

What **long-term behavior** for **nuclear waste packages**...



P. Stroppa/CEA

Investigations on the long-term behavior of high-level waste packages are carried out on the C18-C19 shielded line of the **Atalante** facility, at Marcoule.

Investigation of long-term behavior is the cornerstone of nuclear waste management studies. The timescales involved are beyond imagining, not however beyond scientific reasoning. On the basis of the major physical-chemical mechanisms that determine this behavior, scientists may derive, if not exact predictions as to the future evolution of waste packages, at any rate estimates, allowing quite substantial safety margins, of the extent of radionuclide release, for a variety of timescales.

For the purposes of dimensioning a **nuclear waste storage** or **disposal** facility, and quantifying its potential impact on the environment, investigations must be carried out as to the behavior of such waste, over timescales that may be counted in tens, or even hundreds, of thousand years (see Box D, **From storage to disposal**). Scientists have sought therefore to ascertain precisely the major physical-chemical mechanisms that determine this behavior. They have integrated these mechanisms into **models**, to obtain a projection over time, and arrive at conservative predictions, i.e. allowing for safety margins, of the source terms in storage or disposal conditions, for all types of **long-lived** intermediate-level, or high-level waste (**HLW** or **ILW-LL**), and for **spent fuel**. While such long-term extra-

polations do not allow making precise predictions as to the future evolution of waste **packages**, they do make it possible, nevertheless, to compare various possible options. These models are designed, as may be seen from their conservative character, for integration into performance models, for the purposes of safety calculations.

Timescales beyond imagining... but not beyond scientific reasoning

Research programs on the long-term behavior of radioactive waste packages are of a specific nature: they must enable extrapolations over very long timescales, of the order of some tens of thousand to seve-



Nanoscale to assist for the very long term

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In **storage** or **disposal** conditions, **waste packages** will be subjected to **radioactive** decay of **radionuclides**, tending to disrupt the containing **matrix** [**self-irradiation** effects]. The main sources of irradiation in nuclear glasses result from **alpha** (α) **decays** from **actinides**, **beta** (β) **decays** from **fission products**, and **gamma** (γ) **transitions** accompanying β and α decays. Radiation emission is concomitant with generation of heat. α decays, characterized by production of a **recoil nucleus**, and emission of an α particle (with an energy of 4–6 **MeV**), yielding a helium atom at the end of its path, are the main cause of atom displacements. In particular, recoil nuclei shedding large amounts of energy over a short distance result in atom displacement cascades, thus breaking large numbers of chemical bonds (depolymerization). This type of decay is thus the main cause of potential long-term damage. Understanding such processes on the **nanome-**

ter scale, or even on the atomic scale, makes for robust predictions of behavior over several hundreds of thousand years. Development of a method based on coupling between a binary-collision approach and classical **molecular dynamics** was required to compute atom displacement cascades at real energies (of the order of 70 **keV**) in simplified nuclear glasses. This novel method is intended as a substitute for pure molecular dynamics at high energies, to speed computation.

After a validation stage, on a 10-keV cascade, in which findings yielded by this method were seen to be consistent with those from the reference molecular dynamics, a 70-keV cascade was simulated [see Figures 1 and 2]. The readings are continuous over the energy range (0–70 keV). There is systematic temporary depolymerization, of intensity linear with energy, followed by gradual restoration of the glass structure. Numbers of atom displacements also show linear increase with projectile energy.

Structure restoration is not homogeneous, in that the less hot regions are less fully annealed (defect reconstruction is less extensive), compared with hotter regions. No alteration, whether in polymerization or atomic density, remains after passage of the cascade through the warmer regions, whereas an atomic density deficit, sometimes associated with residual depolymerization, may occur in the cooler regions, i.e. at the extremities of the cascade. Local agitation in the warmer regions is swiftly dampened as it propagates, only minimally affecting adjoining regions, hence the less satisfactory annealing achieved in regions subjected to intermediate-energy projectiles.

Further future use of this method is being considered, on the one hand, to achieve a more detailed understanding of the effects of recoil nuclei in glasses, and, on the other, to investigate ways of transferring such understanding to other oxide confinement matrices.

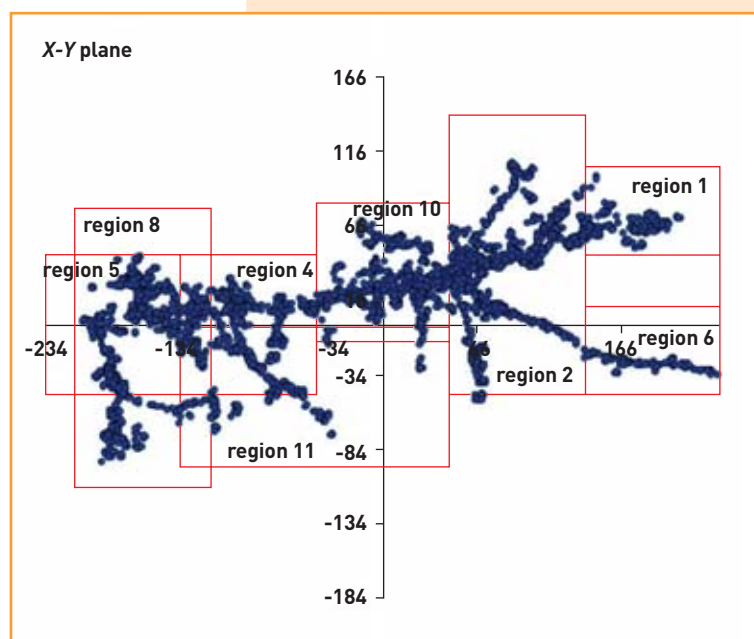


Figure 1. Morphology of a cascade initiated by a 70-keV recoil nucleus. Dots stand for the ensemble of displaced oxygen atoms. Distances are shown in angstroms (10^{-10} m). Only part of the partitioning into regions is shown.

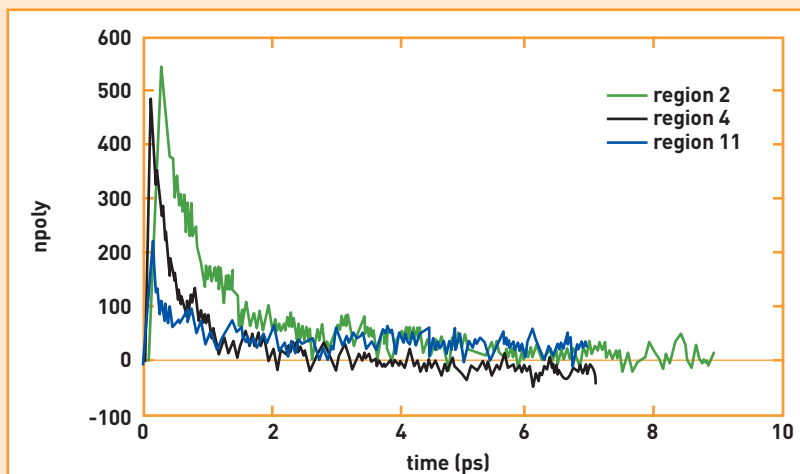


Figure 2. Evolution of depolymerization over time in three regions selected to illustrate the diversity of behaviors. npoly stands for the number of chemical bonds in the glass lattice that are opened at a given time. A positive npoly value thus indicates glass depolymerization.

ral hundred thousand years, to arrive at an opinion as to the durability of the functions assigned to the packages, in the main situations being contemplated (storage, **geological disposal**). In such conditions, mere extrapolation of laboratory findings is inadequate. This must be complemented by the understanding, identification, and hierarchical ranging of the various physical–chemical processes involved, within the packages, while taking on board their interaction with the environment. Knowledge of the mechanisms driving the packages’ long-term evolution is used to draw up operational models, allowing predictions of long-term package behavior, with regard to the essential functions assigned to them. These functions include **confinement**, on a scale of centuries, in long-term storage conditions (see *How may storage and the long term be made compatible?*), the ability to retrieve waste packages after long-term storage, or during the **reversible** disposal stage, and confinement on a scale of thousands of years in geological storage conditions.

A methodology has been developed, to ensure rigorous scientific exactness for predictions of long-term behavior. This approach combines *experiment* (external irradiation, or investigation of **doped** materials, **leaching** in aqueous solutions, or in the presence of various environmental materials, use of numerous techniques for the fine analysis of condensed matter), and **modeling** (from **molecular dynamics** to more integrated models, describing the evolution of a package in a disposal facility) (see Box 1). Further, it includes an aspect of investigation of archeological or geological analogs, such as, e.g., volcanic glasses or the elements found in the Oklo (Gabon) natural nuclear reactors. Characterization of these various materials provides supporting evidence for the validation of long-term behavior models (see Box 2). A variety of environmental conditions are taken on board in these investigations: the closed system, water-unsaturated open system, and water-saturated open system cases.

In the *closed system* case, the package exchanges only energy with the outside, in the form of heat or **radioactivity**. The system may also be subjected to external mechanical stress. On the other hand, it exchanges no solid or liquid matter with the outside environment. Such conditions are the nominal in-storage operating conditions. They are also the conditions prevailing during the initial stage of deep geological disposal, as long as the **container** has not corroded.

In the *water-unsaturated open system* case, degradation of the container has occurred, and the package may thus exchange matter with the outside (along with the energy exchanges mentioned for the closed system case). Package alteration is governed by exchanges of matter with the aqueous and gaseous phases coming into contact with the package. This represents, e.g., degraded storage conditions.

The *water-saturated open system* is an extreme case, where the container has lost all of its confinement properties. The system thus exchanges energy and matter with the surrounding medium, essentially through alteration of the aqueous phase. This represents the nominal long-term disposal situation, after

degradation of the various **confinement barriers**, and after the package has come into contact with water from the site.

The types of package investigated include packages of cemented waste, bituminized sludge, vitrified waste, and spent fuel, and standard compacted waste packages (CSD-Cs) (see Box A, *What is radioactive waste?*).

The findings arrived at subsequent to the Act of 1991

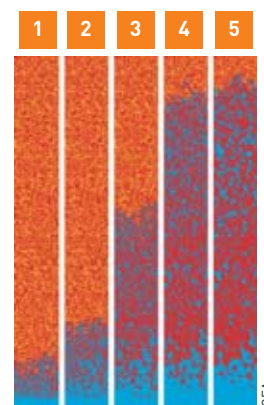
The framework laid down by the French Act of 1991 (see Box 2 in *Radioactive waste management research: an ongoing process of advances*) provided an opportunity for a comprehensive survey, and synthesis, of available knowledge on long-term package behavior, and to complement it, to provide operational models. The latter are simple, robust, conservative tools, underlying the calculations of package “source terms,” i.e. the release of **radionuclides** by the package, in the various storage or disposal situations anticipated. For each package type, these findings were gathered in a synthesis dossier.⁽¹⁾ This collates, on the one hand, the Phenomenological Reference Dossier (*Dossier de référence phénoménologique*), a set of reference data describing the state of scientific knowledge as to the long-term behavior of the package subject to investigation, setting out, in particular, the basis for the operational model; and, on the other, the Operational Dossier (*Dossier opérationnel*), providing a presentation of, and justification for, the operational model for the package being investigated, in the various generic conditions the packages may encounter in the course of their future evolution.

Simple, robust, conservative operational models

The operational models make no claim to predict the *precise* behavior of packages, over extremely long periods, which may extend to one million years, since this involves making extrapolations over timespans incommensurably greater than those accessible to experiment. They neither reproduce, nor predict, the full complexity of the processes involved, rather they aim to provide an upper-bound estimate for the source terms. Thus, they allow for larger amounts of radionuclides released, or shorter lifespans for confinement **matrices**. This approach was systematically used for all package types.

ILW-LL waste packages

For *cemented packages*, in a closed or water-unsaturated open system, the main risk that must be taken on board is that of concrete fissuring, as a result of its physical–chemical evolution. This risk may be reduced by means of an appropriate concrete **formulation**, and suitable fiber or reinforcement material. In a water-saturated open system, the major process affecting cement material behavior is chemical degradation under water (**hydrolysis**–decalcification), this being highly dependent on sulfate (SO_4^{2-}) and carbonate (CO_3^{2-}) **ion** content. Various models have been developed, to reflect the coupling between material che-



Monte-Carlo modeling of the alteration of a simplified nuclear glass, as a function of time.

- 1: $t = 25,000$ s;
- 2: $t = 50,000$ s;
- 3: $t = 125,000$ s;
- 4: $t = 250,000$ s;
- 5: $t = 450,000$ s.

■ (1) http://www.cea.fr/fr/sciences/dechets_radioactifs/



Understanding the past to predict the future

2



A volcanic glass (left) may be seen as a natural analog of nuclear glass of the R7T7 type (right).



The alteration product of basaltic glasses, palagonite, is an analog of the alteration gel from borosilicate nuclear glasses.

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Investigations carried out on natural analogs contribute to the validation of **models** of long-term evolution. Indeed, in-depth examination of such analogs allows their history to be pieced together. The scientist then knows the outcome of the material's evolution over very many years (from a few thousand to several million years), and may thus verify how consistent that evolution is with the body of knowledge gained in the laboratory.

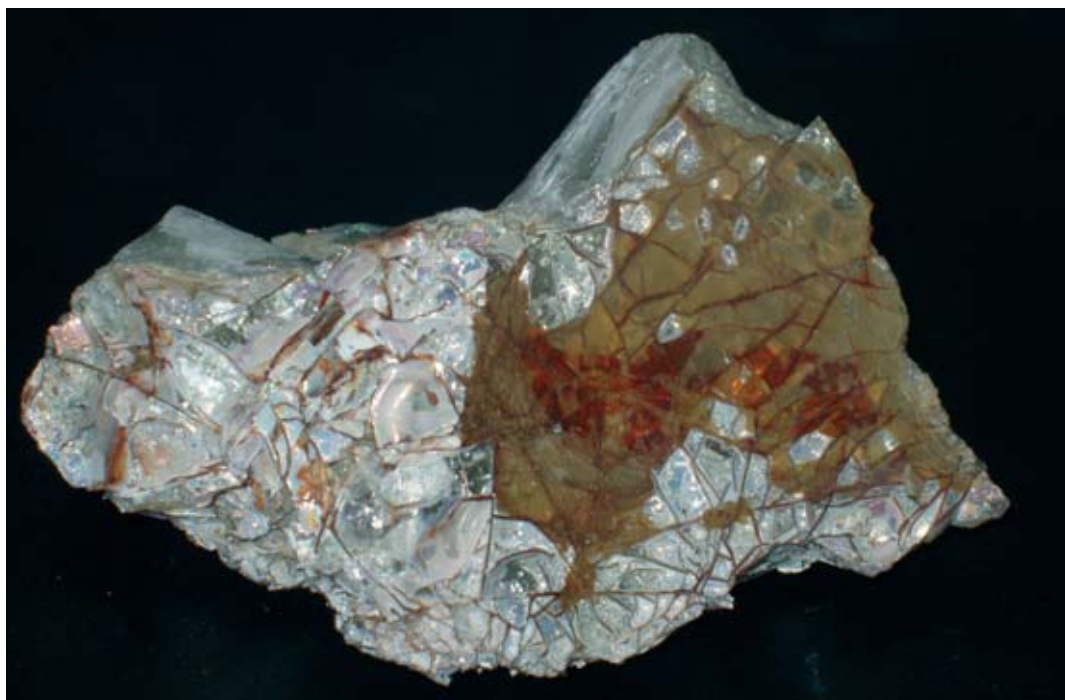
For instance, with respect to **vitrified waste**, analogs may consist in volcanic glasses (together with their alteration product, known as *palagonite*, this being an analog of the alteration gel), or archeological glasses. In particular, thoroughgoing investigation of the alteration of basaltic glasses, exhibiting characteristics very close to those of borosilicate nuclear glasses, allowed verifying that the alteration kinetics for basaltic glasses was exactly similar to that of nuclear glasses (with successive initial rate, diminishing rate, and finally residual rate regimes). Further, apparent average alteration rates for basaltic glasses, over several million years – arrived at by dividing altered depth (measured under a microscope) by sample age, obtained from the geological data – are close to the residual alteration rate measured in the laboratory.

Keeping to the topic of analogs, the investigation of archeological glass blocks that had remained for nearly 2,000 years at the bottom of the Mediterranean brings information as to the influence of glass fracturing: not only does the very existence of such blocks demonstrate that a fractured industrial glass block is able to withstand, over a timespan of millennia, highly aggressive conditions (seawater being constantly renewed), but equally that fissures occurring in blocks may be filled in over time.

mical degradation, radionuclide migration within the material, and package mechanical strength. These models allow, in particular, predictions of changes in radioactive element confinement, in the event of the external alteration of a concrete container due to water. As regards *bituminized sludge packages*, two major processes may significantly impact their long-term evolution: matrix swelling, and radionuclide release due

to alteration by water (in a water-saturated open system). The former process is caused by generation of **radiolysis** gases, this becoming negligible after 1,000 years, according to findings from the JACOB-2 operational model. The second process in turn is the outcome of the coupling of various processes, both chemical, such as water absorption by soluble salts, and hydrodynamic, such as water diffusion into the bitu-

An archeological analog, which remained close to 2,000 years at the bottom of the Mediterranean. Its examination yields evidence as to the influence of glass fracturing (see Box 2).



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men matrix. The COLONBO operational model is applicable, for conditions representative of deep disposal, to the packages, both produced and to be produced, from the La Hague plants, and to a major part of the packages fabricated by the effluent treatment stations at Marcoule and Saclay. The model predicts a release ratio of 90% of the initial radionuclide stock, after 2 million years.

Finally, the operational model developed for *standard compacted waste packages* is based on the location of the radiological inventory in the various materials in the package (fines and deposits, zirconia, Zircaloy®, stainless-steel and Inconel® structural materials), to which a certain confinement performance is associated, except for fines and deposits, and for zirconia, whose radiological inventory is deemed labile, in an upper-bound approach.

Vitrified waste packages

For **vitrified** waste, the evolution in a closed or water-unsaturated open system, under the influence of **self-irradiation** or thermics, induces no significant consequences, in terms of radionuclide release over the long term. The main processes that may significantly impact long-term behavior are thus solely to be found in a water-saturated open system. In such conditions, various regimes may be observed (see Figure 1).

The interdiffusion regime. Interdiffusion consists in an exchange between the glass's alkaline constituents and water hydronium (H_3O^+) ions. It is dominant at the onset of alteration, soon being overshadowed, however, by the hydrolysis of the glass network (after a few hours to several days).

The initial rate regime. Governed as it is by hydrolysis of the glass network (breaking of Si–O–Si chemical bonds), this regime corresponds to the maximum rate that may be observed. The value for this rate only depends, in the first order, on glass composition, **pH**, and temperature. This regime is sustained as long as no protective gel has formed.

The diminishing rate regime. *In situ* reprecipitation of certain hydrolyzed species leads to local transformation of the glass into an **amorphous** material, known as a **gel**. This process is systematically observed in the alteration of nuclear glasses. With silicon as its majority constituent, the gel forms a diffusive barrier between the bulk glass and the solution, thus considerably slowing down the rate of alteration. Gel evolution over time has been investigated in the laboratory, showing that, in a confined system such as a disposal facility, it is possible not only to guarantee gel durability but also to predict an enhancement of its protective ability over time. Findings from modeling have shown that, in disposal conditions, the diminishing rate regime persists for a relatively short time (a few years at most), and that the amount of glass altered in this period may be deemed negligible.

The residual rate regime. Rates measured in such conditions are very small, close to experimental uncertainties, of the order of $6 \cdot 10^{-6} \text{ g/m}^2 \cdot \text{d}$ at 25°C for **R7T7 glass**, meaning it will take 1,000 years for the glass to be altered to a depth of one **micron**. This regime is governed both by diffusion processes in the alteration layer, and by the very slow **precipitation** of secondary phases. The residual rate regime is the expected regime for the long term, in disposal conditions.

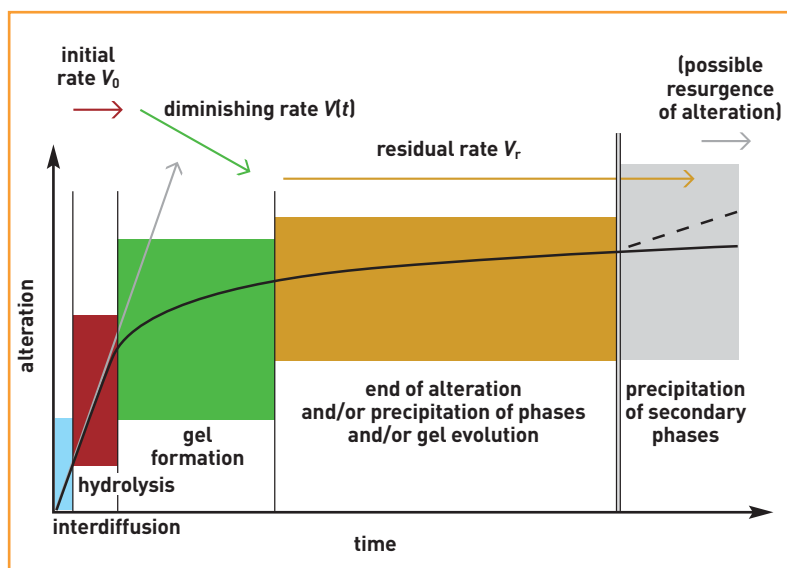
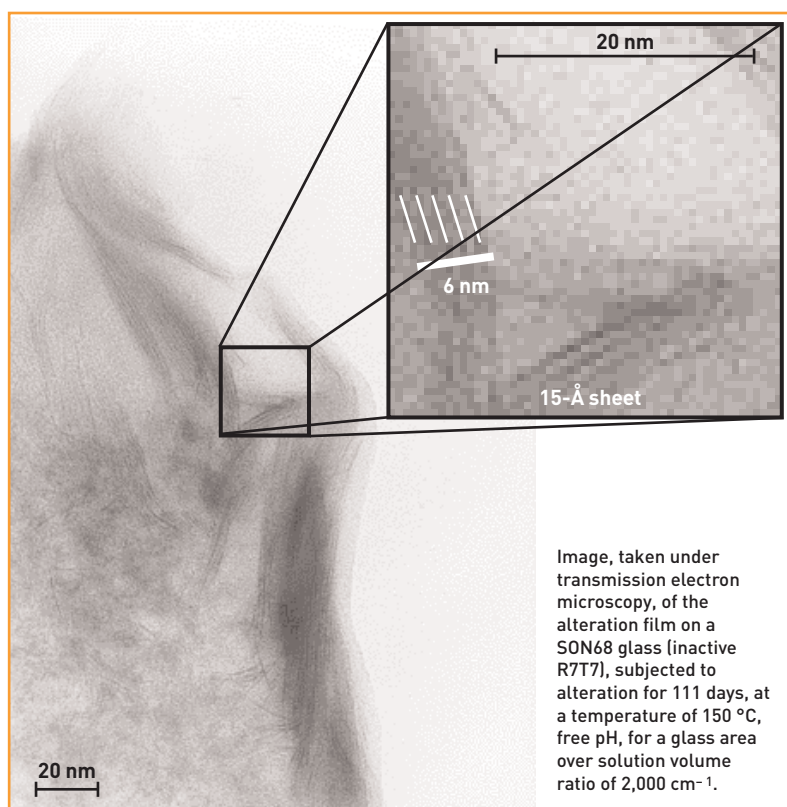


Figure 1.
Diagram showing the various regimes for the alteration rate of nuclear glasses by water.

The alteration resurgence regime. This is observed only in very special conditions, solely in alkaline conditions (for a pH higher than 10). This is due to precipitation of fast-kinetics secondary phases. In this case, the rate may be close to the initial rate.

Aside from knowledge of glass alteration kinetics as a function of time, and of alteration conditions, determining the source term for glass requires taking on board a number of considerations. First, radionuclide retention within the gel and alteration products, through **adsorption** or precipitation, is a major process in nuclear glass alteration. The extent of such retention depends both on alteration conditions, and the nature of the radionuclides involved. Indeed, it



Image, taken under transmission electron microscopy, of the alteration film on a SON68 glass (inactive R7T7), subjected to alteration for 111 days, at a temperature of 150°C , free pH, for a glass area over solution volume ratio of $2,000 \text{ cm}^{-1}$.

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is very high for **actinides** and **rare earth** elements, lower for **fission products** such as cesium. Second, accessibility by water in fissures or fractures in the glass block plays a prevailing part in controlling the rate of alteration. This characteristic leads to defining the concept of “effective surface area,” making it possible to arrive, from the alteration rate, at the quantity of glass altered, on the scale of an industrial block. Finally, the presence of environmental materials that react with glass has a significant impact on the rate regime. Thus, rapid **sorption** of the gel’s constituent elements on some clays, or on corrosion products from containers, may sustain relatively high alteration rates, close to the initial rate, so long as their reactive capacity is not saturated. Once saturation occurs, the rate falls off to the level of the residual rate.

Two operational models have been developed, drawing on these findings. The V_0S model is the simpler of the two. Its main assumption is that glass constantly alters at the initial rate – a maximum rate that can be measured – allowing for pH and temperature

conditions. This introduces, as a result, considerable margins. The $V_0 \rightarrow V_r$ operational model is more realistic, allowing as it does for the various regimes of glass alteration rate. Alteration rate is assumed to be equal to the initial rate, as long as the ambient medium is reactive, swiftly falling off thereafter to the level of the residual rate. Computations carried out with this operational model lead to lifetimes, for R7T7 vitrified waste packages, that may exceed several hundred thousand years, depending on alteration conditions. Nevertheless, this model is also quite conservative, since it does not integrate the processes of radionuclide retention within alteration products. The margin thus induced, in terms of **potential radiotoxicity**, has been evaluated at over one order of magnitude (see Figure 2).

Going beyond the various experimental findings available, investigations of natural analogs of glass (basaltic, or archeological glasses), or of gel (palagonite) further contribute to the validation of models for the very long term (see Box 2).

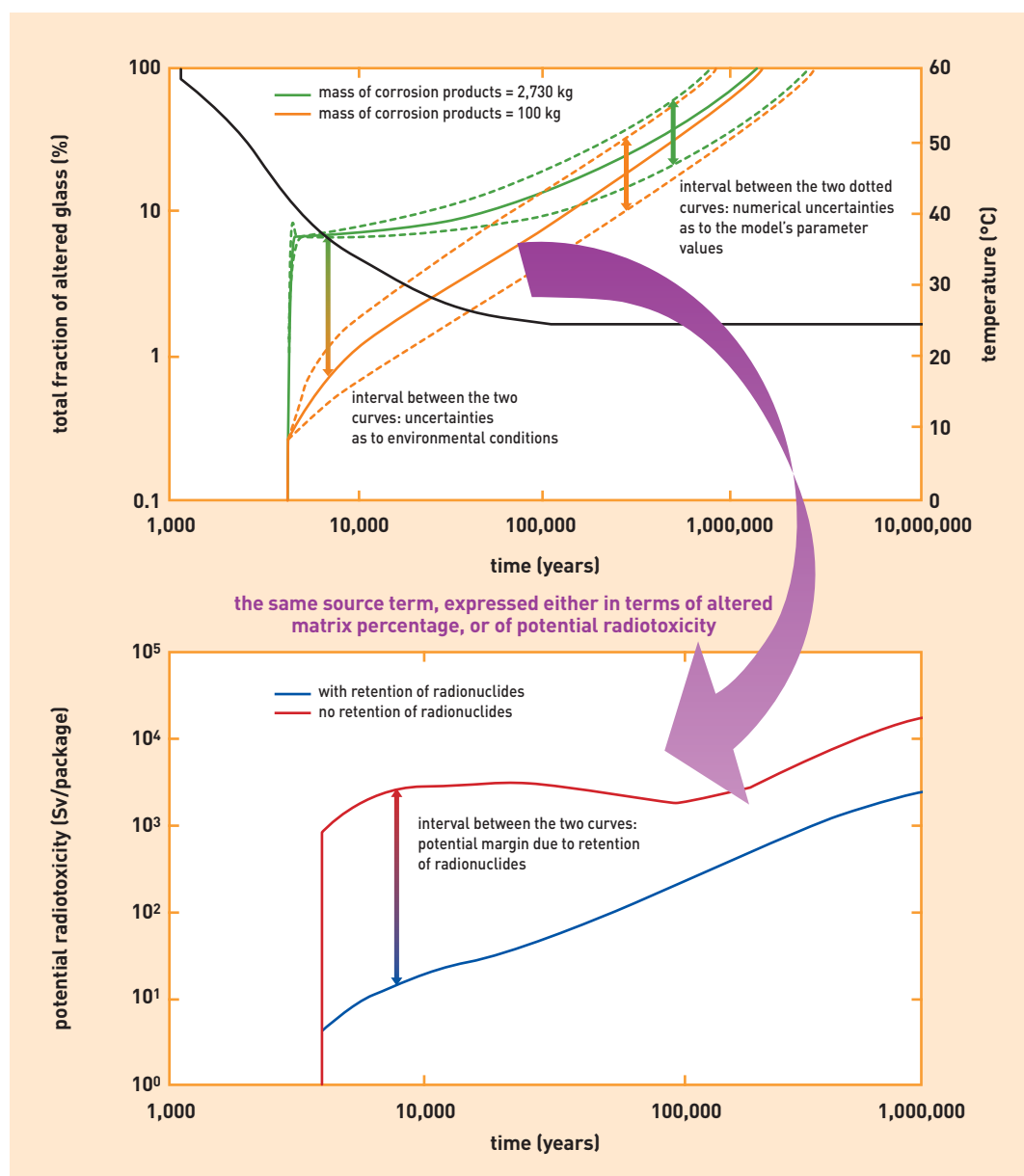


Figure 2.
Computation of the
source term for glass, by
means of the $V_0 \rightarrow V_r$
operational model,
for the case of
an R7T7-type glass.

A What is radioactive waste?

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

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

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

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

- **very-low-level waste (VLLW)**; this contains very small amounts of radionuclides, of the order of 10–100 Bq/g (**becquerels** per gram), which precludes considering it as conventional waste;
- **short-lived low and intermediate level waste (LILW-SL)**; radioactivity levels for such waste lie as a rule in a range from

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

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

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

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

* renamed Areva NC in 2006

Table 1.

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

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

What forms does it come in?

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

- **cementitious waste packages:** ILW-LL waste packages employing hydraulic-binder based materials as a conditioning matrix, or as an immobilizing grout, or yet as a container constituent;
- **bituminized sludge packages:** LLW and ILW-LL waste packages, in which bitumen is used as confinement matrix for low- and intermediate-level residues from treatment of a variety of liquid effluents (fuel processing, research centers, etc.);
- **standard compacted waste packages** (CSD-C: *colis standard de déchets compactés*): ILW-LL packages obtained through compaction conditioning of structural waste from fuel assemblies, and technological waste from the La Hague workshops;
- **standard vitrified waste packages** (CSD-V: *colis standard de déchets vitrifiés*):

HLW packages, obtained mainly through vitrification of highly active solutions from spent fuel reprocessing;

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

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

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

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

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

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

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

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

Table 2.

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

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

B Waste from the nuclear power cycle

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

The fuel's composition alters, during its irradiation inside the reactor. Shortly after discharge, fuel elements contain, on average,⁽¹⁾ some 95% residual uranium, 1% plutonium and other **transuranic elements** – up to 0.1% – and 4% of products yielded by **fission**. The latter exhibit very significant radioactivity levels – to the extent this necessitates management safety measures requiring major industrial resources – of some 10¹⁷ **Bq** per tonne of initial uranium (tiU) (see Figure 1). The **uranium** found in spent fuel exhibits a makeup that is obviously different from that of the initial fuel. The greater the irradiation, the higher the consumption of **fissile** nuclei, and consequently the greater the extent by which the **uranium** will have been **depleted** of the fissile **isotope** ²³⁵U. Irradiation conditions usually prevailing in reactors in the French fleet, with an average fuel residence time inside the reactor of some 4 years, for a

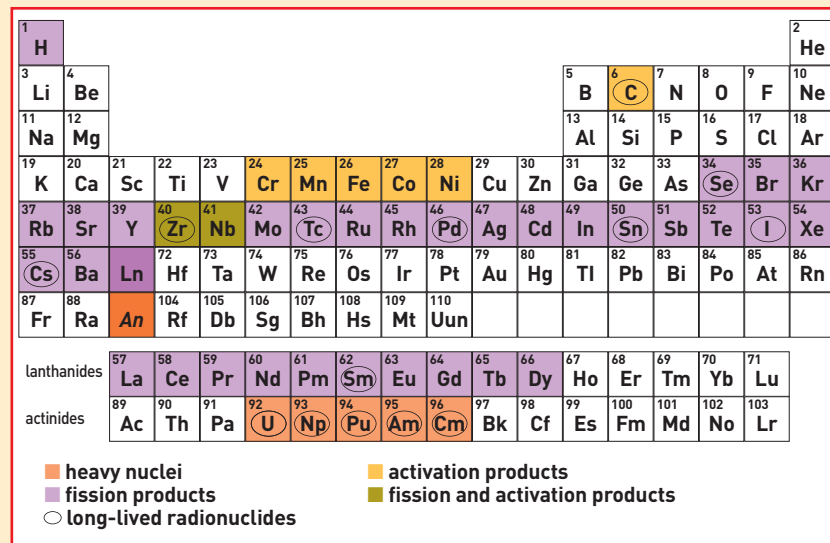


Figure 1.
The main elements found in spent nuclear fuel.

burnup rate close to 50 **GWd/t**, result in bringing down final ²³⁵U content to a value quite close to that of **natural uranium** (less than 1%), entailing an energy potential very close to the latter's. Indeed, even though this uranium remains slightly richer in the fissile isotope than natural uranium, for which ²³⁵U content stands at 0.7%, the presence should also be noted, in smaller, though significant, amounts, of other isotopes having adverse effects in neutronic or radiological terms (²³²U, ²³⁶U), that had not figured in the initial fuel (see Table 1).

The **plutonium** present in spent fuel is yielded by successive **neutron capture** and **decay** processes. Part of the Pu is dissipated through fission: thus about one third of the energy generated is yielded by "in situ recycling" of this element. These processes further bring about the formation of **heavy nuclei**, involving, whether directly themselves, or through their daughter products, long **radioactive half-lives**. These are the elements of the **actinide** family, this including, essentially, plutonium (from ²³⁸Pu to ²⁴²Pu, the odd-numbered isotopes generated in part undergoing fission themselves during irradiation), but equally neptunium (Np), americium (Am), and curium (Cm), known as **minor actinides (MAs)**, owing to the

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

element	isotope	half-life (years)	UOX 33 GWd/tiU (E ²³⁵ U: 3.5%)		UOX 45 GWd/tiU (E ²³⁵ U: 3.7%)		UOX 60 GWd/tiU (E ²³⁵ U: 4.5%)		MOX 45 GWd/tihm (Ei Pu: 8.65%)	
			isotope content (%)	quantity (g/tiU)	isotope content (%)	quantity (g/tiU)	isotope content (%)	quantity (g/tiU)	isotope content (%)	quantity (g/tihm)
U	234	246,000	0.02	222	0.02	206	0.02	229	0.02	112
	235	7.04·10 ⁸	1.05	10,300	0.74	6,870	0.62	5,870	0.13	1,070
	236	2.34·10 ⁷	0.43	4,224	0.54	4,950	0.66	6,240	0.05	255
	238	4.47·10 ⁹	98.4	941,000	98.7	929,000	98.7	911,000	99.8	886,000
Pu	238	87.7	1.8	166	2.9	334	4.5	590	3.9	2,390
	239	24,100	58.3	5,680	52.1	5,900	48.9	6,360	37.7	23,100
	240	6,560	22.7	2,214	24.3	2,760	24.5	3,180	32	19,600
	241	14.4	12.2	1,187	12.9	1,460	12.6	1,640	14.5	8,920
	242	3.75·10 ⁵	5.0	490	7.8	884	9.5	1,230	11.9	7,300

Table 1.
Major actinide inventory for spent UOX and MOX fuel after 3 years' cooling, for a variety of enrichment and burnup rates. Burnup rate and quantity are expressed per tonne of initial uranium (tiU) for UOX, per tonne of initial heavy metal (tihm) for MOX.

B (next)

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

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

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



Magnum/Harry Gruyaert

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

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

Table 2.
Breakdown by chemical family of fission products in spent UOX and MOX fuel, after 3 years' cooling, for a variety of enrichment and burnup rates.

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

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

Such U and Pu recycling is the foundation for the **reprocessing** strategy currently implemented in France, for the major part of spent fuel (some two thirds currently).

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

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

c What stands between waste and the environment?

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



A. Gomin/CEA

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

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

When in the state they come in as supplied by industrial production, they are known as **primary packages**, the pri-

mary container being the cement or metal container into which the conditioned waste is ultimately placed, to allow handling. The container may act as initial confinement **barrier**, allotment of functions between matrix and container being determined according to the nature of the waste involved. Thus, the whole obtained by the grouping together, within one container, of a number of primary

c (next)

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

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

In storage or disposal conditions, the waste packages will be subjected to a variety of aggressive agents, both internal and external. First, radionuclide

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

Stored waste packages will also be subjected to the effects of water (**leaching**). Container canisters may exhibit a degree of resistance to corrosion processes (the **overpacks** contemplated for glasses may thus delay by some 4,000 years the arrival of water), and the confinement matrices must be proven to exhibit high chemical stability.

Between the containers and the ultimate barrier provided, in a radioactive waste deep disposal facility, by the geological environment itself, there may further be interposed, apart, possibly, from an overpack, other barriers, so-called **engineered barriers**, for backfill and sealing purposes. While these would be pointless as backfill in clay formations, they would have the capability, in other environments (granite), of further retarding any flow of radionuclides to the **geosphere**, notwithstanding degradation of the previously mentioned barriers.



Technological demonstrators of ILW-LL packages for bituminized sludges.

D From storage to disposal

The object of nuclear waste **storage** and **disposal** is to ensure the long-term **confinement** of **radioactivity**, in other words to contain **radionuclides** within a definite space, segregated from humankind and the environment, as long as required, so that the possible return to the **biosphere** of minute amounts of radionuclides can have no unacceptable health or environmental impact.

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

Used from the outset of the nuclear power age, industrial storage keeps spent fuel awaiting reprocessing, and conditioned **high-level waste (HLW)**, or **long-lived intermediate-level waste**

(**ILW-LL**) in conditions of safety, pending a long-term management mode for such waste. Retrieval of stored packages is anticipated, after a period of limited duration (i.e. after a matter of



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

years, or tens of years).

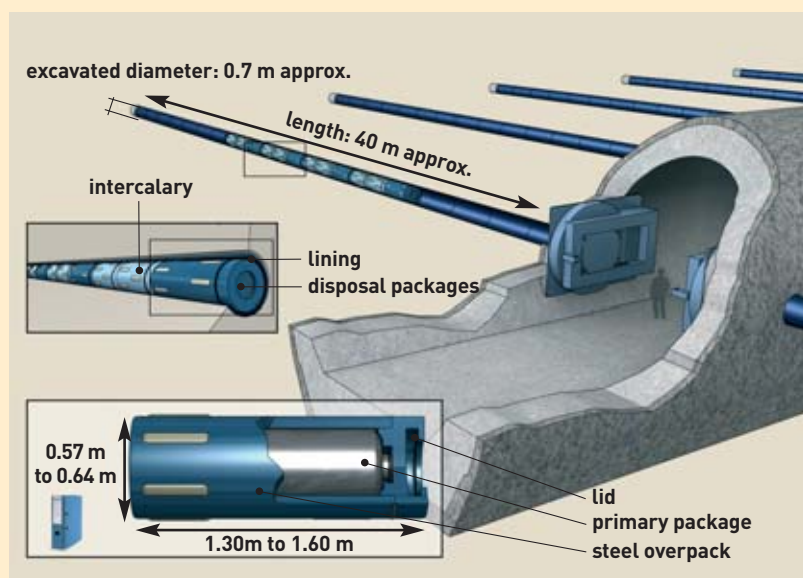
Long-term storage (LTS) may be contemplated, in particular, in the event of the deferred deployment of a disposal facility, or of reactors to carry out

recycling-transmutation, or simply to turn to advantage the natural decay of radioactivity (and hence the falling off of heat release from high-level waste), before putting the waste into **geological disposal**. By "long term" is meant a timespan of up to 300 years. Long-term storage may take place in a **surface** or **sub-surface** facility. In the former case, the site may be protected, for instance, by a reinforced-concrete structure. In the latter case, it will be located at a depth of some tens of meters, and protected by a natural environment (for instance, if buried in a hill-side) and its host rock.

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

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

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



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

E What is transmutation?

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

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

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

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

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

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

- by expelling particles (a neutron, e.g.), while possibly releasing radiation;
- by solely emitting radiation; this is known as a *capture reaction*, since the neutron remains captive inside the nucleus;
- by breaking up into two nuclei, of more or less equal size, while releasing concurrently two or three neutrons; this is known as a *fission reaction*, in which considerable amounts of energy are released.

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

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

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

E (next)

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

For a neutron having an energy lower than 1 **eV** (in the range of slow, or thermal, neutrons), the capture cross-section

prevails; capture is about 100 times more probable than fission. This remains the case for energies in the 1 eV–1 **MeV** range (i.e., that of **epithermal neutrons**, where captures or fissions occur at definite energy levels). Beyond 1 MeV (fast neutron range), fissions become more probable than captures.

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

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

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

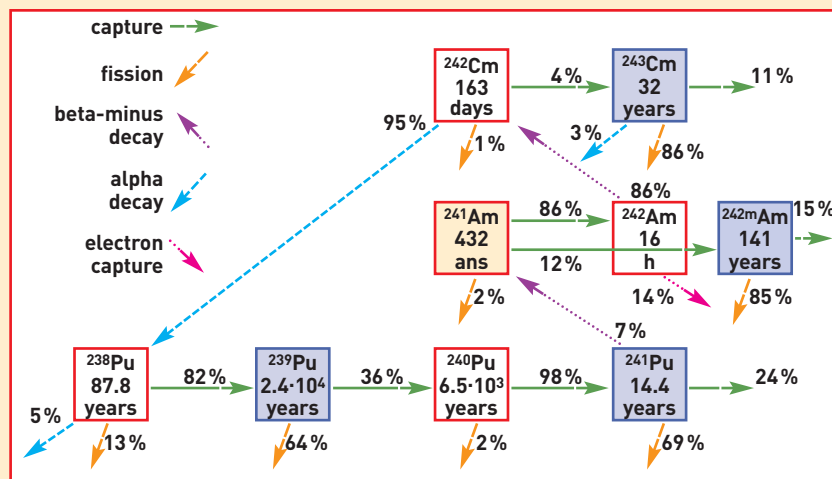


Figure.

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

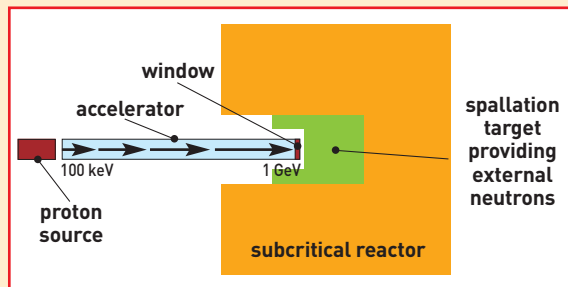
F What is an ADS?

An **ADS** (accelerator-driven system) is a **hybrid system**, comprising a nuclear reactor operating in **subcritical** mode, i.e. a reactor unable by itself to sustain a **fission chain reaction**, “driven” by an external source, having the ability to supply it with the required complement of **neutrons**.⁽¹⁾

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

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

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



Principle schematic of an ADS.

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

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

ter then go on to interact with the fuel of the subcritical neutron **multiplier medium**, yielding further neutrons (fission neutrons) [see Figure].

Most hybrid system projects use as a core (of annular configuration, as a rule) **fast-neutron** environments, since these make it possible to achieve neutron balances most favorable to **transmutation**, an operation that allows waste to be “burned,” but which may equally be used to yield further **fissile** nuclei. Such a system may also be used for energy generation, even though part of this energy must be set aside to power the proton accelerator, a part that is all the higher, the more

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

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

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

The industrial context

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.