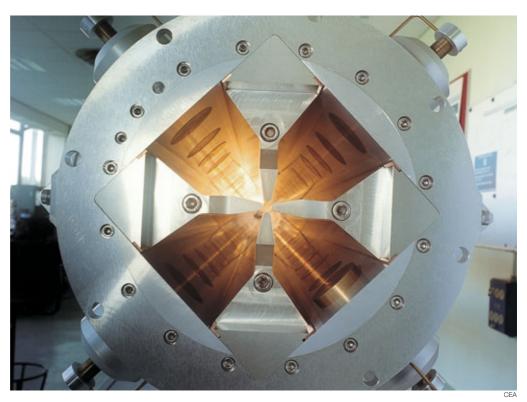
FROM THE CRITICAL REACTOR TO THE SUBCRITICAL HYBRID SYSTEM: TRANSMUTATION TOOLS

Before addressing the problem of minor actinides and long-lived fission products, the plutonium problem must be settled. This energy-rich raw material must go through a "multirecycling" process, and its stocks managed in "just-in-time" mode, to avoid build-up. The next step is to transform the long-lived radioactive nuclei found in nuclear waste into short-lived or even stable products. We now know that this is feasible. Transmutation must be performed in facilities where waste generation is already at a minimum level in order to stabilize stocks of final radioactive waste. Various methods, that may be combined in various ways, have been studied, ranging from the use of existing reactors to that of dedicated hybrid systems, combining a particle accelerator with a subcritical reactor. Assessing the latter method meant learning more about a particular nuclear reaction, known as spallation, which produces neutrons to drive a transmutation reactor.

Full-scale model of the RFQ, a radiofrequency quadripole device, designed to accelerate and focus particles emitted by Silhi, the high-intensity, light-ion source of the Iphi (High-intensity proton injector) prototype, developed by CEA and CNRS for design work on proton accelerators upstream from future hybrid systems.



Transmutation possibilities

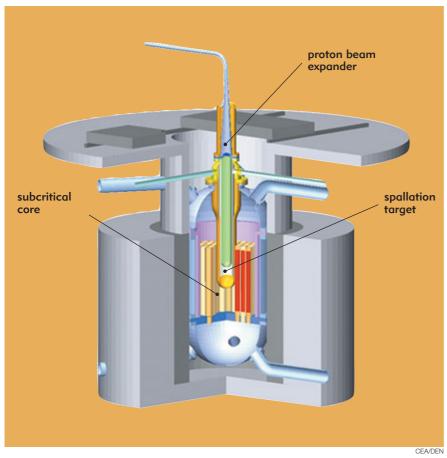
The **transmutation** process, a term covering all transformations of nuclei induced by nuclear reactions in reactors, is the focus of considerable research activity, seeking to transform **long-lived radioactive** nuclei found in spent **fuel** into shorter-lived elements, by **recycling** them in power reactors or in certain specific systems. Research teams have already demonstrated that trans-

mutation shows considerable potential as a means to minimizing the possible long-term impact of final waste on human beings and the environment (see *Transmutation is technically feasible*).

Of the different elements found in spent fuel, actinides (plutonium, neptunium, americium, and curium) account for most of the potential harmful effects in terms of radiotoxic inventory. Long-lived fission products (principally technetium-99, iodine-129, and cesium-135) must also be taken into

consideration. Although they only account for a minute fraction of this inventory (approximately 10⁻⁵), they are more soluble and therefore tend to move about more in various geological environments. In the very long term, when located in the vicinity of a disposal site, they can thus represent a source of residual toxicity in the same way as **heavy nuclei** (see box C, *Radiotoxicity of spent fuel*). Transmutation is aimed at reducing as far as possible potential and residual detrimental effects relating to reactor operation,

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Schematic view of a hybrid system demonstrator dedicated to incineration. It combines a gas-cooled fast-neutron subcritical reactor, and a high-energy proton accelerator (not shown) whose beam arrives at the upper part of the reactor.

while minimizing the quantities of actinides and long-lived fission products liable to be ultimately placed in **disposal** sites.

Stabilizing the plutonium inventory

Transmutation involves putting the elements concerned through a preliminary advanced separation process (see chapter I). The effectiveness of this step determines the residual quantities in the waste packages. It then involves a multiple-recycling (multirecycling) operation, which is meant to stabilize the inventories in recycled elements, starting with plutonium.

As plutonium accounts for about 90% of the radiotoxic inventory of spent fuel, consuming it, especially in today's reactors, takes priority over any strategy aimed at transmuting minor actinides, which only account for 10% of this inventory. For this reason, preference will be given to those plutonium-recycling strategies that minimize the production of minor actinides.

As regards long-lived fission products, iodine-129 transmutation is complicated by the low probability of neutron **capture** on this **isotope**, and the very long residence time in the reactor that would be required (currently at least 15 years simply to reduce it by half). On the other hand, effective transmutation of cesium-135, an isotope representing only 10% of fission products, requires

a preliminary isotope-separation stage, at least to eliminate the 133 and 134 isotopes which, when exposed to neutron flux, lead back to the formation of the 135 isotope. In order to reduce the harmful effects of these two fission products, it might be preferable in practice to adopt a strategy of separation, followed by **conditioning** to ensure **containment** for very long periods of time, rather than transmutation.

Fast spectra preferred for minor actinides

Transmuting long-lived radioisotopes is an additional constraint for the fuel cycle. Its impact in terms of the complexity of the processes used and cost should therefore be limited. A common indicator can be used to search for the best strategy: the quantity of material transmuted per unit energy produced. This indicator takes into account the intensity and spectrum of the neutron flux and the physical possibility of loading the material to be transmuted in the reactor, as well as the neutron balance aspect, which shows whether or not the fuel must be further enriched with fissile isotopes to offset the additional absorption caused by the products to be transmuted.

This indicator alone is not enough, however, and other aspects must be taken into consideration, such as additional requirements imposed on the fuel cycle, including the need to reinforce radiation-protection measures for making and reprocessing the fuel and the capsules (or "targets") carrying the material to be transmuted.

For minor actinides, the goal is to promote fission rather than neutron capture. There are three reasons for this: to convert the initial nucleus into relatively short-lived fission products, to limit the negative impact on neutron balance (table, p. 43), and to limit the chain production of minor actinides that to varying degrees are more toxic than the initial nucleus. This is why preference is given to **fast neutron** spectra.

For long-lived fission products, which can only be transmuted by neutron capture, intense neutron fluxes are preferred, wherever possible in the **epithermal** and **thermal** energy range, where the probability of neutron capture is highest.

Double strata or double component?

In addition to the various options distinguished at reactor level, others exist at system level, depending on the role played by facilities wholly or partially devoted to transmutation operations. The strategies considered (see *More than one way to control nuclear waste upstream*) involve existing PWRs, other reactors based on "mature" technologies, such as fast-neutron reactors (FNRs) and, in the longer term, gas-cooled

When designing a spallation target for this type of system, simulation programs are used to optimize performance, particularly in terms of neutron production, and to assess the constraints on the materials making up the target and the surrounding structures. For example, it is necessary to predict what different nuclei will be produced by spallation reactions in the target and in the interface separating it from the vacuum of the accelerator, generally consisting in fine tungsten or steel foil. These nuclei can in fact modify the chemical composition of the material, possibly leading to a risk of embrittlement or, in a liquid target, corrosion. Furthermore, as many of these nuclei are radioactive, estimations are required of how the activity of spallation-module materials evolves with time. This type of information is important, for example, for predicting the time before maintenance or for ensuring long-term management of the various components.

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Figure B. Productionrate measurement of the many nuclei produced in a spallation experiment, using a 1 GeV per nucleon, lead-208 beam. The empty squares represent stable nuclei.

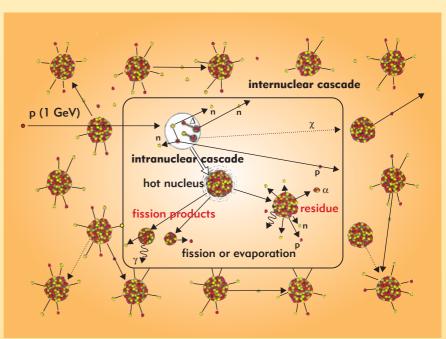
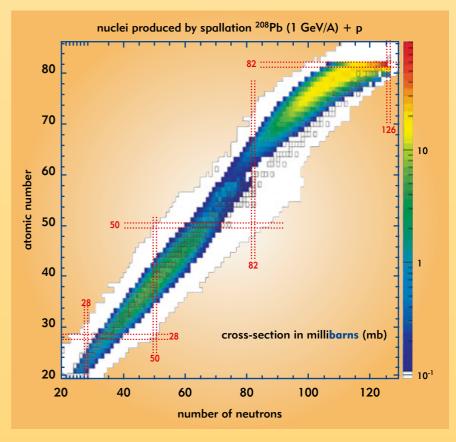


Figure A. Intranuclear cascade.

Before using a simulation program, its reliability must first be guaranteed and its accuracy estimated. This type of computer codes describes the elementary interaction between a nucleon and a target nucleus using nuclear-physics models that generally present spallation as a two-step process. The first step is fast, consisting of a sequence of collisions between nucleons, like billiard balls hitting each other. This is called the intranuclear cascade (figure A). At the end of this process, the incident energy is distributed throughout all the nucleons of the nucleus, which is now in



an excited state. The second step, which is much slower, is *de-excitation*, leading to the emission of low-energy particles or, in some cases, to fission into two smaller nuclei. It is first of all essential to check the quality of these models by comparing the predictions they give with experimental results.

The Nuclear Physics Department of CEA, together with other French and European laboratories, is conducting experiments into the elementary spallation mechanism. This work has included measuring the nuclei produced by spallation, using the reverse-kinematics method, at the GSI accelerator in Darmstadt, Germany. This method involves the use of a heavy-ion beam to bombard a liquid-hydrogen proton target. Its advantage is that the nuclei produced have a strong forward focus owing to the entrainment speed. This makes it easy to identify them using a series of magnets and detectors (figure B).

The experiments carried out have revealed serious shortcomings in some of the models used in simulation codes. At the same time, work is under way with theorists to improve these models, of which the new versions are far more consistent with experimental data.

A spallation target made from liquid leadbismuth, a material often considered for hybrid-system targets, and capable of withstanding a 1 MW beam, is also being made as part of the Megapie collaborative project. Set up as the target for the Paul Scherrer Institute SINQ neutron source in Zurich, Switzerland, it will be used to perform fullscale studies of material problems.

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radionuclide	thermal spectrum	fast spectrum
²³⁷ Np	0.9	- 0.6
²⁴¹ Am	0.1	- 0.6
²⁴⁵ Cm	- 2.3	- 2.7
⁷⁹ Se	2.1	2.1
⁹⁹ Tc	1	1.1
126Sn	1.1	1.2
129Г	1	1
135Cs	1	1

Table. Net neutron balance associated with transmutation of a radionuclide. A value of 1 represents a single neutron capture. Higher values indicate spurious captures induced in the daughter products of the radionuclide in the flux. Negative values indicate fission reactions leading to a neutrongenerating net balance.

fast-neutron reactors (GCRs), or hybrid systems combining a subcritical core and an accelerator-driven neutron source, and using spallation reactors (see box D, *Spallation*, box E, *What is a hybrid system?*, and box F, *PWR*, *FNR*, *and GCR*).

These strategies are built around single or double strata scenarios, according to whether the radionuclides to be transmuted are recycled at the same time as plutonium in existing nuclear facilities (single stratum) or in specific facilities (FNR or hybrid systems). Depending on the scenario adopted, plutonium recycling can be carried out in a single reactor system (e.g. PWR) or in two complementary reactor systems (PWR and FNR) from the first "stratum". The second option is referred to as a "double-component" strategy.

Transmutation could be carried out either in a minimum number (5 to 10%) of dedicated systems concentrating all the materials to be transmuted that are difficult to recycle (americium, curium, and long-lived fission products) in "double strata mode", or in an additional component to the existing PWR plants, accounting for 20-30% of electrical generating capacity, where all the actinides and long-lived fission products are recycled at the same time ("double-component" transmutation). In principle, subcritical systems offer more flexibility for the first approach, because in theory they can be used to manage cores with a high minor-actinide load (approx. 50%), whose neutron characteristics would be incompatible with stable operation in critical mode.

Homogeneous or heterogeneous process?

Various options are also available at the fuel level. How should the elements to be

transmuted, minor actinides and long-lived fission products, be added to the fuel? They can be arranged in homogeneous or heterogeneous mode. In the former case, the products to be transmuted are mixed with standard fuel. In the second case, however, they are separated out and placed in specific capsules (or "targets") that house all the products to be transmuted, while the core is only partly made up of the usual UO₂ or MOX fuel rods, containing no products to be transmuted.

The heterogeneous mode offers the advantage of reprocessing fuel and targets separately, with the result that the targets can go through the reactor once only (monorecycling) or several times (multirecycling), if their radiation resistance allows, or if the products to be transmuted are suitably reconditioned.

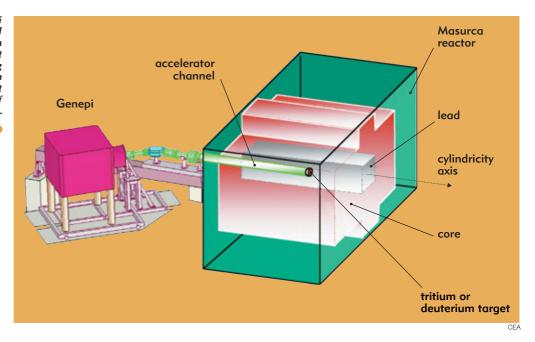
Homogeneous mode for neptunium

For neptunium, recycling is always envisaged in homogeneous mode in plutonium fuel. Americium and curium, on the other hand, can be recycled either in homogeneous mode, or in specific targets to minimize the need for fuel-fabrication operations calling for highly reinforced radiation-protection measures owing to the intense gamma and neutron emissions from these actinides. The targets can be optimized to burn 90% of minor actinides in one pass or to be prepared for further, more advanced multiple-recycling transmutation at a later stage.

Multirecycled targets for fission products

Fission products can only be transmuted as multirecycled targets, either in a PWR, within the limits imposed by the neutron bal-

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ance, or in fast-neutron systems, in conjunction with materials that slow down neutrons (ZrH₂ or CaH₂). The advantage of the second scheme is that it exploits the intense neutron fluxes of such systems, and the neutron energy levels found there are closer to those favorable to neutron capture on these fission products.

Other options, such as α thorium support for actinide transmutation, or boiling-water reactors, which combine a thermal neutron spectrum at the bottom of the core and a fast spectrum at the top, could also be contemplated. Thorium, which drastically reduces the production of neptunium, americium, and curium, does not offer any advantage over inert-matrix transmutation. Furthermore, boiling-water reactors do not have any decisive edge over PWRs in the thermal spectrum or fast-neutron systems, that would

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argue in favor of their deployment as a new system in France, unless further justification is found

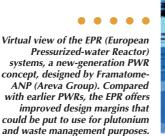
Overview of initial results

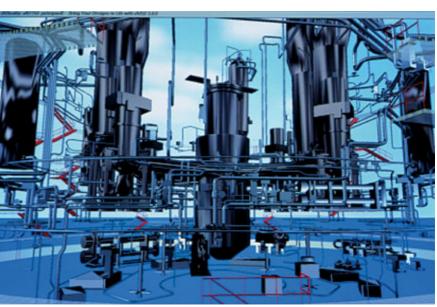
Examination of the various scenarios presented shows that multiple recycling of plutonium is essential. Implementing this process increases the production of minor actinides, which limits the reduction factor. Recycling is necessary to reduce the radiotoxic inventory even further. As recycling neptunium is only beneficial in the very long term, priority must be given to studying americium and curium recycling.

Multiple recycling of plutonium on a uranium support can be achieved in fast neutron reactors and contemplated in PWRs (at least a few other recycling operations with the development of specific fuel assemblies using the Corail concept) (box). Recycling plutonium on an inert matrix, for increased consumption, is an area in which research work is concentrating on the APA/Duplex concept (box).

An FNR component for multiple recycling?

Multiple recycling of minor actinides and certain long-lived fission products can, together with that of plutonium, be envisaged in a "component" made up of fast neutron reactors added to existing PWRs operating on uranium fuel. This component would represent about 25% of all plants. Exploratory studies are focusing on the ability of PWRs to transmute some of these long-lived radionuclides, americium and techne-





Framatome-ANF

Controlling the plutonium inventory in a PWR

Plutonium control is aimed at keeping plutonium in the **fuel** cycle, while stabilizing the overall inventory associated with the operation of nuclear reactors. These objectives are built around a **"reprocessing-recycling"** strategy. They also imply the development of advanced fuels and suitable reprocessing methods, allowing multiple recycling of the plutonium contained in spent fuel, under technologically and economically acceptable conditions.

Monorecycling of plutonium, as practiced today in the 30%-MOX cores of the PWR 900 series, reduces by about 30% the amount of plutonium produced every year by nuclear power plants (some ten metric tons). This means that monorecycling plutonium alone cannot stabilize its inventory. Unless some specific action is taken, 670 metric tons of plutonium will have built up in France by 2070. Multirecycling plutonium in PWRs in MOX assemblies would soon come up against such safety constraints that the objective could not be met.

Several solutions have been thought up to overcome this problem. The first solution, known as MIX, involves replacing the **depleted uranium** of the MOX rod with **enriched uranium**, to obtain more homogeneous neutronic behavior. A second method is to abandon the all-MOX assembly concept in favor of the Corail solution, where uranium-oxide (UOX) and MOX fuel rods are placed in carefully calculated positions within the assembly.

From the MOX to the APA assembly

The third solution is far more ambitious. It entails using UOX rods and plutonium rods, with no uranium, while increasing the cross-section of the **coolant** channel. Increasing the quantity of coolant in the assembly in this way leads to reactor operating parameters that are closer to those encountered in uranium cores, which is not the case of MOX assemblies. This "Advanced Pluto-

nium Assembly", or APA, would thus improve plutonium combustion while increasing neutron **moderation**.

In the first solution, all reactors are loaded with MIX assemblies, and all fuel rods contain mixed plutonium and uranium oxides. Corail, the second solution, also applies to all the reactors, but each fuel assembly only contains 30% of MOX rods. In the third solution, 30% of reactors in the system are loaded with APAs, with each assembly containing 25–50% plutonium rods.

All fuel rods containing plutonium in a Corail assembly or APA are identical as regards enrichment. This means that there is no assembly "zoning" as in MOX assemblies. The Corail solution requires no new technological developments, whereas the APA calls for a new fuel rod and adjustments to assembly design. The APA plutonium rod could be a **cermet** metallurgically alloyed with the cladding, making it a particularly attractive "cold" fuel. Reprocessing this plutonium rod would require adjustments to the process currently used.

In a nuclear power-plant system currently generating 400 TWh, the MIX and APA solutions would lead to an inventory of 230 metric tons, i.e. a reduction by a factor of 3 by the year 2070, compared with the current monorecycling scenario. The Corail solution would stabilize the inventory at around 400 metric tons, thus reducing the mass of plutonium by 40% by the same date. A system based on multirecycling plutonium with APAs would not generate any more waste, either in terms of mass or radiotoxicity, than the current method of storing MOX fuel, which produces americium through the decay of plutonium-241. Lastly, the APA would save around 20% of uranium (natural and enriched) throughout the PWR system.

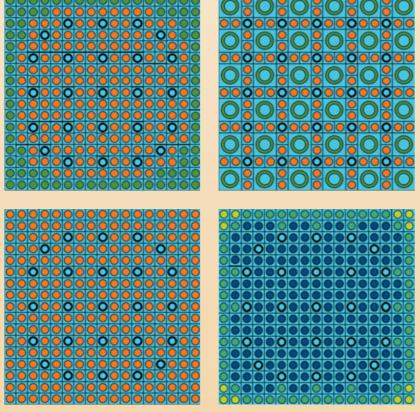


Figure. Cross-sectional views of a fuel assembly presenting various plutonium (Pu) consumption configurations in a pressurized-water reactor. In the Corail configuration (top left), 84 fuel rods (shown in green) contain plutonium on a depleted-uranium support. In the APA concept (top right), the Pu is contained in 36 annular rods on an inert matrix. In the MIX concept, as in standard fuel (bottom left), all the rods are identical, whereas MOX (bottom right) features three different enrichment zones.

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End of a shipping cask for a fuel element containing an americium capsule for the Ecrix transmutation

experiment in the Phénix reactor. The capsule is an example of a specific target carrying material to be transmuted.



tium-99, in particular by adapting APA/Duplex assemblies.

Transmuting a concentrated flux of minor actinides (about 50%) and certain long-lived fission products in a second "stratum" of the cycle, representing only 5% of electrical generating capacity, is hard to achieve in conventional reactors, because the neutron characteristics are not suited to stable operation in critical mode (delayed neutrons fraction, Doppler effect, etc.), unless very small models are designed, in the region of 150 thermal megawatts. Subcritical systems, which rule out these risks of instability by design, can be considered for "double-strata"

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schemes, provided that special steps are taken to guarantee subcriticality in all normal and accident operating states. This requirement is of major concern for systems designed with only a 2% subcriticality margin (approximate multiplication factor $k_{\rm eff} \sim 0.98)$ to restrict accelerator performance specifications to realistic levels.

The desire for subcritical cores with neutron properties that simplify the management of any critical accident situations, and the difficulties encountered in implementing fuels containing high concentrations of minor actinides are some of the reasons why transmutation of minor actinides should be envisaged in fuels with a higher plutonium concentration as part of an "extended doublestrata" scheme, representing 10-15% of nuclear facilities in the country. The contribution of subcritical systems to this type of transmutation strategy, and comparison with the "double-component" approach, remain to be analyzed.

Minimizing long-lived waste

The study of scenarios, going hand in hand with programs to develop computation schemes, and of fuel technologies required for the core configurations considered, is

What is a hybrid system?

A hybrid system consists in a nuclear reactor operating in subcritical mode, meaning that it is incapable of sustaining a fission chain reaction on its own, and an external neutron source capable of providing the required complement(1).

In the scenario preferred by researchers, this source is fed by a high-energy proton **accelerator**. The protons strike a heavymetal (e.g. tungsten) target, and generate

an intense neutron flux *via* a **spallation** reaction (see box D, Spallation).

The accelerator can be a linear accelerator or a cyclotron. As for the actual reactor, most hybrid-system projects make use of **fast-neutron** subcritical neutron-multiplication media, as fast neutrons offer the most favorable neutron balances for **transmutation**, an operation used not only to "burn" waste, but also produce new fissile nuclei, if requi-

red. A hybrid system, which by its principle offers a very wide margin with respect to reactivity accidents, could also be used to produce energy, even if some of this must be reserved to power the proton accelerator. However, the extra investment required for the accelerator means that these systems would be unable to compete with reactors. For this reason, they are best suited to specific applications, such as transmutation.

(1) See Clefs CEA No. 37, p. 14.

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PWR, FNR, and GCR

Three main nuclear-reactor systems (independently of hybrid systems; see box E, *What is a hybrid system?*) are currently being considered in studies of plutonium and waste management scenarios. The three reactor systems are: PWRs, FNRs, and GCRs.

PWRs (pressurized-water reactors), which form the foundation of the French nuclear power-plant system, belong to the "slow" or "thermal" neutron reactor family ("thermal" means in thermal equilibrium with the reactor material in which they are diffused). Water is used as the moderator to slow down these neutrons. The water is maintained at high pressure in the primary system to prevent it from boiling, and also serves as a coolant, transferring the heat from the core to a secondary system via an exchanger. It is this secondary system that supplies steam to the turbine generators producing electrical power. PWRs are members of the large family of light-water reactors that also includes boiling-water reactors

In FNRs (fast-neutron reactors), most fission reactions are generated by neutrons with energy levels of the same order of magnitude as when they were produced by fission. These reactors exploit the improved efficiency, in terms of fission, of neutrons that maintain the speed acquired from previous fissions. Sometimes loosely referred to as "fast"

reactors, they accept a wider variety of fuel isotopes than PWRs at the cost of a higher neutron flux in the reactor, and a high concentration of fissile isotopes in the fuel. This family includes the Phénix (currently operating at low power) and Superphénix (decommissioned) experimental reactors. In the absence of water, which would act as a **moderator**, and designed with a high power density, these reactors require an effective coolant. Sodium is currently used, but other liquid metals can be considered, or even gas with a lower power density.

The GCRs (gas-cooled reactors) considered today form a range of high-temperature reactors (850 °C) with a high energy efficiency, capable of adapting to a wide variety of applications and fuel cycles. The "hardening" of their neutron spectrum in favor of fast neutrons makes it possible to optimize the use of fissile and fertile materials, while minimizing the net production of long-lived waste through *in situ* incineration.

The GCR development plan provides for a technological design and development reactor at the Cadarache site by 2012. The vocation of this experimental reactor will be to validate, among other technologies, all fuels required for this range of reactors, focusing particularly on the validation of a first generation of fuel for fast-neutron GCRs.



Concept for a direct-cycle, helium-cooled, high-temperature reactor.

directing research work along the two main lines most likely to lead to concrete applications. The first concerns a strategy for plutonium control and transmutation that could be decided upon after 2006, to optimize the contribution of PWRs in reducing to a minimum the quantity of long-lived radionuclides in their own waste. The second focuses on the study of future nuclear systems capable, according to the terms of the Interministerial Committee of June 1, 1999, of "minimizing the production of long-lived radioactive waste"; to do this, these systems should be given the same transmutation capabilities as dedicated **incinerators**.

CEA is actively involved in both these lines of research. In the first case, it is imagining ways to go from the current method of monorecycling plutonium in PWR 900 reactors (using MOX fuel), to multiple recycling in Corail or APA/Duplex assemblies in a new-generation series of PWRs (EPR), coming into service as of 2015, and simultaneously proceeding with the advanced separation of minor actinides for storage until

effective transmutation technologies are developed for them. Compared with previous PWRs, the EPR offers design advantages that could be exploited for plutonium and waste management. For example, PWRs running on 36% APA/Duplex fuel could be used, with a multiple-recycling strategy, to stabilize the plutonium and americium inventories associated with their operation, while limiting the build-up of curium inherent in actinide recycling in PWRs.

Vitrified FPs: the only remaining final waste?

This strategy means that the final waste from PWRs would virtually be limited to vitrified fission products alone. Potential radiotoxicity would be reduced by a factor of 100 compared with that of spent fuel as is, and would be brought down to a level comparable with that of initial uranium ore after a few centuries, instead of a few hundred thousand years.

CEA is launching an R&D program on future nuclear systems, focusing on technologies that, in theory, appear most likely to reconcile the need to minimize long-lived waste, and the progress required in areas such as economic competitiveness, safety, and optimum use of available fissile and fertile fuel resources.

The extensive common core between this R&D objective and research into dedicated transmutation systems could lead to common developments in an experimental reactor scheduled for 2012 at the Cadarache center. Furthermore, the first generation of future systems, to be deployed after 2035, and intrinsically designed for transmutation, could adopt a "double-component" approach to burn separated and stored minor actinides from PWRs, while minimizing their own production of long-lived waste.

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