Idetermination 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 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 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.



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, 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

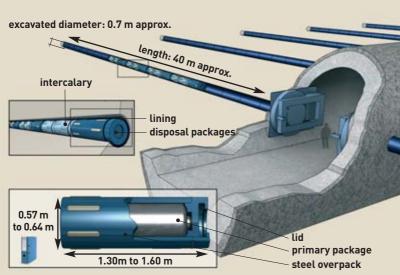
recycling-transmutation, or simply to turn to advantage the natural decay of radioactivity (and hence the falling off of heat release from high-level waste), before putting the waste into geologi-

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 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 wellspecified 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.

ANDRA

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...

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.

[(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-sec-

tion 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 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.

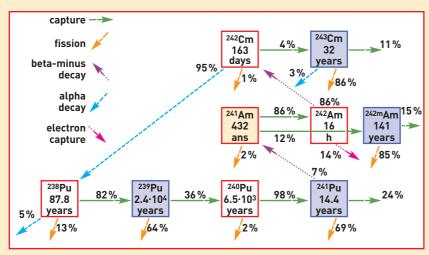


Figure. Simplified representation of the evolution chain of americium 241 in a thermal-neutron reactor (shown in blue: radionuclides disappearing through fission). Through capture, ²⁴¹Am transforms into ^{242m}Am, this disappearing predominantly through fission, and into ²⁴²Am, which mainly decays (with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

What is an ADS?

n 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 Principle schematic of an ADS.

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 keff, 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 keff 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 keff 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.

window spallation accelerator target providing **->**-100 keV external neutrons proton source subcritical reactor

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.