RADIOACTIVE WASTE MANAGEMENT RESEARCH: today's results bringing tomorrow's solutions

Research on radioactive waste management has made considerable progress over the past decade. Decision makers will have to choose the most suitable solutions from those for which governing principles have already been set, especially in the context of high-level, long-lived radioactive waste. France stands at the forefront of this effort and has its own legislation to fund specific research in this area. In return a comprehensive report on the research findings is to be delivered to Parliament in 2006.

CEA

View of the Atalante laboratories, at CEA/Valrhô-Marcoule, where the main hot experimental work is carried out on the processing and conditioning of radioactive waste.

The national and international background

France, as one of the world's major producers of nuclear electricity has been conducting research into radioactive waste management since the commercial nuclear program started many decades ago (box A, ***What is radioactive waste?***). Currently **long-lived** waste products are managed in safe but non-permanent **storage** facilities. So far no country has implemented a high-level, long-lived **radioactive** waste management option (box H, ***What are other countries doing?***). Sweden and Finland, among the most advanced countries, are committed to direct

deep **disposal** of spent fuel in geological formations. In the United States medium-level waste started being stored in the WIPP deep disposal facilities in 1999. In France, the law not only requires that research work be conducted on the deep geological disposal concept, which research is under the responsibility of Andra (the French National Agency for Radioactive Waste Management), but also that two additional avenues be looked at under the responsibility of CEA (box 1) : the feasibility of **transmuting** waste elements and the long-term interim storage concept.

The results of ten years of research

Many CNRS and university research scientists are working together with CEA and Andra teams (box 2). Major breakthroughs were achieved spawning new concepts and "long-term storage" is now a science in its own right. The prospects for radioactive waste really have evolved over the past few years.

Turning to waste volumes, process efficiency gains at the La Hague **processing** plant have enabled liquid effluent volumes to be reduced tenfold and solid waste volumes threefold. Scientists, on their side, have demonstrated that by using the so-called **advanced partitioning** process (see chapter I, *Sorting*), the most radiotoxic elements can be extracted from waste streams, for onward transmutation (see chapter II, *Transforming*) or specific **conditioning** (see chapter III, *Conditioning*): the feasibility of transmutation in various types of reactors has now been proven. Lastly the ability to predict how radioactive waste **packages** will behave over a very long period of time has been obtained (see Chapter IV, *Storage and/or Disposal*).

E. Joly/CEA

Remote-control manipulators of the Alceste unit for characterizing category A and B waste by destructive and non-destructive methods at CEA/Cadarache.

The 1991 Act

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Act No 91-1381 of 30 December 1991 (also known as the "loi Bataille" from the name of its mover), now incorporated into the French Environment Code covering the research on radioactive waste management resulted from the difficulties encountered during the 1980s by the French National Agency for Radioactive Waste Management (Andra) when conducting siting for the future installation of a high-level, long-lived radioactive waste-disposal facility. Coming at the same time as a fifteen-year moratorium on decision-making about the long-term future of this waste, the Act outlined a research program to be carried out during the moratorium and stipulated that Parliament should be presented with an overall appraisal report on this research in 2006.

Responsibility for line 1 (*research into solutions enabling long-lived radioactive elements present in this waste to be partitioned and transmuted*) was entrusted to CEA, while that of line 2 on *the study of the possibilities of reversible or irreversible disposal in deep geological formations, particularly through the construction of underground laboratories*, was entrusted to Andra.

Work on line 3 (*study of conditioning and long-term surface storage processes for high-level, long-lived waste*) is also being led by CEA. The French government moreover asked CEA to look into subsurface storage concepts.

The Act introduced parliamentary control of research work via the Parliamentary Scientific and Technological Choice Appraisal Office to which an annual progress report is submitted, drawn up by the *Commission nationale d'évaluation* or CNE (National Appraisal Commission) comprising national and international experts designated by the French Government, National Assembly and the Senate.

What is radioactive waste?

According to the International Atomic Energy Agency (IAEA), "*radioactive waste is any material, containing a concentration of* ***radionuclides*** *greater than those deemed safe by national authorities in materials appropriate for unchecked use, and for which no use is foreseen.*" In practical terms these substances must be conditioned in such a way that the radionuclides present are **contained** until their **radioactivity** is extinguished and so that substances forming a shield stop the emission of the ionizing radiation, which is potentially hazardous for living organisms.

In France the production of radioactive waste per inhabitant is about 1 kg per annum (of which only 20 g is high-level waste) compared to total waste production (including industrial) of roughly 2,500 kg, of which 100 kg is "eternal" toxic chemical waste (in particular, heavy metals), for which no removal or disposal industry exists at the present time.

The various types of waste

Radioactive waste is essentially characterized by the type of elements it contains and by the **activity level** per unit of volume or mass (expressed in becquerels, the number of spontaneous **disintegrations** per second, it enables the quantity of atoms of radionuclides contained in the waste to be assessed). Each of these radionuclides has a corresponding **radioactive decay** **half-life**, which indicates the time it takes to reduce its activity level by a factor of 2, a radiation emission type (**alpha**, **beta**, **gamma** or **neutrons**) and the energy transmitted by this radiation. The characteristics of the waste thus evolve over time to such an extent that account has to be taken of radionuclide **daughter products**. The phenomenon of spontaneous disintegration leads to radioactive nuclei giving birth not only to other stable nuclei, but also to other radioactive nuclei that will in turn disintegrate as dictated by their own characteristics. That explains the notion of how **radioactive chains** exist, in particular for natural elements such as uranium or thorium.

Antoine Gonin/CEA

Overboots and gloves, being taken off here by a technician leaving the controlled area of the OSIRIS reactor, at CEA/Saclay, end their life as category A waste.

Depending on the type, activity level and lifespan of the radionuclide constituents, radioactive waste has been classified into different categories, from **VLLW** waste (very low level) to high-level waste (**HLW**) with intervening low-level (**LLW**) and intermediate-level waste (**ILW**). It is said to be **long-lived** when its lifespan exceeds 30 years, as opposed to short-lived if the reverse applies.

In France, the distinction is made by the Nuclear Safety Authority between four major categories of waste along the lines of how they are managed in the long term (see table). Specific (glass, concrete) containment systems have been developed for each type of waste to provide suitable management. For some of them these are contingent on decisions committing them to the very long term. All these types of waste are finally placed in **packages** which can be grouped into five main types: packages of bituminized waste, packages based on hydraulic binders (cement, mortar, concrete), packages of vitrified waste, packages of compacted waste and packages of spent fuel, the latter being subject to research.

• Most **very-low-level waste**, or VLLW, comes from dismantled nuclear facilities (250,000 m^3 is expected between now and 2020 and 1,000,000–2,000,000 m^3 altogether). The final status of this waste is still under study. The options are to recycle it or dispose of it in a dedicated repository (as proposed by the Omega-tech project, from the name of the partnership formed by ANDRA (the French National Agency for Radioactive Waste Management) and the company France-Déchets. The approach is based on managing waste by categories, in line with its different characteristics. There are no generic waste release thresholds. CEA (the French Atomic Energy Commission) stores its very-low-level waste at Cadarache (Bouches-du-Rhône). The mean and maximum specific activity of this waste is respectively 10 and 100 Bq/g for beta-gamma emitters, and 1 and 10 Bq/g for alpha emitters.

• **Low-level** waste (classified in category **A**, which also includes some medium-level waste) for the most part contains **radioelements** that emit beta (β) and gamma (γ) radiation for a period less than or equal to 30 years, and thus presents a low-to-medium-level nuisance that becomes negligible after 300 years (10 periods), and alpha (α) or long-lived emitters with an activity level less than or equal to 3,700 Bq/g after 300 years. It originates from nuclear facilities (gloves, filters resins, etc), research laboratorand medical and industrial users of radioelements. By 2020 its volume will reach some 500,000 m^3, that is nigh on 90% of the total radioactive waste volume. It is stored in France in a surface **disposal** site. Andra manages two such sites: the La Manche Center (CM), which is full, and the Aube Center (CA), which is currently accepting waste. According to Basic Safety Rule (RFS) No 1-2 dated 19 June 1984, the combined average mass α-emitter activity level of the packages at the disposal center must not exceed 370 Bq/g at the end of the monitoring period (300 years). As for long-lived

low-level waste (**LLLLW**) such as radiferous waste and graphite waste (less than 15,000 m^3 in 2020), dedicated disposal sites are under consideration.

• **Medium-level** waste (category **B**, which also includes some low-level waste) contains significant quantities of radioelements, generally **actinides**, emitters of alpha radiation of a higher level than 3,700 Bq/g and for a period of over 30 years, that may go up to several tens of thousands of years without releasing much heat. This **MLLLW** (medium-level, long-lived waste) originates mainly from fuel-cycle plants (fabrication, reprocessing) and CEA research centers. It is the remains of structures that have contained nuclear fuel (box B, ***Waste from the nuclear power cycle***) and solid technological waste of various types and also the sludge from effluent-treatment plants (from treatment operations and CEA research laboratories), stabilized in **bitumen** matrices, a solution with increasingly limited uses, or **cement**-based matrices. The quantity of this waste is estimated at 60,000 m^3 in 2020. It is currently in intermediate and interim **storage** awaiting an as yet undefined outlet to fit the terms of the research recommended in the Act of 30 December 1991 (box 1).

• **High-level** waste (category **C**) containing alpha-, beta- and gamma-emitting radioelements with a life of over 30 years. It is made up of **fission** and **activation products** solutions from spent-fuel reprocessing. It accounts for by far the main source of waste *in terms of activity* although in volume it will only represent an accumulated volume of about 5,000 m^3 in 2020, equivalent to a 17-m-sided cube.

CEA

Example of waste embedded in a concrete matrix.

France currently produces a total of less than 200 m^3 of high-level waste per annum, equivalent to a 6-m sided cube. High-level waste is currently **vitrified** and is subject to intermediate and interim storage in appropriate facilities as category B waste.

The activity contained for this high-level waste varies from 10^{10} to some 10^{13} Bq per liter of glass depending on the solutions when the glass is produced. It then decays to return to a level close to the original mineral level after 10,000 years. The main sources of radiation in nuclear glass result from β decay originating from fission products such as cesium-137 (30-year period) and strontium-90 (28-year period), α decay arising from actinides (Am, Cm, Pu…) and the γ transitions that accompany α and β decay.

	short-lived main elements < 30 years	**long-lived** main elements > 30 years
very-low-level waste (VLLW)	dedicated disposal and recycling systems*	conversion to disposal of current interim storage*
low-level waste (LLW)	(category A waste) surface disposal at the Aube Disposal Center (Andra)	dedicated disposal * (radiferous waste, graphite)
medium-level waste (MLW)	dedicated disposal for tritiated waste *	(category B waste) systems under consideration (Act of 30 December 1991)
high-level waste (HLW)	systems under consideration (Act of 30 December 1991)	(category C waste)

* Management systems under consideration.

Table. Classification of radioactive waste in France and their management system status.

Different categories of waste

Waste is classified by physical type, in view of the treatment it has to undergo prior to storage and/or disposal, by distinguishing between solid and liquid waste. Spent fuel is not considered as waste.

Solid waste

Incinerables (protection accessories, demineraliser resins, etc.), metal parts that have been used in reactors and in "hot" laboratories or from the processing of fuel elements and other materials that belong to the category of **technological waste** (cables, miscellaneous apparatus, soil, etc.) fall into this category. Also included is waste with specific behavior (sodium, graphite, radiferous waste, boron carbide and toxic metals) as well as radioactive sources.

Liquid waste

Liquid waste comprises the aqueous liquids produced in laboratories, reactors and reprocessing facilities (fission-product solutions), which may contain different salts (nitrates, borates, phosphates, sulfates…) in varying concentrations, and organic liquids (oils, reprocessing solvents [tributylphosphate, dodecane] and others, scintillant liquids).

Spent fuel

Most spent fuel – from the nuclear power generating industry – is not considered as waste since it can be reprocessed using the Purex process in the Cogema workshops at La Hague. The quantity of spent-fuel output by the French nuclear power generation industry is currently 3,500 metric tons of heavy metal (box B, ***Waste from the nuclear power cycle***). Waste from experimental and prototype reactors such as Phénix and reactors of the old natural uranium/graphite/gas and heavy-water lines are also reprocessable as opposed to the spent fuel from some other experimental reactors (silicide fuels or irradiated oxides in Rhapsodie, France's first rapid-neutron reactor) and irradiated laboratory samples encapsulated in araldite.

The main joint research projects

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Radioactive waste management research being conducted by Andra (French National Agency for Radioactive Waste Management) and CEA principally takes the form of joint programs at a national, European or international level.

The partners working on the national program in public research institutes are the CNRS (National Center for Scientific Research) and the universities, which coordinate their actions in this area with the **Pace** program (*Programme sur l'aval du cycle électronucléaire*, or Nuclear power cycle downstream program) organized around five major research consortiums (GdR, for *Groupements de recherche*) on important research themes. This work also has input from members of the nuclear industry: EDF (Electricité de France), Cogema (Compagnie générale des matières nucléaires) and Framatome. The latter two are both subsidiaries of the Areva Group. The CEA takes a direct part in four out of these five RCs:

Practis (Physico-chimie de Radioéléments, des ACTinides, aux Interfaces et en Solutions) [Physical-chemistry of Radioelements, ACTinides, Interfaces and Solutions] (CEA, CNRS, EDF and Andra);

Gedeon (GEstion des DEchets par des Options Nouvelles) [Waste Management through New Options] to focus particularly on the subcritical systems assisted by accelerator and thorium-based fuel (CEA, CNRS, EDF and Framatome);

Nomade (NOuvelles MAtrices DEchets) [New Waste Matrices] to study new conditioning matrices (CEA, CNRS);

MoMaS (MOdélisation MAthématique et Simulations numériques) [Mathematical Modeling and Digital Simulations] to develop methods to simulate radioactive waste management problems (CEA, CNRS, Andra, BRGM, EDF);

A fifth RC, **Forpro** (FORmation géologique PROfonde) [Deep Geological Formation], brings the CNRS and Andra together on research into underground qualification laboratories.

The CEA is working together with EDF on the **Precci** program (Programme de Recherches sur l'Evolution à long terme des Colis de Combustibles Irradiés) [Long-term Evolution of Irradiated Fuel Packages Research Program].

On a European level, programs are currently under way under the aegis of the aegis of FP5 (the 5th Framework Program for research and development that covers the period 1998–2002), such as **Partnew**, a program on advanced partitioning which has taken over from **Newpart** of FP4, **Calixpart** on the selective extraction of radioelements by specific molecules and **Spire** on the effects of radiation on materials for hybrid subcritical systems with accelerator. CEA acts as coordinator on some of these research topics, while other bodies also coordinate, such as the European CERN, FZK from Germany, ENEA of Italy, SCK/CEN of Belgium, NRG from Holland and KTH from Sweden. The European Union's Common Research Center, GSI from Germany, ILL – a Franco-German-British body – and bodies outside the community such as the Paul Scherrer Institute (Switzerland) are also taking part in research programs.

Joint projects with the other major nuclear countries cover the three major lines of research involving Japanese partners such as JAERI (Japan Atomic Energy Research Institute) and JNC (Japan Nuclear Cycle Development Institute), the Russians (Minatom, IPC, Vernadsky Institute…) and the Americans (Department of Energy).

The international cooperation network supported by the CEA, Andra and the CNRS extends to international bodies such as the NEA, the OECD's Nuclear Energy Agency, especially on basic nuclear data and IAEA (the International Atomic Energy Agency) for biosphere modeling.

The nuclear energy industry can show that in addition to its particular benefits (see foreword), it can recycle its own highly **radiotoxic** waste using advanced partitioning. As a result of this research, the resulting and ultimate waste components are reduced to rather short-lived **radionuclides** that can be conditioned effectively to prevent migration into the **biosphere**.

CEA teams of physicists and engineers are working towards these goals in conjunction with their peers in all the major nuclear energy-producing countries (in Europe, Japan, USA) and are collaborating particularly closely with the USA, which has resumed its nuclear energy research work in the context of the "*Generation IV*" Forum.

Actinide-free vitrified waste

The greatest success was reached in the area of waste processing. So by 2006 it will be possible to make the decision to extract not only plutonium and uranium, as is routinely done at La Hague, but also americium, curium and neptunium. Altogether they make up all the **alpha**-emitting **actinides**, which are highly radiotoxic. Radioactive waste loses its thermal and radiotoxic load much quicker when the actinides are eliminated and their radiotoxicity level drops after a few centuries to that of the initial mined uranium.

Fission products

A few very-long-life **fission products** such as iodine-129, cesium-135 and technetium-99 remain. These three elements are important because of their chemical mobility and abundance in spent **fuel**. Research has shown that they too may be selectively extracted.

In the end, the residual waste will be suitable for either deep disposal, that is under conditions that will not require monitoring over time, or alternatively interim storage, a solution that by definition is not final. The golden rule of interim storage is that waste packages must be protected to remain intact and be retrievable at any time for reprocessing, reconditioning or transfer to deep disposal.

What should be done with the elements that have been separated out?

What will the various extracted elements become? **Reprocessed** uranium builds up strategic reserves. The plutonium will have to be incinerated as it becomes available through the extraction process. This is currently the practice for most of it, while the **minor actinides** can be partitioned for possible transmutation or specific conditioning should this option be chosen.

Transmutation

The possibilities of transmutation have been closely examined by simulating the operation of reactors loaded with minor actinides. Missing basic nuclear data have now been measured. Capsules (known as "target batches" in specialist jargon) containing americium have been prepared and, in some cases, tested in various reactors. A system design has been developed linking a **subcritical reactor** to an accelerator.

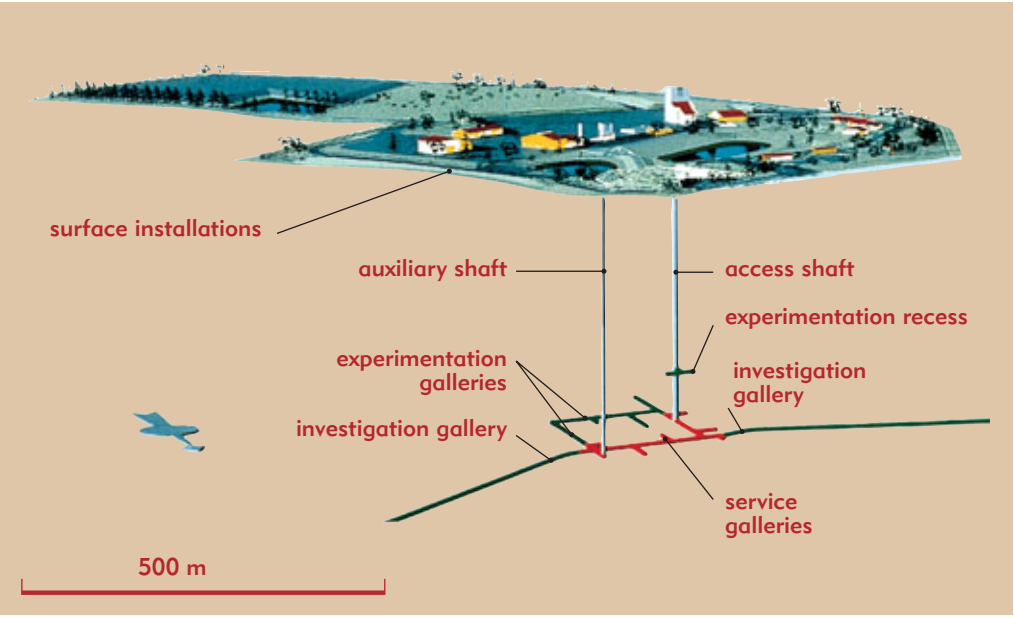
Research has now shown that the predicted transmutations are physically feasible, but that it could take about 100 years to reach a balance between the production and the destruction of radionuclides. Moreover because the handling of substantial quantities of highly radioactive elements is very onerous, the prospect is difficult in practical terms. Finally, controlling reactors loaded with minor actinides in significant quantities could turn out to be a delicate operation. Nevertheless, none of these objections is insurmountable, and there are reactors already capable of recycling minor actinides.

Les films de Roger Lennhardt/Cogema

A standard compacted waste package being stored at the La Hague plant compacting shop. Waste volume reduction is an area where progress has been made in recent years in addition to theoretical advances on radioactive waste management.

Specific conditioning

Because it is amorphous, glass displays great properties for the conditioning of a variety of elements present in spent fuel, and the latest research shows that it is an exceptionally durable **matrix** for use in a disposal. Small quantities of chemically pure substances are made available if advanced partitioning is carried out. These substances



Andra

Computer-generated picture of the underground research laboratory of Meuse/Haute-Marne, being constructed at Bure.

3

Waste volumes are dropping all the time

Apart from the work being carried out on long-term, or even permanent solutions, the French radioactive waste management industry has concentrated in recent years on drastically reducing waste volumes. It has achieved this either by compacting (metallic waste), incinerating or by applying other processes. Thus for the bulkiest, low-level **short-lived** waste (category **A**) (accounting for 90% of the total volume but less than 1% of the **radioactivity**), accrued production is now estimated at 500,000 m^3, whereas it was forecast as recently as the mid-1990s at 900,000 m^3 for the year 2000 horizon!

The main breakthrough is the elimination of **bitumen** that was used for conditioning sludge produced by fuel-cycle liquid-effluent treatment, the decontamination of **technological waste** (embedded in concrete blocks) and the compacting of hulls and end-pieces (metal parts left over from spent fuel) that were previously cast in cement. This research has enabled the overall volume of **long-lived** waste originating from **processing** operations to be reduced by a **factor of 5** between the time the plants were designed in the 1980s and 2001, and by a **factor of 3** since 1991 (figure 1 illustrates the progress made in the UP3 plant at La Hague where Cogema processes imported spent fuel).

The production of bitumen in the UP3 plant (about 0.6 m^3 per reprocessed metric ton) came to a halt in 1995 when a new system came on stream for managing liquid effluents generated by the treatment process that enables all radioelements to be vitrified. Research focused on process optimization aiming to limit the presence of mineral elements in the reagents used (sodium in particular), so that the concentration of effluents could be boosted and moreover guarantee that they would be incorporated properly into the glass, making for more reliable radioelement containment. Shutdown of the "bitumen" shop not only resulted in reducing waste volumes, it also reduces activity dispersal in waste packages of various types (medium-level, long-lived waste inventory reduced) and significantly reduces the residual activity of effluents originating from processing associated with bituminisation, discharged into the sea.

Another line of research has focused on perfecting the recovery of plutonium fixed on various types of technological waste.

The reduction in volume of "hulls" (metal fuel-cladding sections) has also been the subject of much research. CEA has developed a cold-crucible casting process that not only reduces volumes but also concentrates **transuranic element** activity in the casting area, which results in some degree of decontamination.

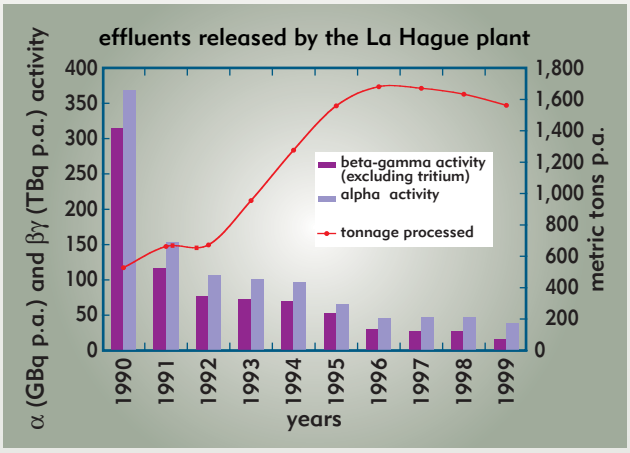


Figure 2. Evolution of liquid waste at La Hague highlighting how volumes have plummeted despite the increase in tonnages processed.

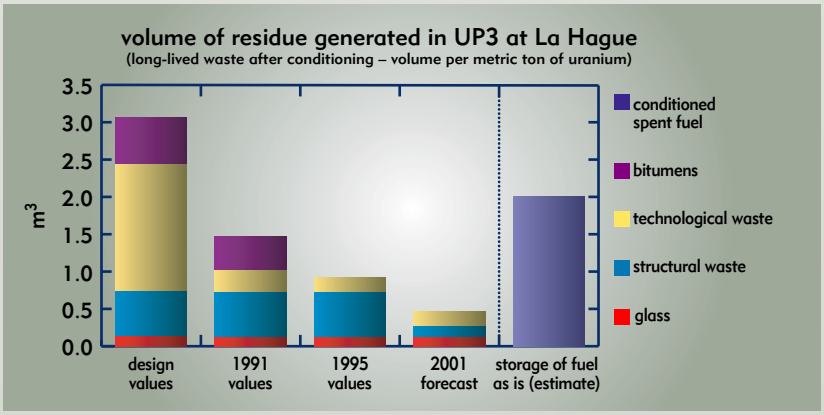
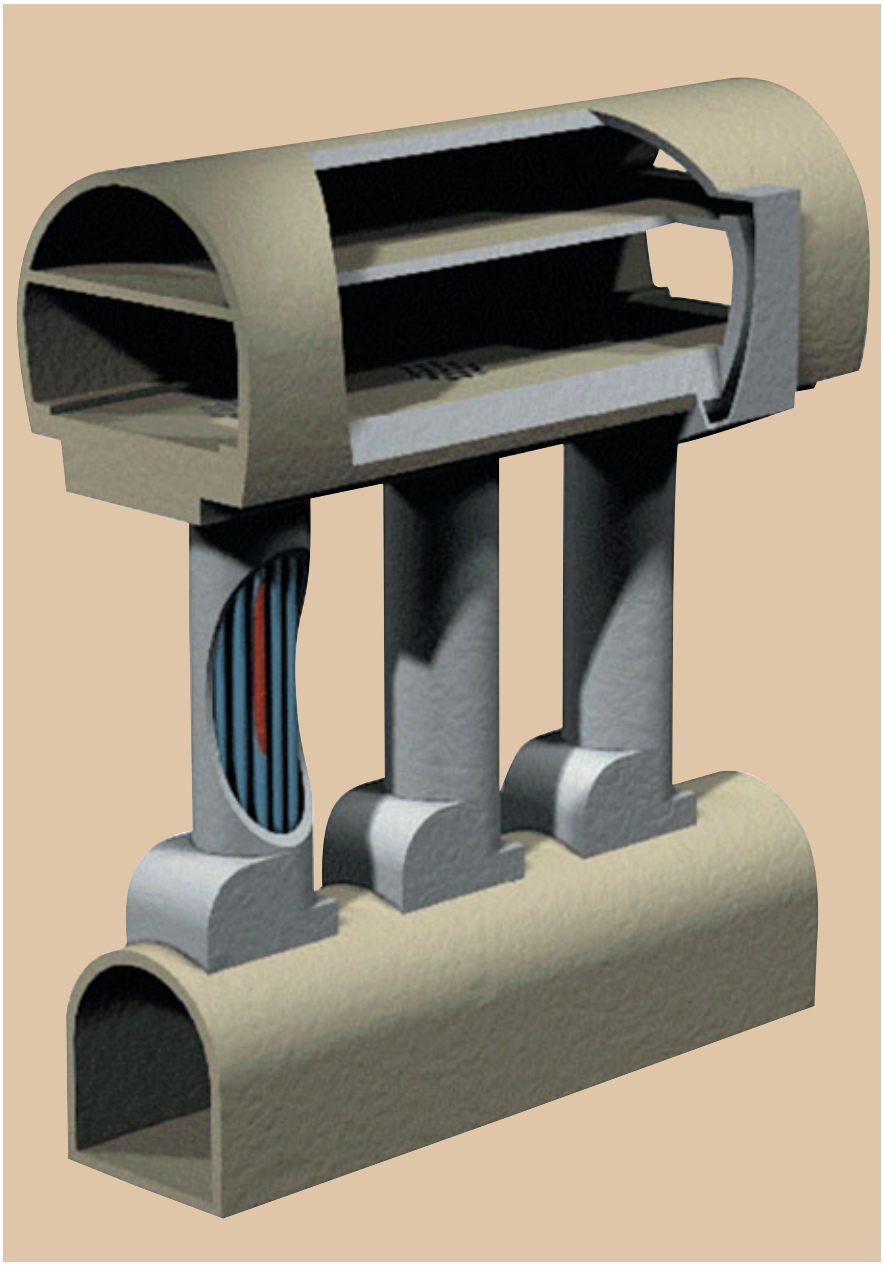


Figure 1. Volume of residue generated in the UP3 plant at La Hague broken down by type of waste.

Reduction of releases

Research into liquid effluents has basically centered on developing assisted-filtration technology to perfect purification of effluent radiation contaminants discharged into the sea. This process, adapted to the treatment of a very high volume of very-low-level contamination effluents, aims to reduce the residual concentration of contaminants by a **factor of 10** by adsorbing them on macromolecules prior to filtration. Model scale tests have tested the feasibility of such a process. The interest must lie in boosting facility-draining operations, which should generate a major flow of effluents. Furthermore a variety of specific initiatives have been conducted, such as on the treatment of effluents resulting from analytical work which largely contribute to the concentration of alpha-emitters in effluents. Thus in the space of a decade, all the above has led to the reduction of activity released by a factor of almost 10, despite the very considerable increase in the tonnages processed (figure 2).

Research on gaseous effluents has basically centered on trapping iodine (**fission product**). A process has been developed to recover the iodine deposited on solid adsorbant filters and also a trapping process in hydroxyl ammonium nitrate which could reduce concentrations of the sodium used to destroy the acid compounds produced by breaking down the processing solvents in effluents destined for vitrification.



CEA

Subsurface vitrified-waste storage concept.

thereafter can be introduced into a specifically designed **ceramic** that would resist the leaching effect of water until their **radioactivity** is extinguished. The potential for the elements trapped in this ceramic to return to the biosphere would be significantly reduced by an equivalent factor. Research carried out by the **Nomade** joint project is encompassing a number of options.

Medium-level waste

While high-level, long-lived waste poses the most acute problems, it only accounts for a very small proportion of long-lived radioactive waste (1,550 m^3 of the 30,000 m^3 accumulated in France between the beginning of the nuclear era and the year 2000), (box 3), the remainder of the long-lived waste is made up of medium-level waste presented in a large variety of forms (box B, ***Waste from the nuclear power cycle***). The most recent is conditioned in standardized form; the older waste is in the process of being reconditioned. Characterization methods have been developed to help in the selection of the most appropriate conditioning systems for each waste form, and followed by storage or disposal.

Waste restrained in a matrix and enclosed in a sealed **package** loses all effective harmfulness. Research carried out in the past decade has shed light on the long-term behavior of the main conditioning systems: glass, bitumen, concrete, uranium-oxide matrix (UO_2) and its cladding for spent fuel, etc.

Deep disposal in geological formation

Whatever decisions are made they will necessitate the creation of **near-surface**,

Waste from the nuclear power cycle

High-level, **long-lived radioactive** waste (HLLLW or category "**C**" waste according to the French classification system) basically originates from the spent **fuel** of the nuclear power plant industry.

Irradiation of fuel in the reactor leads to the internal formation of very diverse **radionuclides**, resulting from nuclear **fission** or **capture** reactions involving **heavy nuclei** (uranium and plutonium), and to a lesser degree the **activation** of other elements present in the fuel or surrounding structures (table 1). Thus for their part the "**hulls**" (metal cladding sections that contained pressurized-water reactor – **PWR** – fuel) and "**end-pieces**" of their fuel assemblies make up category "**B**" waste.

High-level, long-lived radioactive waste is only a small part of the fuel mass (about 4% for uranium oxide-based fuel as used in the majority of EDF's 58 PWRs). It is made up of products from nuclear fission, so-called **fission products** (FP) and **minor actinides** (neptunium, americium and curium) so described because of their low concentration (less than 0.1% of the spent fuel mass). A limited number of these so-called long-lived elements have a high radioactive decay time span, that is over 30 years. Less than 10% of FPs falls into this category and some have a life span of several thousand or even several million years (table 2). It should be noted that a very long life span is not a disadvantage where very low-level activity waste is concerned. Minor actinides (MA), which are heavy nuclei (in contrast to FPs which are fragments of them), are for their part made up of elements containing at least one **isotope** or a long-lived **daughter product**.

This category "C" waste is highly radioactive and thermogenic during the first decades of its existence as it also contains high concentrations of short-lived fission products such as strontium-90 and cesium-137. It is considered as the ultimate waste of the nuclear-power fuel cycle, as it is totally depleted of usable energy.

This does not hold true for 96% of the mass of spent fuel represented by uranium (95%) and plutonium (1%), both **actinides** described as major. These two elements can be reprocessed to produce energy whereas the three metric tons of spent fuel that contain them have already produced 1 billion kWh, enough to cover the whole annual electrical power requirements of about 70,000 people!

Nowadays plutonium is recyclable in the form of **MOX** fuel in some of the power industry reactors (applicable to some twenty reactors). The residual uranium can for its part be re-enriched (and recycled in lieu of mined uranium) but it appears most of all as a very important future potential reclaim resource with the arrival of new reactor technologies.

The recycling of uranium and plutonium (immediate or deferred) forms the basis of France's current **processing** strategy, and is applied to most of the spent fuel (two-thirds of the 1,200 metric tons of fuel annually discharged by power stations). "Ultimate" waste amounts to less than 20 kg of fission products and less than 500 grams of minor actinides for the 500 kg or so of uranium initially contained in each fuel element and after separating the 475 kg of residual uranium and about 5 kg of plutonium.

The Purex uranium and plutonium extraction process

Spent-fuel reprocessing essentially relies on the Purex (Plutonium Uranium Refining by Extraction) hydrometallurgical process implemented at Cogema's La Hague plant. The fuel is dissolved in boiling nitric acid. The uranium and plutonium are selectively extracted by a **solvent**, tributylic phosphate (TBP), with a recovery rate of almost 99.9%. The waste from these operations, apart from the metallic remains of fuel assemblies (see above) is dissolved at a rate of about forty kilograms to 600 liters of nitric acid solution. It is calcined then blended with 200 kg of glass, which once solidified, represents a volume of 110 L. It is in this

1 H																	2 He
3 Li	4 Be	heavy nuclei fission products (long-lived radionuclides)									activation products fission and activation products		5 B	6 (C)	7 N	8 O	9 F	10 Ne
11 Na	12 Mg												13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 (Se)	35 Br	36 Kr	
37 Rb	38 Sr	39 Y	40 (Zr)	41 Nb	42 Mo	43 (Tc)	44 Ru	45 Rh	46 (Pd)	47 Ag	48 Cd	49 In	50 (Sn)	51 Sb	52 Te	53 (I)	54 Xe	
55 (Cs)	56 Ba	*Ln*	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn	
87 Fr	88 Ra	*An*	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Uun									

lanthanides	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 (Sm)	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
actinides	89 Ac	90 Th	91 Pa	92 (U)	93 (Np)	94 (Pu)	95 (Am)	96 (Cm)	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

Table 1. Main elements present in spent nuclear fuel.

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category	element	isotope	period (years)	mass (g/t)	isotope content
minor actinides	Np	237	2,140,000	430	100%
	Am	241	432	220	67%
		243	7,380	100	31%
	Cm	243	28.5	0.3	1%
		244	18.1	24	94%
		245	8,530	1	5%
fission products	Se	79	65,000	4.7	9%
	Zr	93	1,500,000	710	20%
	Tc	99	210,000	810	100%
	Pd	107	6,500,000	200	16%
	Sn	126	100,000	20	40%
	I	129	15,700,000	170	81%
	Cs	135	2,300,000	360	10%

Table 2. Inventory of the main long-lived elements present in uranium oxide (UOX) spent nuclear fuel discharged at 33 GW·d/t.

matrix with its proven durability properties that it is **stored** on site, in metal containers inserted in specially designed and controlled shafts.

Alongside this *Colis standard de déchets vitrifiés* (Standard vitrified-waste package) Cogema has developed a package of the same format without a matrix, the *Colis standard de déchets compactés* (Standard compacted-waste package), for hulls, end-pieces and other metal technological waste items.

Sidney Jezequel/Cogema

Loading glass containers into a TN 28 cask at Cogema's La Hague plant.

subsurface and/or deep disposal sites to accommodate radioactive packages. This last solution presents definitive advantages. Deep disposal effectively provides a geological **barrier** to isolate the packages from the biosphere, high chemical stability of groundwater to stem matrix corrosion and lessens the risk of human intrusion, which reduces the obligation for monitoring.

Deep waste disposal is no innocent gesture, as it must be based on a great level of trust between all parties concerned. Sweden appears to be ready to take the step but France has not got to that stage yet. Introducing the notion of **reversibility** could be pivotal in the decision.

Various solutions for disposing of radioactive waste have been devised only to be abandoned not so long ago: sending it to the Sun, depositing it in a continental-plate subduction area or even into sub-oceanic sedimentary beds. We are left today with well-controlled disposal, in deep, stable geological formations and the current industrial solution: interim near-surface or subsurface storage. Cogema already operates an interim storage facility for vitrified waste at La Hague which, with rather minor changes in the design, would meet the criteria developed by CEA for the long-term "quality label". As one of them, the need for monitoring means that this waste management method cannot be permanent. CEA has conducted in-depth research to design conditioning methods and storage facilities capable of lasting centuries and requiring minimal monitoring.

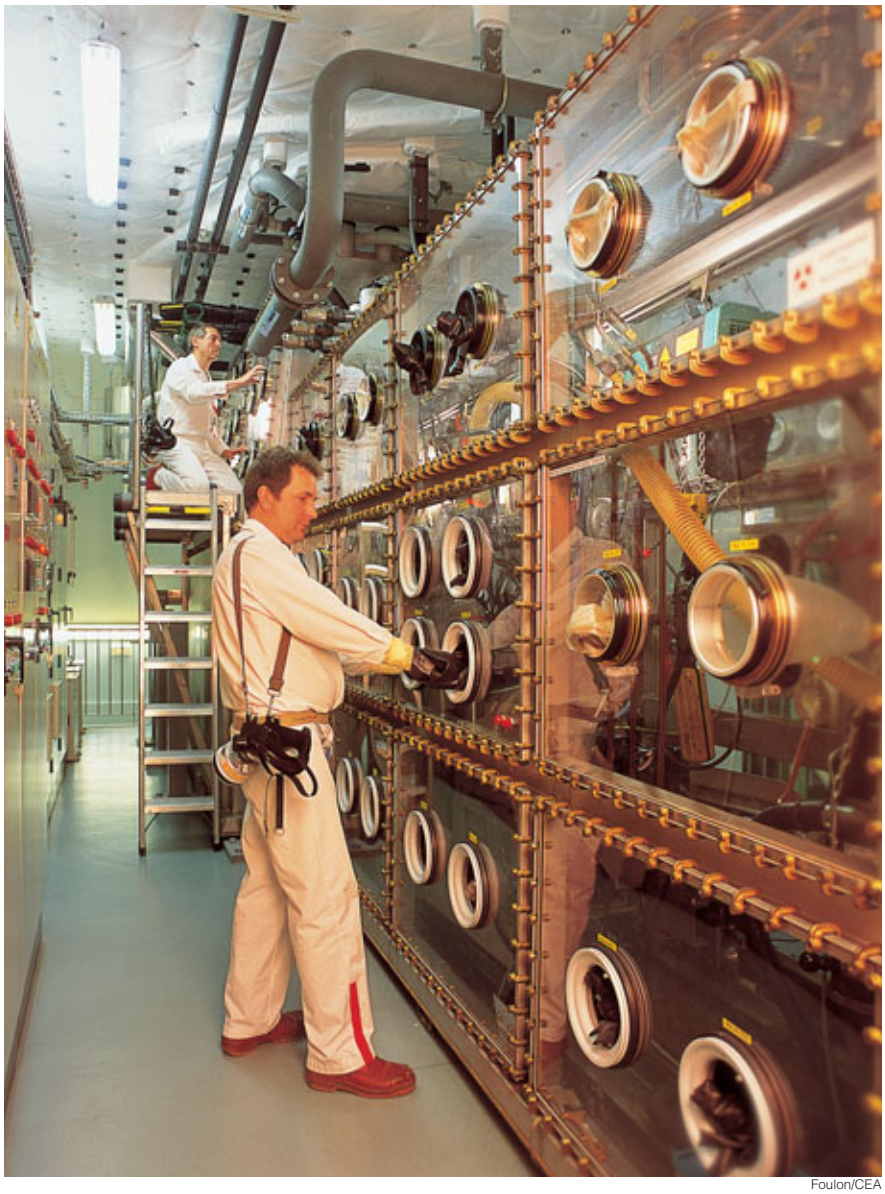
Spent-fuel processing without partitioning

If the decision was made not to process all or part of the spent fuel, it would have to be stored or disposed of as is. It would then retain its plutonium, an important energy resource and the main radiotoxic component. CEA is carrying out major research to identify the best conditions for interim storage, as spent-fuel pools are not a long-term storage solution. In 2006 CEA will produce concepts for a canister-packaging system and for long-term interim storage, both near-surface and subsurface solutions. CEA is also working together with Andra on deep disposal research.

The radiotoxicity of waste

Radiological protection standards have drawn from the findings of epidemiological studies on historical cases of exposure. Current radioactive waste management is such that these standards are being met by a wide margin. However the standards were not designed to cover disposal or long-term storage situations. This explains why CEA is

The alpha incinerator of CEA's Military Applications Division at Valduc (Côte-d'Or) uses glove boxes to process plutonium-contaminated combustible waste (PVC, neoprene, latex, polyethylene and cellulose) whose activity level is such that it cannot be stored in near-surface facilities. This brainchild of the Iris process (Installation for research into incineration of solids), developed at CEA/Marcoule in collaboration with CEA/Valduc and built by SGN, aims to reduce conditioning volumes and provide safe storage without compromising the reversibility of the conditioning and recovery of the plutonium at a later date. Following pyrolisis of the waste and calcination of the resultant pitch, post-combustion is carried out on the gases produced in the electrically-operated rotary furnaces. After cooling at 165 °C, it undergoes pre-filtration through electrostatic filters, filtration through very-high-efficiency filters and dry chemical purification using spongiacal lime. This process produces a gaseous effluent that complies with the legislation on releases into the environment. Since it was started up in 1999, the facility has enabled waste volumes to be reduced by a factor of 20 in the form of ash that traps about 93% of the initial activity, the remainder of the plutonium is subsequently recovered in the dust of the electrostatic precipitators and cleaning residues.



Foulon/CEA

continuing its research into radiotoxicology and radiobiology, especially on the problem related to chronic effects caused by low doses.

The choices for 2006

Given the initial profusion of research angles, it is now important to focus on a clear presentation of the findings and thus change course slightly and pursue those lines of research whose findings will carry most weight for the decisions to be made in 2006. By defining priorities one must avoid two pitfalls: narrowing the range so much that no choices would be left ; leaving the field so wide open that no well-informed decisions could be made.

A proposal with possible secondary options will be put forward for each type of waste. Each will be backed up with judgmental arguments so that informed opinions and appraisals can be made on the net benefits and the possible implementation difficulties along with the costs involved. Therefore, for each proposal put forward, the research program will have explored the most realistic routes possible to provide solutions to the problem of radioactive waste management.

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