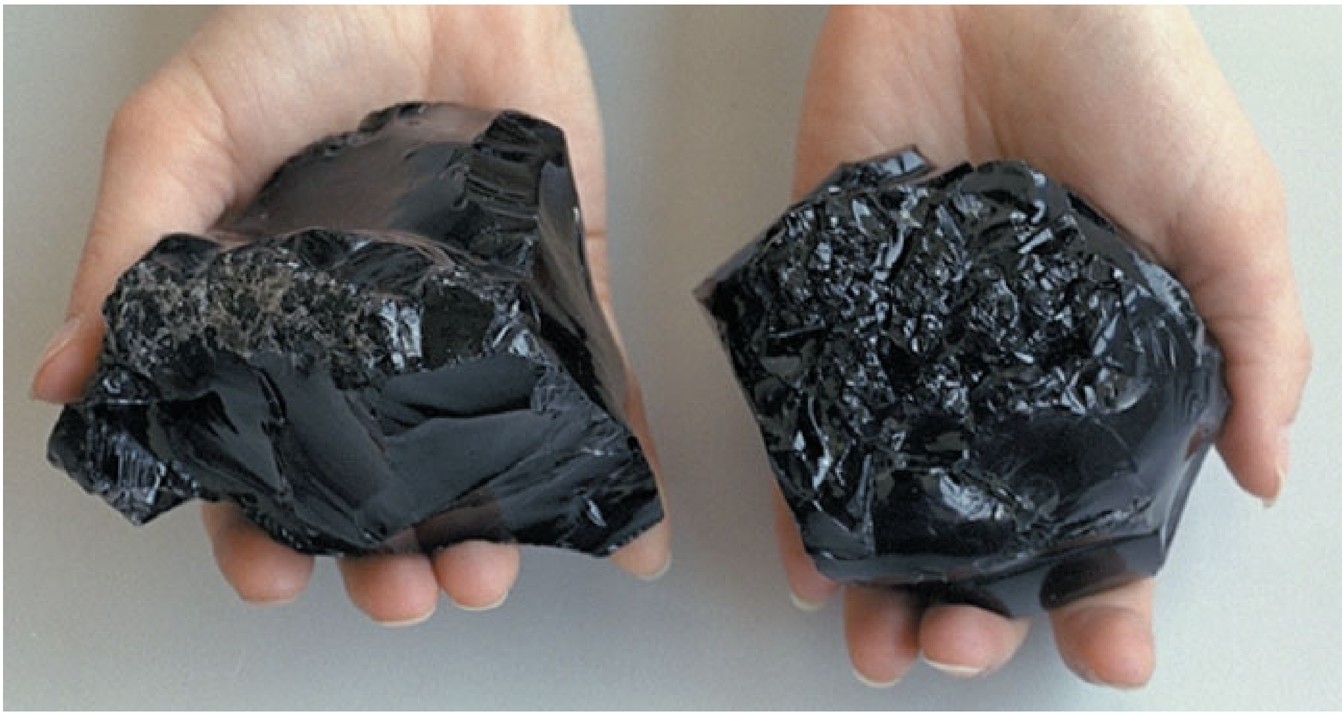
PREDICTING THE LONG-TERM BEHAVIOR OF WASTE PACKAGES

Scientists have undertaken to accurately establish the main physical and chemical mechanisms that govern the long-term behavior of radioactive waste packages stored or disposed of in a given environment, since they are unable to experiment directly on them. They have integrated these mechanism into models to make projections over time and arrive at representative orders of magnitude for the elements stored or disposed of, for all categories - spent fuel stored as is, vitrified-waste packages and radioelements incorporated into other matrices, such as ceramic matrices. The validity of extrapolations provided by these models is subject to in-depth analyses. While they cannot guarantee to predict the fate of a specific waste package exactly, they can be used to forecast the consequences of what might happen to it. The conclusion of this work is that any release of a minimal quantity of radionuclides would not occur before tens of thousands of years have elapsed.



Thierry Foulon/CEA

Volcanic glass, such as the chunk shown on the left, can be considered as a natural analogue of type R7T7 nuclear waste glass (on the right).

While the initial purpose of conditioning waste in the form of **packages** is to make handling possible, the main purpose is to guarantee powerful and durable **containment** of the **radionuclides** it contains (see box G, ***Conditioning, a vital phase***). One peculiarity of **radioactive** waste is that with the passage of time it loses its harmfulness, but this decrease can be very slow. Therefore the packages must remain isolated from the **biosphere** for very long periods.

CEA is conducting a research program into the long-term behavior of waste packages so that it can guarantee the performance of conditioning for **long-lived** waste and enlighten future decisions on its management. This program seeks to establish how waste packages evolve over time, focusing on their behavior in reaction to the various internal or external aggressions to which they will be subjected in **disposal** or **storage** conditions. Advances in simulation techniques and familiarity with the effects of these aggressions in the long term are making this work possible. It is being carried out in conjunction with the industrial producers of the waste, and allows to organize results in a shared knowledge base. This approach, which is founded on a structured scientific approach that calls on many specialties, makes it possible to assess, then share, the degree of confidence with which these forecasts can be viewed. Finally it results in developing behavior **models** ("ope-

Core of zone 10 of the Oklo natural reactor, in Gabon. Research on it allows to assess the future of radionuclides over very long periods in an environment that presents similarities to geological disposal.

Hosatte/CEA

rational" models) simple enough to be incorporated in the general management-method assessment procedure. By 2005 a standard database on the long-term behavior of waste packages will be available for reference purposes.

Determining the mechanisms and extrapolating over time

The research programs looking into the long-term behavior of radioactive-waste packages are special in that they must enable extrapolations to be made over time and make pronouncements on the lifespan of the functions assigned to the packages in the main contexts planned for (storage, deep geological disposal). That is why research workers are seeking to establish the main physical and chemical mechanisms that govern package behavior and then build them into the models which will be used to project over time.

In the first stage research workers set out to understand these phenomena, either by conducting laboratory experiments or observing comparable natural phenomena (volcanic glass, elements found in the Oklo natural reactors in Gabon, etc.). The main phenomena studied are those that involve water (in liquid or vapor form), heat and the radiation of the packages themselves. Then they develop mathematical models that will enable them to predict the long-term behavior of the package, and then validate them by means of these natural analogues or calling on outside scientific expertise.

Sampling a solution during an oven leaching test at 50 °C as part of the research conducted at CEA/Marcoule on the long-term behavior of waste-package matrices.

Thierry Foulon/CEA

Three main types of environmental conditions

Long-term behavior of waste packages is routinely focused on three major types of environmental conditions: closed system, open system non-saturated with water, and open water-saturated system.

Closed-system evolution

First of all study turns to the closed system evolution of the **packages**, in which the package casing is intact and containment is totally ensured. The package undergoes internal transformation, but does not exchange any substances with its environment. It can thus undergo chemical or **radiolytic** reactions that may trigger internal corrosion and swelling through the effect of gas. This evolution may put the casings of these **containers** under stress, thus jeopardizing future handling of the packages and the containment of the radioelements. Management of these phenomena is now well under control for many industrial packages as scientists can simulate them and thus estimate orders of magnitude for the relevant protection time scales.

Evolution in a non-saturated open system

The next stage is to assess the effects of an aggression to the **matrix** from the outside environment providing that it is not saturated with water. The stressors are air of varying degrees of dampness, and any corrosive gases that form by radiolysis of air when it comes into contact with the packages. Specific packages or matrices, which are sensitive to these stressors, may transform then lose all or some of their containment properties. Uranium oxide is a particular case in point. This phase occurs in scenarios where abnormal or degraded situations are studied to see how packages will alter in storage or during the presealing phase of a deep disposal (see box I, ***How can disposal be reversible?***).

Evolution in a water-saturated environment

Lastly attention turns to the evolution of packages in a water-saturated environment, which could be the conditions prevailing in a deep disposal facility, several thousands of years after a site has been sealed off, once the water pressures have readjusted in the geological formation. It is during this phase that corrosion and aqueous transitions in the containers and matrices will develop. Then once the containment **barriers** have fallen, the **radionuclides** will be able to migrate. Confidence is highest in this area of scien-

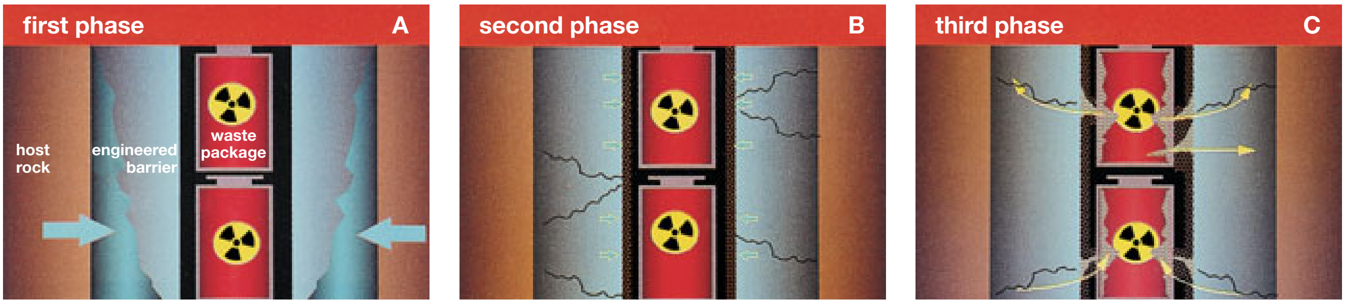


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Figure. The three phases, schematized, of late evolution of waste package near field in a disposal facility in geological formation: waterlogging (A), physical and chemical evolution of the engineered barrier and corrosion (B), package alteration and radioelements migrating (C).

tific knowledge, as research workers can quite accurately assess the modes and kinetics of corrosion and alteration states of the container and matrix materials in the well-defined chemical conditions of deep disposal (figure).

Near field and far field

In the case of deep disposal, waste packages will be surrounded by so-called **engineered barriers** that provide additional containment (see *Potential storage and/or disposal strategies*). Whatever type of infill material is placed between the packages and the geological barrier makes up the *near field*; the *far field* is the geological environment undisturbed by the underground engineering structure.

Scientists have been forced to take into account coupled interface phenomena between the packages and both near and far environment when simulating the long-term evolution of the waste packages in deep disposal. The far field, which is hydrologically, mechanically and geochemically stable, imposes its conditions at the boundaries of the near field, which will gradually react to readjust to it.

From hot and dry to lukewarm and damp

To put it simply, the near field of high-level waste is initially hot and dry in the center, and then it will cool down as the heat dissipated by the waste decreases over time. Parallel to that the water will gradually invade the near field from the outside. It is important to simulate waste-package far-field reactions over time. While temperatures do not exceed 100–150 °C, it has been shown that the engineered barriers will not have lost their swelling, waterproofing and radioelement-fixing properties.

For ten years and more all these near field package transition phenomena have been the subject of research that is now highly advanced. Researchers are able to simulate these transitions over the very long term and have come to the conclusion that, in every single case, any release of the radioelements outside the near field will not occur until after several tens of thousands of years have elapsed. Furthermore this release would only involve a minute quantity of radioelements, as most of them would have lost all their radioactivity while still contained inside the packages.

The main categories of packages investigated

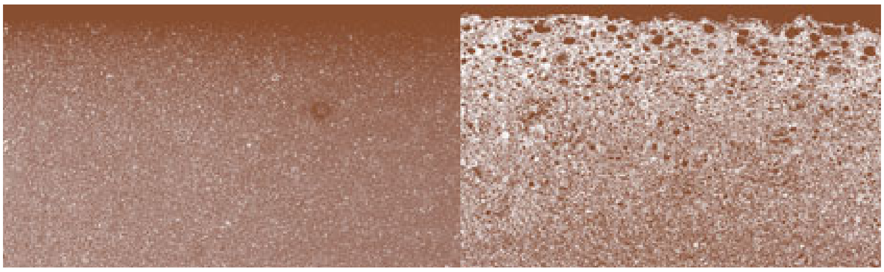
The whole process is repeated for each of the five main waste package categories: vitrified waste packages, particularly of the R7T7 type, compacted waste packages, bituminized(1) waste packages, hydraulic binder-based waste packages (cement, mortar, concrete) and spent-**fuel** packages. The latter include packages that already exist, such as the standard vitrified waste package (produced by Cogema at La Hague), those in the final stages of development (such as the standard compacted waste package also designed by Cogema), or still at design stage (the spent-fuel waste package).

We illustrate this approach by taking the example of vitrified-waste and spent-fuel packages, which differ both in nature and in terms of our knowledge about them, in situations that are specific to them.

A protective layer on glass packages

Cogema produces vitrified-waste packages at Marcoule and La Hague essentially by vitrifying highly active solutions emanating

Cross-sections, obtained using scanning transmission electron microscopy, of a bitumen coating as part of research carried out with Cogema on the long-term behavior of waste packages. The left-hand view was taken prior to leaching, the right-hand one after 20 months of leaching, revealing the formation of a permeable area.



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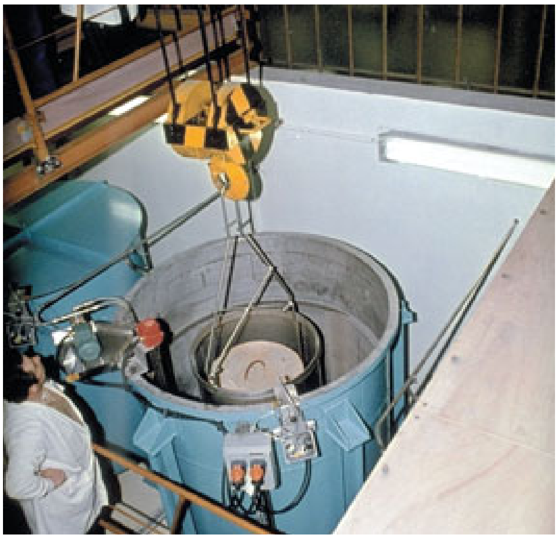
(1) Modeling has in particular allowed to understand and assess the formation of gaseous hydrogen in the liquid-waste packages encased in bitumen (category **A** or **B** waste), in the event of interaction with water, which would lead to deformation of this waste package.

Waste package leaching research installation at CEA/Valrhô-Marcoule.



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from spent fuel reprocessing (see box B, *Waste from the nuclear power cycle*). The radioelements are incorporated in the form of oxides dispersed in a vitreous lattice. Several nuclear waste glass compounds exist, but the bulk of the studies has centered on R7T7, French nuclear glass, by far the most important, (see *Waste vitrification: more than one string to its bow*). Vitrified-waste packages are characterized by a high level of activity and slow radioactive decay. Technically speaking their storage is not a major problem even over the very long term, provided the stainless-steel canisters are monitored for resistance to corrosion. The main operational issue is how long the radionuclides can be contained, in generic disposal conditions, once the site has been resaturated with water. The homogeneous and well-characterized vitreous matrix dominates the behavior of this package, which forms a simple system (see *Glass packages guaranteed for millions of years*). The durability of containment is controlled by the kinetics of glass alteration as water levels on the site are slowly renewed. Moreover this alteration induces the formation of a protective layer at the glass/water interface resulting from the recondensation of the hydrolyzed silica. This layer in turn contributes to slowing down alteration by 2–4 orders of magnitude. So the annual fraction of altered glass is only of the order of 10^{-7} in typical disposal conditions once the site has been water-saturated. Glass, specifically designed to condition high-level waste, turns out to be a very durable matrix indeed.

The most striking results on vitrified waste were achieved in 1999: a long-term evolution model showed that, in deep disposal, 99.9% of the glass remains intact, even after 10,000 years' contact with water. Thus glass turns out to be the ideal matrix because the life span of glass packages can be calculated in millions, and even tens of millions of years in optimum conditions.

The research work outstanding on long-term behavior of vitrified-waste packages will thus focus on extending alteration modeling on the basis of increasingly accurate understanding of the fundamental physical and chemical mechanisms at work. In order to anticipate the formulation evolutions expected because of increased **burnups** and the varied nature of fuel reprocessed, the scope of application will be broadened to include a greater variety of glass categories.

Mascilia experimental device at CEA/Cadarache, used for research into the long-term mechanical and hydromechanical behavior of the geomaterials confronted over time by a wide range of temperatures in high-level radioactive waste storage or disposal facilities.

Emmanuel Joly/CEA

Spent fuel as is

Most of the spent fuel emanating from French power plants, in the current fuel-cycle scheme, is **processed** to extract the materials that can be recycled. The major part of the waste is conditioned in glass packages for geological disposal. Notwithstanding research is going on into storing it over the long term (several generations) and geological disposal (see *Spent nuclear fuel as is: how to dispose of or store it safely*). This begs many operational questions such as: how far can we safely retrieve assemblies after long periods of storage? How should they then be processed? What are the properties of fuel **assemblies**, that were designed only to function in a reactor, not for the long-term conditioning of radionuclides in mind and are fairly complex systems? To find answers to these questions many mechanisms taken from the fields of the physics and chemistry of solids and gases, radiochemistry, mechanics, corrosion and so on will have to be built in. The mechanisms that underlie evolution of the fuel structure itself under the effect of gradual lowering of temperature and the production of helium resulting from **alpha radioactivity**, cladding subjected to overpressure due to the production of gas and the formation of swelling phases in the event of the fuel oxidizing, etc., will have to be incorporated. It happens to be particularly difficult to demonstrate that cladding can be resistant in the long term. That is why a “dual-casing packing system” is recommended for storing or reversible phase disposal (see *Potential storage and/or disposal strategies*).

Post-waterlogging behavior in the deep disposal situation is again dominated by the interaction of the fuel with water. The state of the fuel when this water arrives remains a major unknown. A question mark hangs over the fraction of radionuclides that can be rapidly mobilized, conditioned by the size and cohesion level of the then-aged uranium oxide grains. How radiolysis affects the disassociation kinetics of the oxide matrix that traps the remaining radionuclides is a further research topic.

All in all, research into irradiated fuel must be pursued to enhance the dependability of current models (including by the characterization of industrial fuels), and develop suitable container and store designs. ●

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