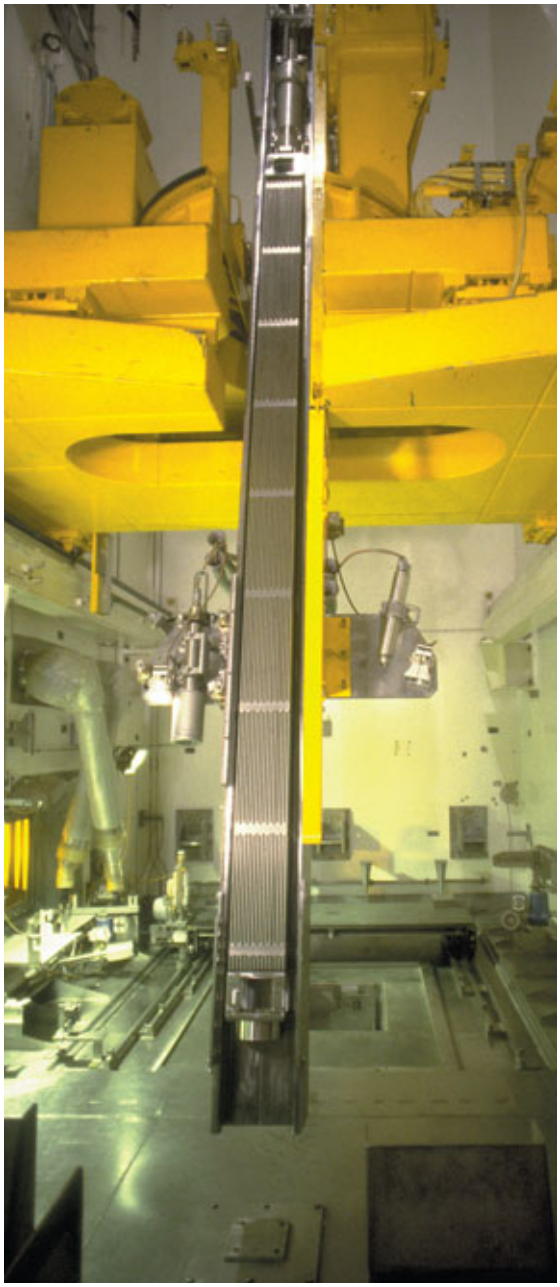
SPENT NUCLEAR FUEL AS IS: HOW TO DISPOSE OF OR STORE IT SAFELY

France has opted to process spent fuel, while other countries such as the United States – which is having second thoughts – Sweden and Finland, have decided not to do so. CEA has embarked on a far-reaching research program working in collaboration with its partners in the nuclear industry on the conditioning and long-term behavior of spent -fuel assemblies kept as is. The purpose is to be able to take the decision on the ultimate fate of these spent-fuel assemblies currently not undergoing, or destined not to undergo reprocessing in either long-term storage or deep geological disposal, with the benefit of full knowledge of the facts.

The French nuclear power industry annually discards some 1,200 metric tons of spent nuclear **fuel assemblies**. Roughly 850 tons' worth goes to La Hague for reprocessing while the remaining 350 tons are stored in EDF's power plant cooling pits pending a decision. All or part of this spent fuel (see *The 1991 Act*) will either remain in industrial **storage** with a view to being **reprocessed** at a later date, or be stored for an undefined length of time pending a final decision on its fate. In the interests of exhaustiveness and comparison with other countries, deep geological **disposal**, possibly incorporating a **reversibility** phase, is also being examined. Viewed from this angle it is vital to have forecasts of how the spent nuclear fuel stored in these sets of conditions will evolve over the long term (100–100,000 years…). Only once society has full knowledge of the facts will it be in a position to take an informed decision on the final fate of this fuel when the time comes to do so.

In 1998 this thinking prompted CEA to embark on the **Precci** (Research program into the long-term evolution of irradiated fuel packages) interdisciplinary research program working closely with EDF, the producer and owner of fuel assemblies, and other players from the industry. It coordinates all the scientific work being carried out by teams of scientists in this sphere in the Saclay, Cadarache and Marcoule centers(1). The wider scope of this program draws on a vast network of both national (CNRS, universities...) and international (Belgium, Germany, Japan, Spain, Sweden,

(1) Seven departments reporting to the Nuclear Energy Division and one department reporting to the Physical Sciences Division are taking part in this research program.



Philippe Lesage/Cogema

Spent-fuel assembly in the Cogema UP2-800 plant shearing shop at La Hague.

Switzerland, the United States…) collaborative work. Much of this is being carried out under the mantle of a European(2) research project piloted by CEA.

Identifying long-term evolution mechanisms

The prime aim of this work is to identify the various mechanisms likely to drive spent-fuel evolution in the long term. These mechanisms will then be quantified to simulate integrated models so that the evolution of the complex system that is nuclear spent fuel can be predicted (figure 1). To ensure a unified and consistent approach, all the work carried out in this field has been organized so that it considers the various potential environmental conditions for the fuel in either of the two options of storage and disposal. This has led to the identification of three evolution modes: as a closed system, or in the presence of a gaseous phase or water.

In **closed system** mode, the spent fuel evolves intrinsically without any material exchange with the outside environment. This evolution is what would normally be expected of "cask" dry storage and in the first life phases of deep geological disposal, namely before the site is water-saturated and the gradual corrosion of the container sets in, which should last several thousand years.

Evolution in the **presence of a gaseous phase**, i.e. in contact with an oxidizing gaseous atmosphere, may represent an incidental situation in dry storage during which the tightness of a container is compromised, leading to the atmosphere altering the fuel.

Evolution in the **presence of water** is when the fuel comes into contact with an aqueous phase. The fuel is slowly dissolved and gradually releases **radionuclides**. This situation is expected in the very long term in deep disposal following the end of the first "closed-system" phase and could also apply to a faulty fuel rod stored in a pool. The **Precci** program aims to develop models for each of these generic environmental conditions, and predict the evolution of the relevant physical and chemical characteristics over the long term so that safe storage or disposal facilities may be designed. On the basis of this assumption, R&D is centered on the evolution mechanisms outlined in the following paragraphs.

(2) "Spent-Fuel Stability Under Repository Conditions" project in which thirteen European bodies representing Belgium, France, Germany, Spain, Sweden and Switzerland are taking part.
(3) Fick's law.

Closed-system evolution (storage and disposal)

The fuel will alter through the combined effect of internal physical and chemical imbalances resulting from its life in the reactor (composition gradients, mechanical constraints…) and the action of **radioactive** and heat **decay**. The main evolutions expected will affect chemical composition, location of the radionuclides and the state of the rod and cladding.

General chemical composition

Radioactive decay will cause chemical composition to evolve. Likewise the chemical form of specific **radionuclides** will change through the gradual drop in temperature, taking the form of a significant amount of helium gas accumulating in the rods resulting from **alpha** (α) emitter decay in the fuel pellets (figure 2).

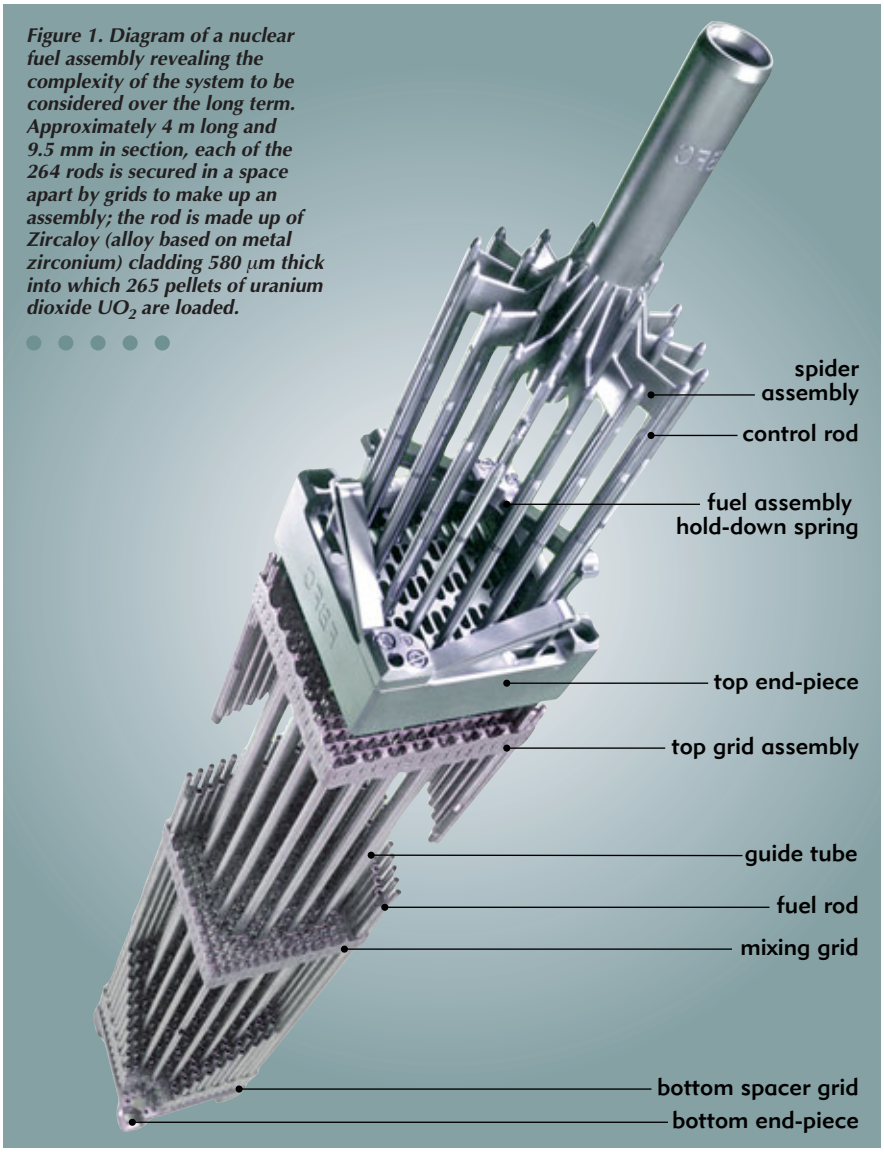
Location of radionuclides

The radionuclides in the rods will gradually change location as a result of thermal diffusion processes(3) and α **self-irradiation**-activated diffusion. Experiments are currently under way to calculate the diffusion coefficients of the main radionuclides involved.

Overall physical condition of the rod

The rod is expected to deteriorate because of a number of factors: the accumulation of large amounts of helium, radiation damage build-up over long periods that is not repaired since temperatures are too low and the gradual deterioration of contact surfaces between the material's constituent grains (grain boundaries) - a mechanical weak spot in pellets. Both experimental work and numerical simulation are used to investigate these transformations (figure 3), so the pellets could gradually lose their cohesion.

Figure 1. Diagram of a nuclear fuel assembly revealing the complexity of the system to be considered over the long term. Approximately 4 m long and 9.5 mm in section, each of the 264 rods is secured in a space apart by grids to make up an assembly; the rod is made up of Zircaloy (alloy based on metal zirconium) cladding 580 μm thick into which 265 pellets of uranium dioxide UO_2 are loaded.



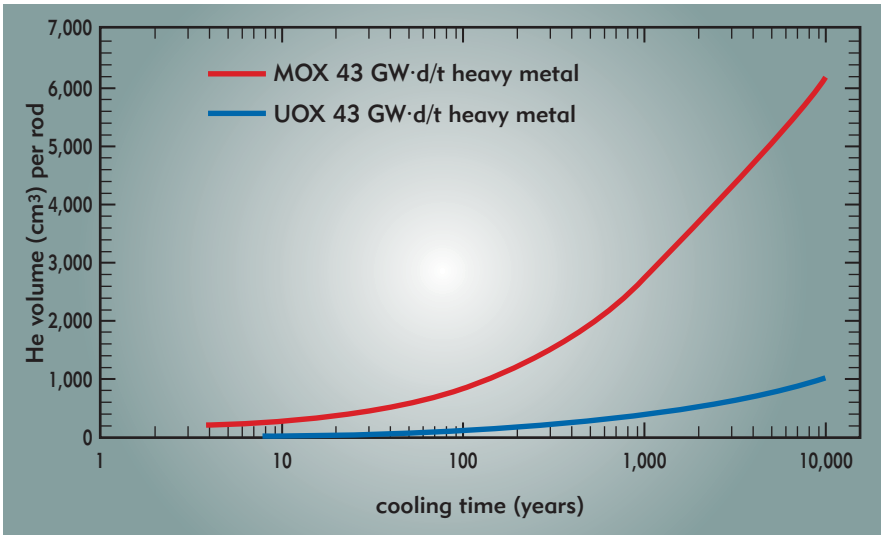


Figure 2. How the quantity of helium produced following the decay of α emitters present in the irradiated fuel pellets in a MOX (red) or UOX (blue) fuel assembly for a mean burnup fraction of 43 GW·d/t alters over time. One of the key challenges of research is to determine the future of this gas in the rods. (Piron et al.*, 2000).*

Mechanical condition of cladding

Cladding will gradually suffer irreversible deformation (so-called "**thermal creep**" mechanism) through the acute mechanical stresses due to the overpressures inside the rods (fission gases and helium) and the temperature. It is important to assess the kinetics of this deformation and gauge whether it is likely to lead to rupture.

Gaseous-phase evolution (incidental storage conditions)

Fuel pellets are made up of uranium dioxide UO_2, a material that becomes highly unstable in the presence of oxidants such as oxygen and water vapor… It tends to transform rapidly into more highly oxidated substances such as U_3O_7/U_4O_9 then U_3O_8. The latter's density is lower than the initial material, which causes the pellets to swell and burst. These transitions are relatively well known for unirradiated UO_2 and at temperatures of over 250 °C. The challenge now on is to extend this knowledge base to more complex cases of irradiated fuel and for the lower temperatures that prevail in storage and disposal situations. The radionuclides released in the event of such transformation occurring must also be identified.

Evolution in the presence of water (long-term disposal conditions)

In the presence of water, spent fuel releases part of the radionuclides not bound by the oxide (the so-called "labile" fraction), then the pellets gradually and slowly dissolve releasing the radionuclides incorporated into the oxide. The dissolution rate is largely bound to the oxidizing/reducing conditions of the environment. The fuel will be altered much faster in an oxidizing environment. Furthermore irradiated fuel is peculiar in that it presents a significant α radiation field on contact with water for several tens of thousands of years, which locally sustains these oxidizing conditions by **radiolysis**, exacerbating the fuel alteration rate. One major task ahead is to quantify this mechanism and find out how long it is likely to govern the dissolution of the fuel. Another vital question is to learn exactly what nature and quantity of radionuclides will be released in the labile fraction.

Predicting the long-term behavior of spent fuel will be kernel to deciding on its future. It calls for R&D to provide answers to very complex questions as they are closely correlated and require an understanding of the basic mechanisms at microscopic scales. By creating the necessary synergies between very diverse disciplines and teams, the Precci program could put forward relevant and sound models by 2006 that could be used for predicting the long-term evolution of spent fuel. These models will provide CEA and its industrial partners with solutions to the operational issues posed by the designers of storage and disposal facility. By taking into account the intrinsic reactivity of spent fuel they will be able to define the safety of these installations.

Christophe Poinssot
Coordinator of the Research program into the long-term evolution of irradiated fuel packages,
chargé d'affaires for Precci
Nuclear Energy Division
CEA/Saclay

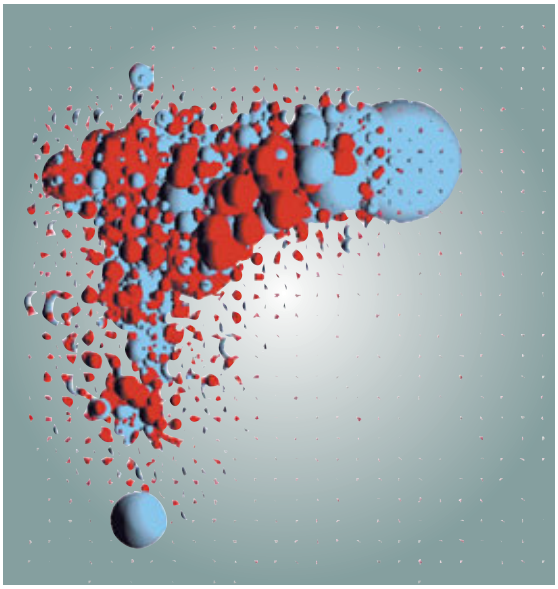


Figure 3. Result of numerical simulations carried out by molecular dynamics on UO_2 to investigate the consequences of α-emitter decay in the material, by determining the influence of atom recoil during α decay on the crystallographic state of the lattice (nature and quantity of faults formed). The figure shows the state of a simulated UO_2 crystal 300 femtoseconds (10^{-15} s) after the impact of a recoil nucleus. Each sphere (or dot) represents an atom (of oxygen in red and uranium in blue) and the size of these spheres is proportional to the speed acquired by these atoms following impact (Ghaleb et al., 2001). From this research we can determine the stable faults formed by α decay that will be accumulated in the lattice in the long term.