



Industrial solutions for long-lived, high- and intermediate-level waste

For several decades, Areva has been reprocessing spent fuel, and conditioning ultimate waste, in its La Hague plants. Bearing in mind “historic,” legacy volumes, and the considerable gains achieved in terms of volume reduction, long-lived and high-level waste yielded by operation of the current generation of reactors now only accounts for an almost marginal flow, relatively to the existing inventory. Such optimization will carry over to the third generation of reactors (EPR). The advent of the fourth generation, and the coming on stream of a new reprocessing plant, which may be located around 2040, may, in the long term, allow further advances.



Areva/La Hague

The La Hague (Manche *département*) site has been host to the gradual construction of its various workshops, spread over time, and space. Optimization of the site, set in place over the past few years, has made it possible to arrive at an homogeneous whole: the exemplar for the 3rd generation of spent fuel reprocessing.

From the outset of nuclear power development, in the 1950s, France opted for the **treatment-conditioning** pathway, to recover such reusable materials, from an energy standpoint, as **plutonium** (Pu) and **uranium** (U). With the first generation of **UNGG reactors**, **reprocessing of spent fuel** was deployed in the initial **UP1** plant, at Marcoule, in 1958, this being followed in 1966 by the **UP2-400** plant at La Hague (Manche *département*).

Following successful operation of these plants, **EDF**, and many customers from other countries (Germany, Japan, Belgium, Switzerland, the Netherlands) firmed up contracts for construction, and subsequent operation, by **Areva** (then trading as **Cogema**), of the **UP2-800** and **UP3** plants, both sited at La Hague. These modern plants came on

stream, respectively, in 1994 and 1989. They allow reprocessing of fuel from second-generation reactors, such as the **PWRs** in EDF's current fleet (see Box 1 in *Radioactive waste management research: an ongoing process of advances*). A further plant of this type is being built at Rokkasho-Mura (Japan), for the reprocessing of fuel from light-water reactors. The coming 20 years will not see any major upheaval in the present system. The aim will be to carry through optimization of these plants, before the possible achievement of technological breakthroughs, coupling, for instance, fourth-generation reactors (see *New systems to curb waste at source?*) and new treatment-conditioning functions, which may include, among other processes, advanced partitioning (see *Less radiotoxic ultimate waste?*).



Glass being cast in the Marcoule Vitrification Workshop (AVM: Atelier de vitrification de Marcoule).

What was the waste management strategy in plants of the past generation?

The main principles of **waste** management at these plants were decided on quite early on. Thus, as early as the 1970s and 1980s, the foundations were laid down of current, modern systems, based on characterization of waste **activity** as early as feasible, concentration of that activity, traceability of **primary waste**, and zoning of installations, along with volume reduction. The government order of 1984 on quality confirmed then-current good practices, and set a framework for the system, such as to vouchsafe the modes of, and conditions for, waste conditioning. One major feature of the options taken at the time was the priority set for waste conditioning, once an operational long-term management pathway had been deployed. An essential component of such pathways is the **outlet**. The first two to be developed were submersion in deep ocean trenches, this being a marginal practice up to the 1980s, for limited amounts of **LLW** waste, and, in the main, final **surface disposal** at the Manche Disposal Center, close to La Hague, initially run by CEA, then by **ANDRA** (see *Industrial solutions for all low-level waste*), when this government agency was made a self-standing organization by law.

The management strategy adopted, for waste for which no long-term management pathway was available at the time, was to **store** it, without conditioning, pending availability of a conditioning technique, and a suitable outlet. A number of exceptions should however be noted. For some of the more highly radioactive waste, conditionings were carried out, to guarantee safe, durable industrial storage of such waste. This was the case for drums of **bitumen-encapsulated waste** at Marcoule, conditioning of which was initiated as early as the 1970s. The most illuminating example of this approach concerns **fusion products (FPs)** from spent fuel from “legacy” nuclear power stations (see Box B, *Waste from the nuclear power cycle*), partitioned at Marcoule and La Hague, and stored for over 20 years, for some of it. This respite allowed deployment of R&D programs to specify the **vitrification** option, develop-

ment of the process at CEA, construction of pilots (Piver initially, then AVM, at Marcoule), followed by industrial workshops, finally the vitrification of these products in forms offering high performance in terms of compactness and durability. Thus, currently, all FPs yielded by older generations of French nuclear reactors have been vitrified, bar a few tens cubic meters, corresponding to reactors using a specific, older technology (so-called “UMo” fuel), and these are to be vitrified in the near future.

Consequently, it now becomes possible to sum up this period, by pointing out that, “historically,” management carried out by industry, at the back-end of the French fuel cycle, was able to combine caution, responsibility, and an urge to advance, to arrive at one of the most enviable positions, internationally speaking. Waste is now either wholly in disposal, at suitable sites, or safely stored under a variety of forms, conditioned or otherwise. In the latter case, retrieval and conditioning programs, for such “legacy” waste, are ongoing, and are due to pick up speed in the coming years (see *Nuclear waste management and processing: between legacy and anticipation*).

The relatively limited amount of electricity generated by UNGG reactors of that historic generation resulted in ultimately quite large masses of spent fuel, owing to the low **burnup rate** possible on these reactors. The inventory of **HLW** and **ILW-LL** waste yielded by that generation thus amounts to several tens of thousand cubic meters.

Since that time, current reactors, and the associated cycle performance, have allowed bringing down by a factor 50 the volumes of HLW and ILW-LL waste, per **kilowatt-hour (kWh)**, relative to that first reactor generation. Thus, to give an order of magnitude as to the advances achieved, in terms of waste volume, had that historic fleet generated 400 **TWh** annually, over 40 years, as planned for the current generation, one should have tabled on one million cubic meters or so of that category of waste. In fact, the national inventory drawn up by ANDRA assesses current HLW and ILW-LL waste at a mere 45,000 m³ or so, owing to the fairly low output from this generation (in effect, a few hundred terawatt-hours only), and the fleet being switched to PWRs and modern reprocessing.

Waste conditioning by plants of the current generation: recent developments

The initial design for current-generation plants, intended to carry out reprocessing of PWR fuel, dates from the 1980s. It is thus based on improvements over then-current processes, with a significant departure in the principles involved, with the in-line conditioning of all waste produced.

What was the performance anticipated at the time they were designed?

When designed, a plant such as UP3 was expected to yield a volume of HLW and ILW-LL waste of some 3 m³/tonne of

Cross-section of a package of solid waste immobilized in a cement matrix. Such waste is compacted, nowadays, to achieve volume reduction.





heavy metal (thm) processed. Such volumes correspond to close on 200,000 m³ for a fleet supplying 400 TWh over 40 years. The gain initially planned for was thus of a factor 5, over the preceding nuclear system. As regards waste directly yielded by spent fuel, **fission products** are conditioned in line, in borosilicate glass: this is a major step, coming after the decades of research mentioned earlier (see *Safe conditionings obtained through constantly improved processes*). **Structural waste** was conditioned, when these plants started up, in **cemented** metal drums. As for waste relating to operation of the installations, the La Hague plant treats and conditions its own **operational waste** and **liquid effluents**. **Technological waste** was initially conditioned in cemented **packages**, with a overpack, for protection and **confinement** purposes, mainly made of fiber concrete.⁽¹⁾ Liquid effluents were decontaminated by fixing their activity in sludges, in accordance with provisions decided on when plant design was drawn up. These sludges were subsequently conditioned by **encapsulation** in a bituminous **matrix** (see above).

What are the advances achieved in the area coming under the Act of 1991?

The Act of 30 December 1991 (see Box 2 in *Radioactive waste management research: an ongoing process of advances*) was passed effectively at the time the plants of the generation currently in service were starting up. The Act covers, essentially, research on high-level and long-lived waste (i.e. HLW and ILW-LL waste), and includes, in particular, an important set of provisions devoted to improvement of industrial practices.

For the major part, the advances achieved since that time are the outcome of the numerous findings from R&D programs set up under the aegis of this Act. A **standard compacted waste container (CSD-C)** (see Box A, *What is radioactive waste?*) was designed for compacted metallic structures and technologi-

(1) Fiber concrete: a mix of concrete and cast-iron fibers, resulting in remarkable mechanical cohesion for the material.

cal waste. *Use of bituminization* was eliminated, through sorting, concentration, and recycling of effluents, and direction of residual streams to vitrification. The *management of technological waste conditioning* was optimized by way of improved plant management, to curb the outflow of waste, and reduce waste activity, and through precise activity characterization, and the specification of conditionings exactly adapted to each activity range. The *E-EV-SE storage facility* (for **standard vitrified waste containers [CSD-Vs]**) and *ECC storage facility* (for CSD-Cs) were targeted by design studies, with reference to criteria of durability and long-term life (see *Is industrial storage suited for long-term management?*). *Models of waste package behavior* in disposal conditions have been arrived at, and have highlighted, in particular, the durability of CSD-V packages.

These results went hand in hand with initiatives to secure ongoing advances, initiated by the operator (workshop zoning, sorting at source, recycling, improved measurement performance...), making a very significant contribution to curbing waste volume. Thus, the annual volume of HLW and ILW-LL waste was brought down by a factor of over 6, relative to design parameters for the reprocessing plants, presently standing at a volume of less than 0.5 m³/thm. Such a figure corresponds to some 20,000 m³ for a fleet generating 400 TWh, over 40 years' operation, as conservatively assumed for assessment purposes.

Bearing in mind that the inventory of HLW and ILW-LL waste is currently assessed by ANDRA at less than 100,000 m³, for all waste arisings, present and future, up to shutdown of the generation lots currently in operation (see *An inventory projecting into the future*), and that mean fleet age stands at some 20 years, one thus finds that projected production of waste packages, accounted for by future operation of the fleet, only accounts for 10% of the total (half of the 20,000 m³ noted above). Even allowing for increased reactor operating life, the advances achieved by the reprocessing industry mean future waste will be but marginal. This corroborates the finding that by far the greater part of the HLW and ILW-LL waste inventory comes from "historic" nuclear power, generated at a time when waste volumes, per kilowatt-hour, were markedly higher than with current flows. Present and future production of ILW-LL waste remains marginal.

Current waste conditioning by third-generation plants: the state of the art

As a result of the above-described developments, waste conditioning has achieved a state of the art, of which the first point of note is the production, from the end of 2002, of the two types of standard **container** (see Figure 1). The advent of the standard waste container (CSD: *conteneur standard de déchets*) has indeed made it possible for all HLW and ILW-LL waste to be conditioned in a single container model, with variants in the form of the two package types. On the one hand, 99% of spent fuel activity is concentrated and cast into the borosilicate glass matrix of CSD-Vs. And, on the other hand, struc-

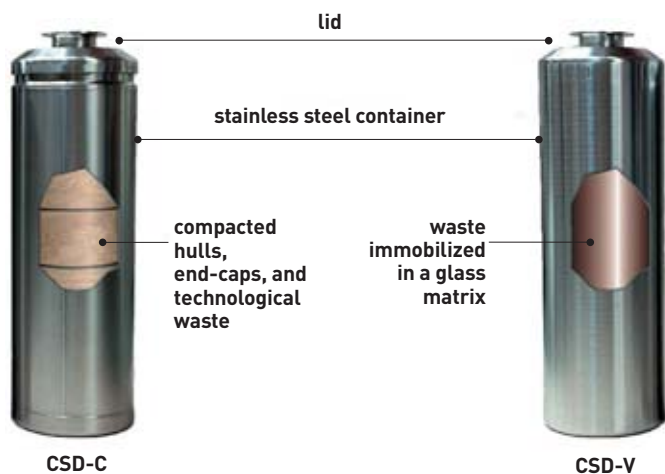


Figure 1. CSD-Cs and CSD-Vs are standard containers, identical as to their nature and dimensions, used to contain most of the long-lived **radioelements** generated. The CSD-C holds compacted structural and technological waste, while the CSD-V holds vitrified fission products and **minor actinides**.

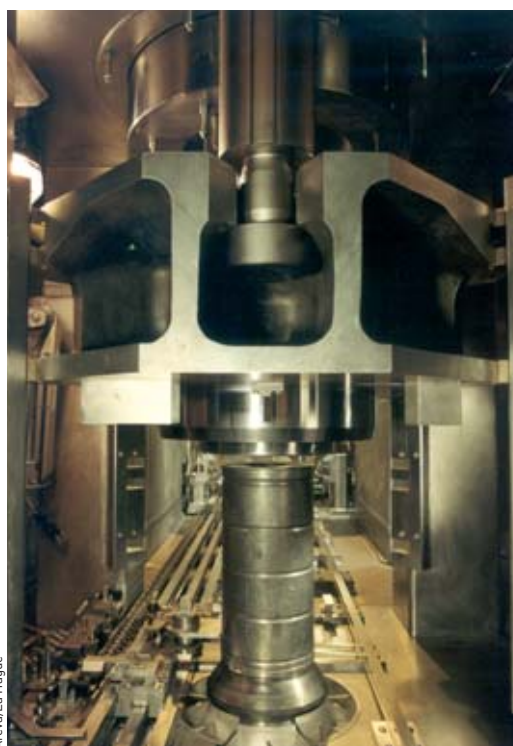


Areva/Philippe Lesage

Compaction of **hulls** and **end-caps**, waste from the structures of spent fuel assemblies, has taken the place of cementation, thus yielding a reduction in volume by a factor 4.

tural and technological waste, for which activity levels are such as to preclude surface disposal, is compacted into CSD-Cs. The two packages share the same dimensions. Standardization thus allows synergies to be derived at every stage in package life. CSDs thus represent a flexible, high-performance conditioning. Standardization of external geometry allows optimization of handling equipment, and shared use of storage and destorage facilities, transport packing, and, when applicable, **geological disposal** installations. These packages are fabricated under a rigorous quality system, including qualification of conditioning processes, a specification for production of the waste packages themselves, and controls at every step in production, under constant monitoring by an outside organization, certifying that production conforms to specifications. Stability is guaranteed over a scale of centuries, this being the timescale for industrial management. CSDs further exhibit high stability on a geological timescale, with a lifespan, for the glass matrix, of the order of several hundred thousand years to one million years for CSD-Vs (see *What long-term behavior for nuclear waste packages... and for spent fuel?*). The various package categories could, in some designs, be used to optimize final disposal, by smoothing overall thermal power output, through use of (cooler) CSD-Cs as “intercalaries.”

CSD-Vs have been in production at Areva's La Hague unit since 1989. To date, over 10,000 containers have been fabricated, with a production ratio close to 0.7 CSD-V/tiU (tonne of initial uranium). This amounts to a volume of less than 2,000 m³ overall. The vitrification technique for FPs in a borosilicate glass matrix has nowadays gained worldwide recognition as the solution best suited to HLW waste conditioning. The French process for vitrified residue fabrication has been approved in many countries, notably in Western Europe and Japan. Returning CSD-Vs to the country of origin of reprocessed spent fuel is now a daily occurrence: over 50% of CSD-Vs, corresponding to their original spent fuel, are already held at the storage facilities of Areva's many foreign customers.



Areva/La Hague

A press in the Hull Compaction Workshop (ACC: *Atelier de compactage des coques*), which came on stream at La Hague in 2002. Mixed hulls and end-caps, or technological waste, are compacted under a load of 2,500 tonnes, at ambient temperature, to form high-density pellets.

CSD-Cs have been in production at Areva's La Hague unit since 2002, after over 10 years' R&D. More than 4,000 CSD-Cs had been produced to the end of 2005, with a production ratio of less than 1 CSD-C/tiU. The techniques used at the ACC compaction workshop are novel, on an industrial scale, for an environment suited to nuclear materials, particularly with regard to the active and passive measurement systems, reaching a high level of sophistication. The fabrication process for CSD-C packages is undergoing approval procedures in many countries, in the wake of the CSD-V.

In-storage management for such CSD packages exhibits, in turn, high performance levels. Thus, the storage buildings for CSD-Vs (E-EV-SE workshop) and CSD-Cs (ECC workshop), at the La Hague site, are

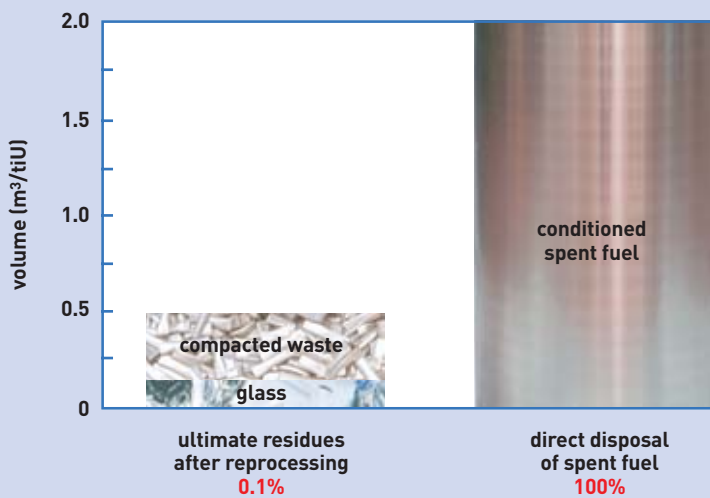


Figure 2. Volume reduction, as between reprocessing of spent fuel, and possible direct disposal of the same, is achieved to a factor of around 5.

compact, fully reversible, and compatible with long-term storage (on a scale of centuries). 12,000 CSD-Vs and 24,000 CSD-Cs may be kept in storage, in a space from 3 to 5 times smaller (depending on designs) than if the spent fuel were stored directly in storage casks. Potential building extensions would allow storage of waste yielded by close to 100 years' nuclear power generation, on a surface the size of two rugby-football pitches.

Referred now to the volumes that would need considering in designs for the direct disposal of spent fuel, the volume reduction enabled by reprocessing reaches a factor of 4 or 5. This obtains after taking into account package thermics, for the dimensioning of the disposal facility (see Figure 2), this being of far less weight for designs involving reprocessing. Consequently, assessments carried out in France or Belgium indicate costs pegged a ratio of some 2.5, in favor of the solution involving conditioning after reprocessing, compared with possible direct disposal of spent fuel.

What are the directions for advances in the two coming decades?

Constant advance, stoked by the R&D programs carried out at CEA and by evolving industrial practices, will allow new gains to be achieved in a number of areas. New margins for advances will emerge with the arrival of the **EPR**, particularly relating to the reactor's characteristics (burnup rate, and efficiency). Beyond this, potentially significant advances could be arrived at with the switch to fourth-generation plants, around 2035–40 (see Figure 3). Aside from carrying through volume reductions, advances will concern other aspects, such as integration of the ensemble of stages in the packages' "lifecycle" (including long-term management, when this is deployed), together with relevant optimizations: dosimetry, safety, costs. Concurrently, programs are being launched, and are due to expand significantly, for the "retrieval," i.e. the taking out of storage facilities, and conditioning of waste present at the La Hague and Marcoule sites, inherited from the previous generation, and not as yet conditioned.



Cross-section view of a standard compacted waste container (CSD-C), showing the stack of pellets of compacted structural waste (hulls and end-caps) or technological waste.

Evolution of the CSD-V "in line"

One major aim, in the short and medium term, is to keep to a ratio of standard containers per tonne of initial uranium below 0.5 m³/tiU, while supporting customer utilities as regards evolutions in **core** management, in particular increased burnup rates, inducing a higher-than-proportional rise in **alpha emitters**.

The limiting rate for FP and **actinide** incorporation into glass is due, on the one hand, to chemical limitations, and, on the other, to the effects of glass **self-irradiation**. The current specification, aside from parameters restricting oxide content and FP (cesium and strontium) activity, sets limits for actinides (uranium, plutonium, curium), on the basis of the reasonably expected "envelope" amounts for **UOX1** fuel. Since the 1980s and 1990s, increased burnup rates for UOX fuel have resulted, for an identical rate of FP incorporation into glass, in a gradual rise in actinide content. This is the case, in particular, for curium (Cm), for which the rise with burnup rate is steepest.

MOX reprocessing, which is currently carried out at moderate intensity, depending on customer requirements, shows similar outcomes.

In the short term, Areva's industrial goal is to raise that limit, to stabilize the volume of vitrified waste produced per terawatt-hour, bearing in mind the considerable amounts of **UOX2** fuel to be

Standard compacted waste container (CSD-C). In like manner to fission product solutions, structural components from fuel assemblies are nowadays conditioned in line, during spent fuel reprocessing operations. One aim is to include in such packages non-metallic, or organic technological waste.



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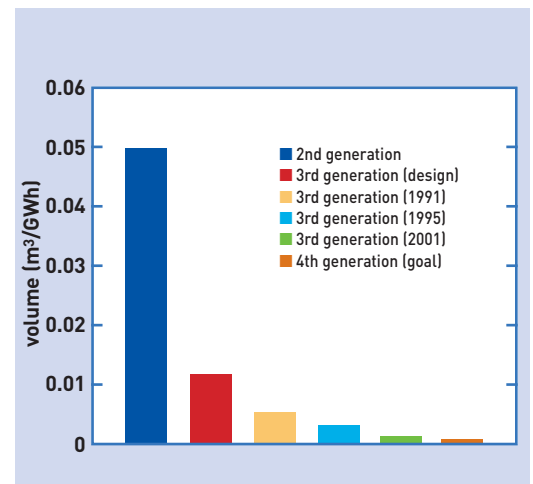


Figure 3. Evolution of volumes for HLW and ILW-LL waste, according to reactor generation (from M.-F. DEBREUILLE and J.-G. DEVEZEUX DE LAVERGNE, *Global 2003*).

reprocessed. This evolution is projected for a constant production technology, provided it is verified that long-term CSD-V behavior is not significantly impacted by higher alpha-emitter content. R&D findings supporting such an increase are already in hand, and a new specification is being finalized.

For the medium term, Areva is investigating further gradual increases in glass alpha-emitter content. Ongoing R&D programs are aimed at further increasing this, to allow reprocessing of **UOX3** fuel in optimum conditions. The research effort now initiated involves two main thrusts. The first one involves direct investigation of the effects of higher alpha-emitter incorporation rates, through the fabrication of ²⁴⁴CmO₂-**doped** glasses. The second thrust is concerned with understanding, at the microscopic level, the processes of alteration, and stabilization, of observed macroscopic properties, on the basis of characterization findings for doped glasses, and findings from work on external irradiation, and from numerical **modeling**. The most

recent findings warrant, at this point in time, a degree of confidence in a successful outcome for this coming stage.

Evolution of the CSD-C “in line”

The current industrial goal is that of following through the coming up to full design production rate of the ACC workshop. This challenge is well on its way to being met,

Standard vitrified waste container (CSD-V). Investigations are ongoing on such packages, to take on board fuel evolution, in particular increased burnup rates.



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and compaction of structural components in line with the shearing workshops is in effect as of now. A further goal, as for CSD-Vs, is that of supporting the increase in burnup rates, beyond 45 GWd/tU, in reprocessed fuel, by guaranteeing full package control, with regard to package safety and content traceability. A third aim consists in opening up the field of CSD-C production, to include ever larger amounts of non-metallic technological waste, with a view to achieve further reductions yet in overall fabricated package volume. Indeed, cementation results in larger package flows, by a ratio of up to 10, relative to CSD-C compaction. The investigations initiated are thus targeted at achieving, ultimately, an annual production of only a few tens of cemented ILW-LL packages. Ongoing programs are being carried out at various levels: R&D on the behavior of technological waste materials in CSD packages, qualification of measurement methods, deployment of methods for the determination of the constituents of technological waste materials, together with the associated traceability.

Evolutions in the conditioning processes used

The directions of advance outlined above, in some cases, go beyond improvements in existing processes, or an understanding of package behavior. Both the future evolutions in fuel and the shutdown of installations in plants of the previous generation (rinsing, decontaminations), or the retrieval and conditioning of legacy waste, mean careful examination should be made of the benefits and drawbacks of strategies involving use of current techniques, or development of new techniques. The general approach is to favor deployment of conditionings in existing workshops. This approach, indeed, has many advantages, since workshops do not have to be built, the techniques are proven, lead times can be tightened, and costs are lower, since the facility has been amortized. Projected programs at the La Hague plant, concerning retrieval of legacy waste, are described in the paper *Nuclear waste management and processing: between legacy and anticipation*.



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View of the outside of the extension to the E-EV-SE vitrified waste storage workshop, at Areva's spent fuel reprocessing plant at La Hague.



Hall of the E-EV-SE facility, where standard vitrified waste containers are stored. Designed on a modular basis, its current capacity stands at 4,320 CSD-Vs. This, however, may be increased by a factor 8, through construction of extra modules.



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In sum, advances should allow near-stabilization of the volumes of HLW waste produced per tonne of spent fuel, in spite of constantly rising burnup rates. This remains an ambitious goal. Further out than a few decades, with the new generation of reprocessing plants, conceivably further volume reductions may be achieved, if that option is taken up, as shown in Figure 3.

Influence of the thinking on future management pathways for HLW and ILW-LL waste

Under the aegis of the Act of 1991 on research on HLW waste, two types of installations, seen as constituent parts of the management solutions being considered, have been the subject of particular investigation, for different purposes: *long-term storage*, and *disposal in deep geological layers* (see Box D, *From storage to disposal*).

One of the major findings arrived at, in the framework defined by the Act, is to show that these management modes, possibly combined with others, are feasible. However, this approach does not allow, at this stage, specifying management pathways that are fully optimized, in the industrial sense. In particular, the inventories being used consist in detailed descriptions of packages produced, or to be produ-

ced, on the lines of previous practice, i.e. in line with the prevailing principles of the past two or three decades (which should not significantly alter for the near future). However, depending on decisions soon to be made under the provisions of the Act, it will gradually become possible to redirect package conditioning, to tend towards such an optimization.

Paths that may be anticipated include, for example:

- fuller knowledge of the inventory of **radionuclides** contained in packages, as and when required, with regard to coming specifications for the design of disposal (or even storage) facilities;
- the continuation, at a level to be specified, of work on long-term package behavior, with possible consequences for conditionings;
- standardization of cemented packages (for legacy waste, in particular), and, more broadly, optimized allocation of functions between waste, matrix (where applicable), canister, overpack or container (as applicable), **engineered barrier** (when applicable), and the geological barrier, in the disposal case.

Such programs should, for a large part, be in a position to achieve results over the coming two decades. Further out, decisions such as deployment of certain forms of advanced partitioning, linked to the fourth generation of reactors, could be taken. These decisions could be based on considerations of optimized disposal costs, in relation to package thermal load. Waste packages would then undergo more or less profound transformations, with regard both to their form (with a new generation of reprocessing plants, and hence of conditioning processes), and content.

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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 **fusion 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 TWh 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 235** (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

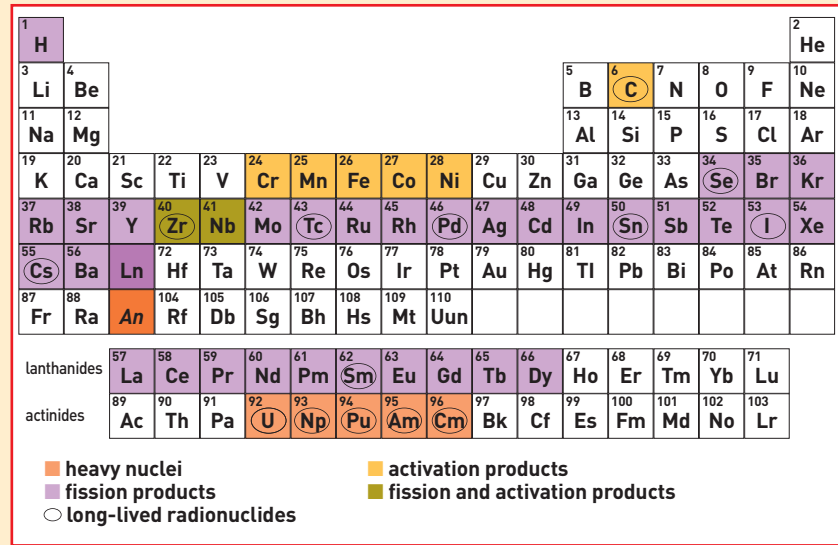


Figure 1. The main elements found in spent nuclear fuel.

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

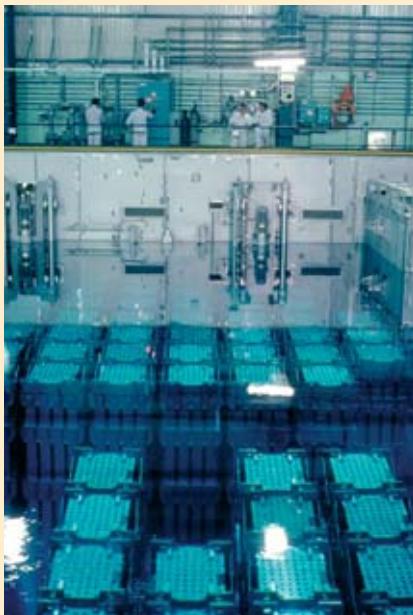
Table 1. Major actinide inventory for spent UOX and MOX fuel after 3 years' cooling, for a variety of enrichment and burnup rates. Burnup rate and quantity are expressed per tonne of initial uranium (tiU) for UOX, per tonne of initial heavy metal (tihm) for MOX.

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

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

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 mode), or to a so-called **open cycle** policy, whereby spent fuel is considered to be waste, and designated for **conditioning** into **containers**, and **disposal** as-is.

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

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).

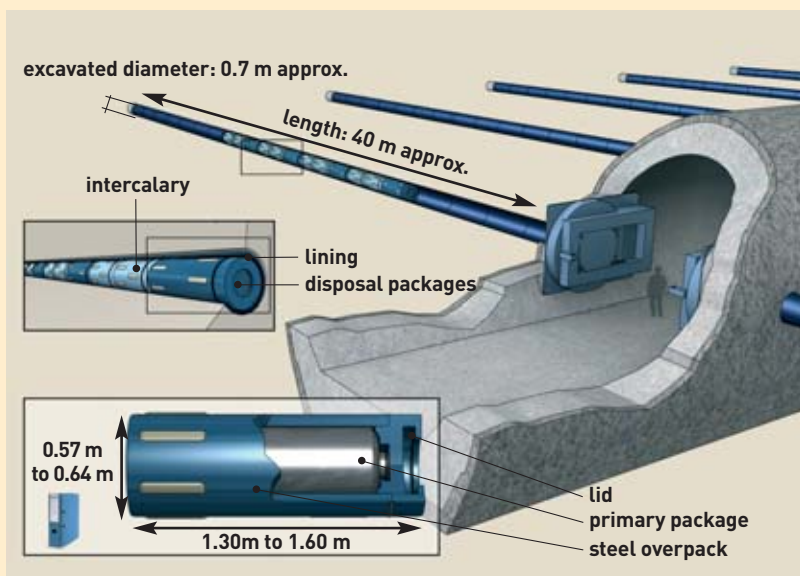
Long-term storage (LTS) may be contemplated, in particular, in the event of the deferred deployment of a disposal facility, or of reactors to carry out

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.

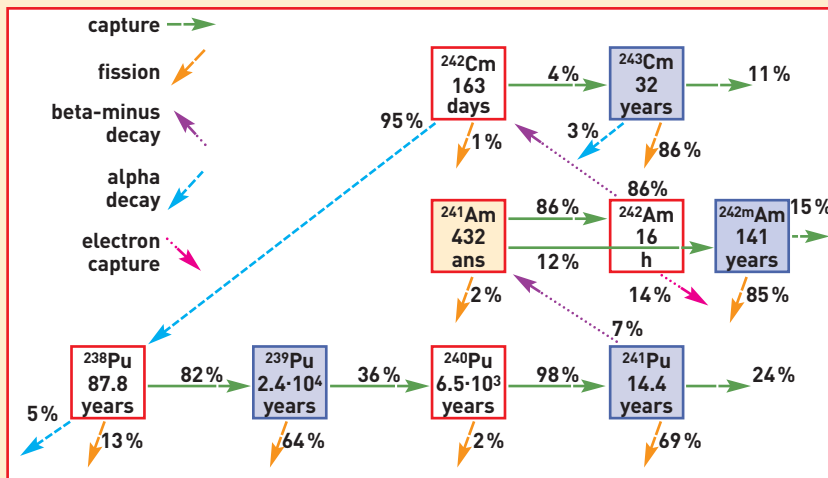


Figure.

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.

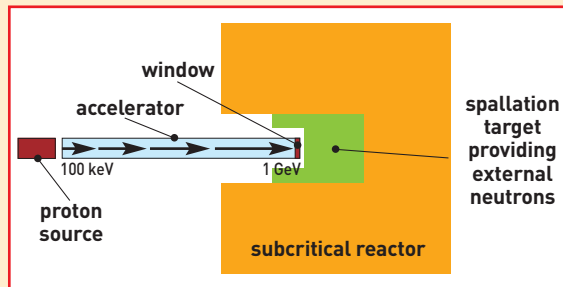
F What is an ADS?

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

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.