The indispensable **disposal** of **ultimate waste**

Whichever management strategy is selected, disposal of ultimate waste will be a requisite. Disposal in a deep geological environment stands as a reference solution, to ensure confinement over several tens of thousand years, or even longer, and to mitigate possible loss of memory by society. CEA is contributing to the research effort, steered by ANDRA, particularly with respect to the issues of identifying, and modeling, the processes governing the long-term evolution of such a disposal facility.

Whichever **radioactive waste** management strategy society goes for, it will remain indispensable to protect the **biosphere** from the residual **ultimate waste**. The nature of the **radioelements** this contains requires a solution that has the ability to ensure their **confinement** over several tens of thousand years, or even longer. On such timescales, social stability is a major uncertainty, which may neither be evaded, nor easily anticipated. Which is why **disposal** in a **deep geological** environment stands as one of the reference solutions, insofar as this inherently allows, by its very design, to mitigate a possible loss of memory on the part of society. Indeed, advances achieved by research show that certain carefully selected geological environments may remain stable, retain their properties, and prevent dispersion of toxic elements for millions of years.

CEA has been contributing to the research effort on the disposal of radioactive waste in a deep geological environment for several decades, this being steered since 1992 by **ANDRA**. These investigations, carried out under the aegis of a partnership agreement, prorogued at regular intervals, and jointly funded, on an equal basis, by both organizations, have mainly focused on the identification, and **modeling**, of the processes governing the long-term evolution of a geological disposal facility. They are concerned more particularly with the design of **engineered barriers**, and the long-term evolution of their properties, characterization and modeling of **radionuclide** behavior in a disposal facility, and development of global modeling tools for the evolution of a disposal facility.

To this should be added the ensemble of investigations on the long-term behavior of waste **packages** in disposal conditions (see *What long-term behavior for nuclear waste packages… and for spent fuel?*).

Long-term evolution of engineered barrier properties

In a deep geological disposal facility, waste packages may, as and when required, be surrounded by a backfill material, known as an *engineered barrier*, having the purpose of restricting water flows around the packages, filling technological voids, delaying package degra-

MJM Production/ANDRA

Constructed at a depth of 500 m, at the boundary of the Meuse and Haute-Marne *départements* (North-Eastern France), in an argillaceous layer with an age of 150 million years, ANDRA's Underground Research Laboratory is a multidisciplinary research instrument, for the investigation of disposal conditions in a deep geological environment for long-lived radioactive waste. Shown here, an experiment carried out at the bottom of the shaft.


CEA

Clay for engineered barriers, compacted in the form of pellets, to facilitate its eventual setting in position inside a disposal facility. The swelling caused by resaturation with water of this material makes it possible to guarantee filling of the voids initially prevailing between pellets.

dation, and, finally, delaying subsequent radionuclide migration, once the package has degraded. The materials must exhibit good thermal–hydro–mechanical (THM) properties, to preclude their becoming destabilized, or degraded, owing to the evolutions over time in temperature, water content, or water chemical composition. Materials thus being considered for such barriers are cement materials, or swelling argillaceous materials.

CEA has been carrying out investigations for over 15 years to characterize, and, most crucially, achieve better understanding of THM property of these materials, while analyzing their degradation modes. The aim of these investigations is to develop robust **models**, allowing predictions of THM property evolution and possible degradation over time, with the ultimate goal of being in a position to vouchsafe their functions may be sustained over the very long term (see Figure 1). On the basis of this expertise, CEA

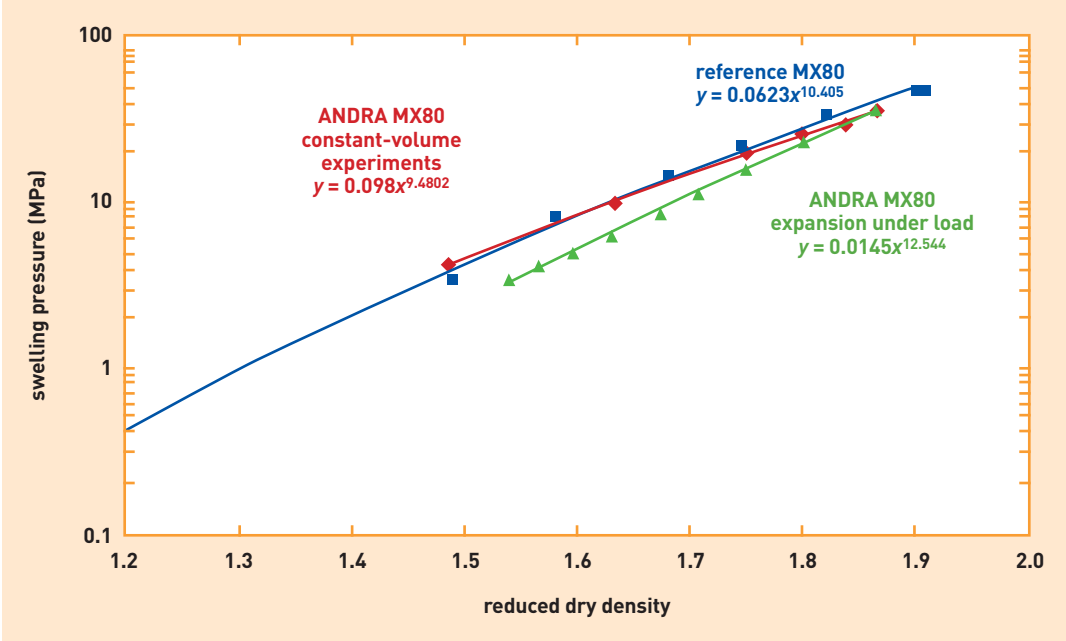


Figure 1. Evaluation of the swelling pressure of a reference clay for engineered barriers (MX80) as a function of its density. Clay's ability to swell when undergoing resaturation by water from the site makes it possible to guarantee filling of all technological voids occurring around waste packages.

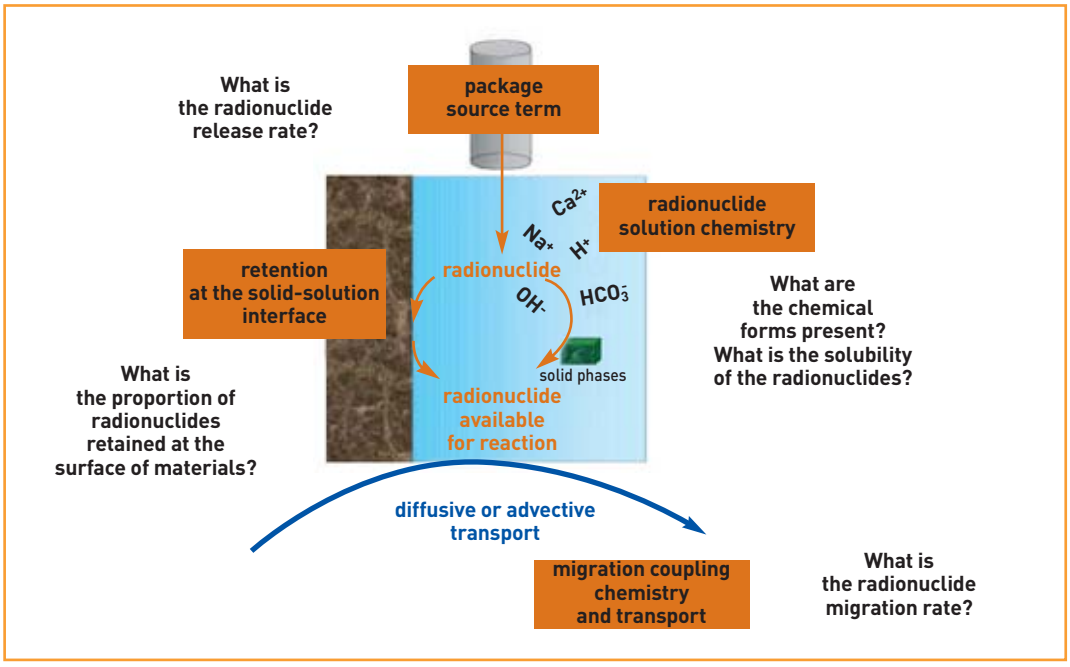


Figure 2. Migration of radionuclides through a complex, porous environment is governed by radionuclide release rate, their speciation in solution, retention at solid–solution interfaces, and by the extent of migration processes, these being overwhelmingly diffusive, in a disposal situation.

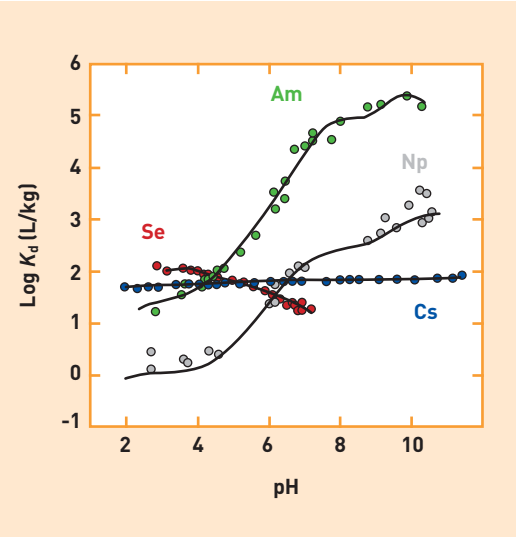


Figure 3.
Evolution of the extent of retention for various radionuclides (americium [Am], neptunium [Np], selenium [Se], and cesium [Cs]), at the surface of a simple argillaceous mineral (smectite), as a function of **pH**. Retention is expressed as a logarithm of the distribution coefficient, this being the ratio between **sorbed** quantity and dissolved quantity. Retention of cations (Am, Np, and Cs) rises with pH, whereas that of anions (Se) falls, owing to the evolution with pH of the chemical form of surface sites bearing hydroxyl –OH groups. Cesium retention, on the other hand, is solely governed by the permanent surface charges exhibited by clays, thus being virtually independent of pH.

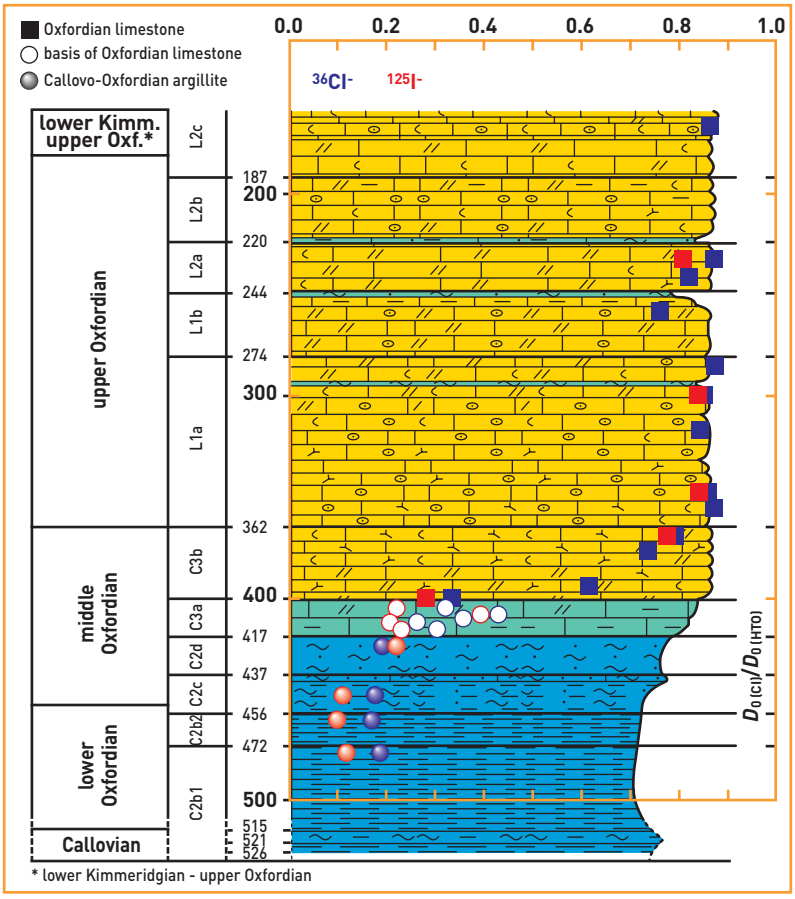


Figure 4.
Evolution of the effective diffusion coefficient of iodine $^{125}I^-$ (shown in red) and chlorine $^{36}Cl^-$ (blue), normalized with respect to the effective diffusion coefficient of HTO **tritium**, as a function of depth, in the East of the Paris Basin (at the site of ANDRA's Meuse–Haute-Marne Underground Research Laboratory). The steep falloff observed below a depth of 400 m correlates with increased presence of argillaceous materials, and existence of an electrostatic repulsion between the negatively charged surface of the clays and the anions.

further carried out, concurrently, investigations of a more technological nature, on the various options for use of such materials, in particular argillaceous materials.

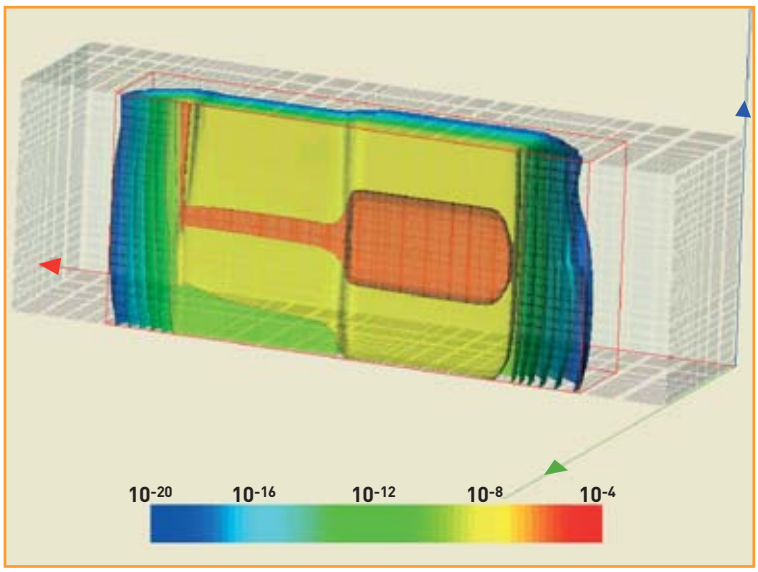
Characterization and modeling of radionuclide behavior in a geological disposal facility

Over the very long term, waste **containers** will unavoidably corrode, under the effects of interstitial water, present in geological environments. Thereafter, the nuclear waste packages will gradually degrade under the effect of water, at very slow rates, as shown by investigations on the science of long-term package behavior (see *What long-term behavior for nuclear waste packages… and for spent fuel?*). This will result in the release of small amounts of radionuclides, around the waste package. The subsequent evolution of these radionuclides will then be governed by three main processes (see Figure 2).

The first process results in the emergence of complex aqueous species, containing the radionuclide and other elements naturally present in the water (carbonate CO_3^{2-}, sulfate SO_4^{2-}, silicate SiO_4^{4-}… **ions**). Thus, the radionuclide will not always remain as an isolated ion in the water, rather it will often form part of more complex molecular structures, the nature of which will depend on the environment. Identification of these chemical species (i.e., ascertaining *speciation*) is crucial, insofar as radionuclide mobility depends on what species are actually present. In this area, CEA has concurrently carried out methodological development work, to avail itself of analytical tools providing the means, in a given environment, to ascertain the various species actually present, and more theoretical investigations, to establish the thermodynamic constants enabling predictions of speciation in environments of such complexity. These constants, once validated, are then integrated into databases of thermodynamic data, the consistency of which is verified, and on which all chemical modeling is based. CEA is thus contributing to the setting up of an international database on **actinide** and **long-lived fission product** chemistry, under the aegis of the **Nuclear Energy Agency**.

The radionuclides released by package degradation reach an ambient medium which is a compact, though porous, material (concretes, clays, rock), its porosities being saturated with water. Radionuclides within these pores (a few **microns** in size) will possibly interact with the surface of argillaceous materials, and become fixed on it. This second process, known as *retention*, contributes very extensively to the confinement capacity of natural rock. CEA is carrying out numerous experiments to establish relevant data, but equally to develop theoretical models. The extent of retention of the main radionuclides on natural or artificial materials present in a disposal facility has thus been measured, depending on the various physical–chemical conditions that may be encountered (see Figure 3). The theoretical models allow predictions of the quantity of radionuclide retained on the surfaces of materials. The findings are then used as input data for the migration models, to evaluate the extent of this process, in the course of safety analyses.

Finally, radionuclides remaining as a solution will gradually *migrate* away from the package, this being the



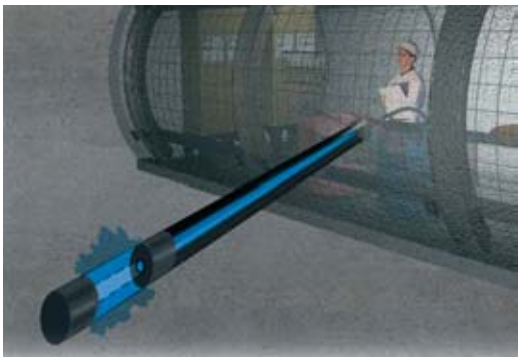
CEA

Figure 5.
Iodine 129 concentration, in mol/m³, around a **spent fuel** package, after 50,000 years, in the case of an altered evolution scenario for a disposal facility. This altered scenario corresponds to the unlikely event of faulty sealing in the galleries.

third process, at a rate that depends on their physical characteristics, and the properties of the rocks in the geological environment. Negatively charged species (anions) further exhibit the special characteristic of only accessing a restricted region of pore space, owing to the existence of an electrostatic repulsion, prevailing with the surface of negatively charged argillaceous materials. In this respect also, CEA, acting as a privileged partner of ANDRA, has developed specific means of characterization, to assess the diffusion properties of the various materials present in a disposal facility, and in the host rock (argillites). Diffusion experiments were carried out, at scales ranging from that of samples measured in centimeters, to that of geological formations (see Figure 4).

Overall modeling of a geological disposal facility

Prudent assessment of very-long-term evolution for a disposal facility entails availability of modeling tools that take into account all of the processes involved. Simplifications are required, not only if computations are to be completed in a reasonable time, but also to avoid arriving at models of such complexity it might hamper achieving a critical perspective, with respect to physical reality. CEA has jointly developed, with ANDRA, from the beginning of the present decade, a modeling platform, specifically dedicated to **storage** and, most importantly, to deep geological disposal. This tool, dubbed ALLIANCES (*Atelier logiciel d'intégration d'analyse et de conception pour l'entreposage et le stockage* – Software Workshop for Integration of Analysis and Design of Storage and Disposal Facilities), allows modeling of the main THM and chemical processes active in a disposal facility, particularly those involved in long-term package alteration, radionuclide release, and radionuclide migration within the **near field**, then in the geological environment (see Figure 5). This has been used by ANDRA to carry out safety calculations, to support its final report on disposal, under the aegis of the French Act of 30 December 1991 (see Box 2, in *Radioactive waste management research: an ongoing process of advances*).



ANDRA

ANDRA

DIR experiments, carried out in the experiment niche, at – 445 m, of ANDRA's Underground Research Laboratory, at Bure (Meuse–Haute-Marne *départements*). Top, the experiment design selected, with injection of tracing agents at the bottom of a borehole. Bottom, experiment cabinets, in which sampling and *in situ* monitoring are carried out, for the experiments currently conducted in the experiment niche.

Underground laboratory experiments

The findings from investigations carried out in the laboratory, on samples taken at various depths (core samples) require validation, and confirmation, at the scale of the geological environment, and in real conditions. This is one of the reasons for the construction, by ANDRA, of an underground research laboratory, at the Bure site, at the boundary of the Meuse and Haute-Marne *départements* (North-Eastern France). CEA is taking part in some of the experiments carried out there, those in particular aimed at confirming the properties of radionuclide diffusion and retention in the geological formation (DIR experiments: *Diffusion de traceurs Inertes et Réactifs* – Diffusion of Inert and Reactive Tracers). Thus, after completing design and dimensioning of these tracing experiments in the underground laboratory, CEA teams are currently conducting them, and interpreting them. Initial findings corroborate the models developed from core samples.
CEA thus stands as a privileged partner of ANDRA, for investigations on deep geological disposal, concerned as it is, on the one hand, with meeting operational requirements, and, on the other, with carrying out relevant, longer-term methodological and experimental development work, such as to assist ANDRA achieve deeper understanding of the evolution of a deep geological disposal facility.

> Christophe Poinssot
Nuclear Energy Division
CEA Saclay Center

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 m^3 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 m^3 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 m^3 annually), technological waste from the operation or routine maintenance of production or fuel-processing plants, from nuclear reactors or from research centers (some 230 m^3 annually), along with sludges from effluent treatment (less than 100 m^3 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 m^3 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 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 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.

<table>
<tr><th></th><th>Short-lived
Half-life < 30 years
for the main elements</th><th>Long-lived
Half-life > 30 years</th></tr>
<tr><td>Very-low-level waste (VLLW)</td><td colspan="2">Morvilliers dedicated disposal facility (open since 2003)
Capacity: 650,000 m³</td></tr>
<tr><td>Low-level waste (LLW)</td><td rowspan="2">Aube Center
(open since 1992)
Capacity: 1 million m³</td><td>Dedicated disposal facility under investigation for radium-bearing waste (volume: 100,000 m³) and graphite waste (volume: 14,000 m³)</td></tr>
<tr><td>Intermediate-level waste (ILW)</td><td>MID volume estimate: 78,000 m³</td></tr>
<tr><td>High-level waste (HLW)</td><td colspan="2">MID volume estimate: 7,400 m³</td></tr>
</table>

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 10^{17} 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 (^{235}U). 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 burnup rate close to 50 GWd/t, result in bringing down final ^{235}U 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 ^{235}U 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 (^{232}U, ^{236}U), that had not figured in the initial fuel (see Table 1).

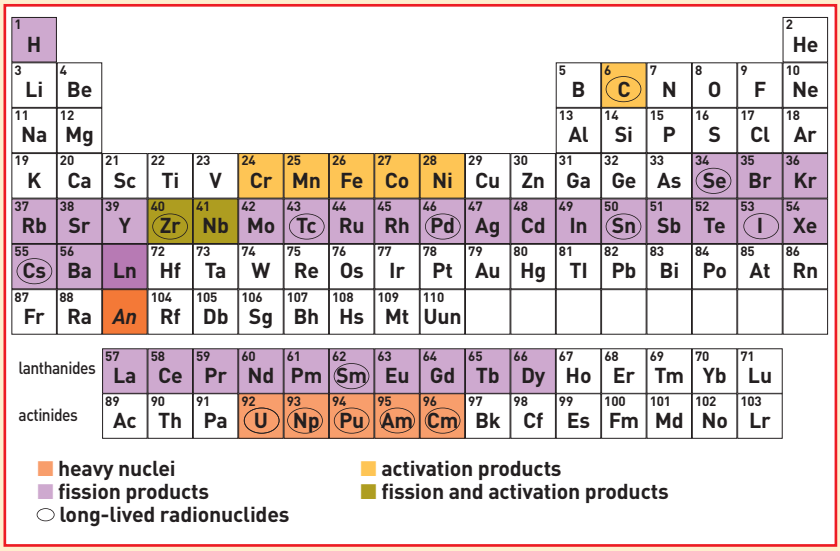


Figure 1.
The main elements found in spent nuclear fuel.

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 ^{238}Pu to ^{242}Pu, 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	$7.04 \cdot 10^8$	1.05	10,300	0.74	6,870	0.62	5,870	0.13	1,070
	236	$2.34 \cdot 10^7$	0.43	4,224	0.54	4,950	0.66	6,240	0.05	255
	238	$4.47 \cdot 10^9$	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	$3.75 \cdot 10^5$	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 (^{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 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 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.

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.

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.

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 **fission product** or **minor actinide** solutions). This matrix contributes to the confinement function. The waste thus conditioned is placed in an impervious **container** (cylindrical or rectangular), consisting in one or more **canisters**. The whole – container and content – is termed a **package**. Equally, waste may be compacted and mechanically **immobilized** within a canister, the whole forming a package.

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

A. Gonin/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.

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.

CEA

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 years, or tens of years).

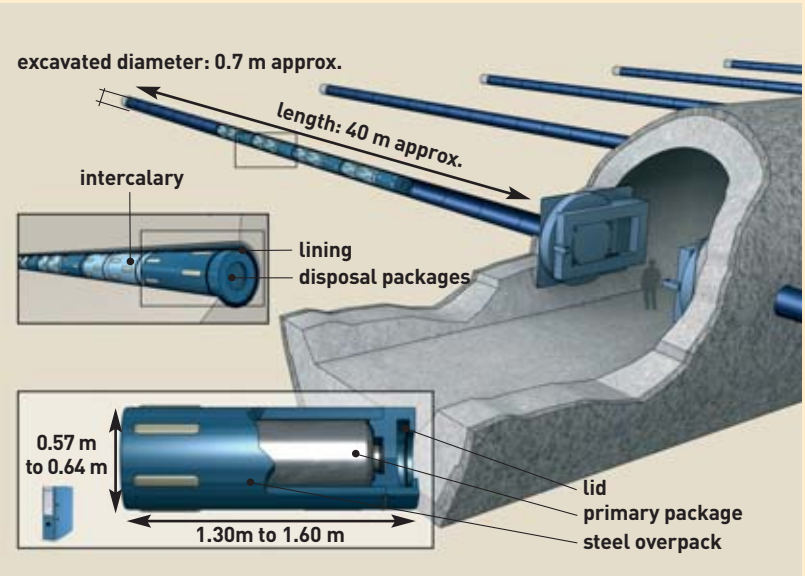
A. Gonin/CEA

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

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 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 hillside) and its host rock.

Whichever management strategy is chosen, it will be imperative to protect the biosphere from the residual ultimate waste. The nature of the radioelements the latter contains means a solution is required that has the ability to ensure their confinement over several tens of thousand years, in the case of long-lived waste, or even longer. On such timescales, social stability is a major uncertainty that has to be taken on board. Which is why disposal in deep geological strata (typically, 500 m down) is seen as a reference solution, insofar as it inherently makes for deployment of a more passive technical solution, with the ability to stand, with no increased risk, an absence of surveillance, thus mitigating a possible loss of memory on the part of society. The geological environment of such a disposal facility thus forms a further, essential barrier, which does not exist in the storage case.

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



ANDRA

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.

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 thermal 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 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.

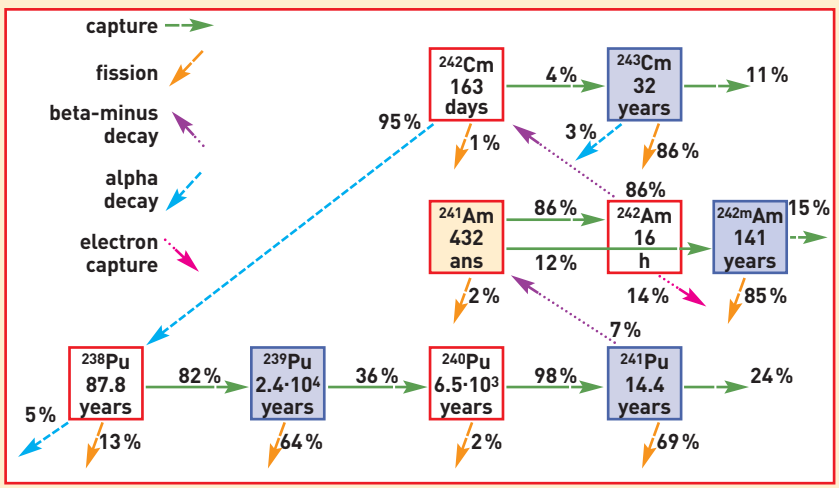
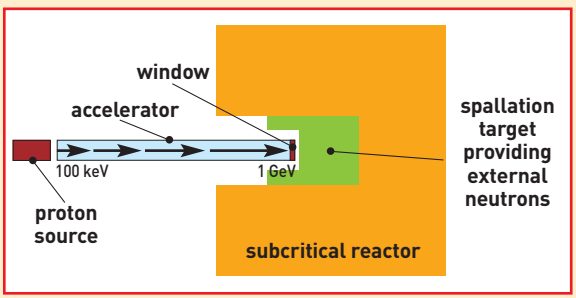


Figure.
Simplified representation of the evolution chain of americium 241 in a thermal-neutron reactor (shown in blue: radionuclides disappearing through fission). Through capture, ^{241}Am transforms into ^{242m}Am, this disappearing predominantly through fission, and into ^{242}Am, which mainly decays (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 $(k_{eff} - 1)/k_{eff}$. 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.

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

The industrial context

1

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.