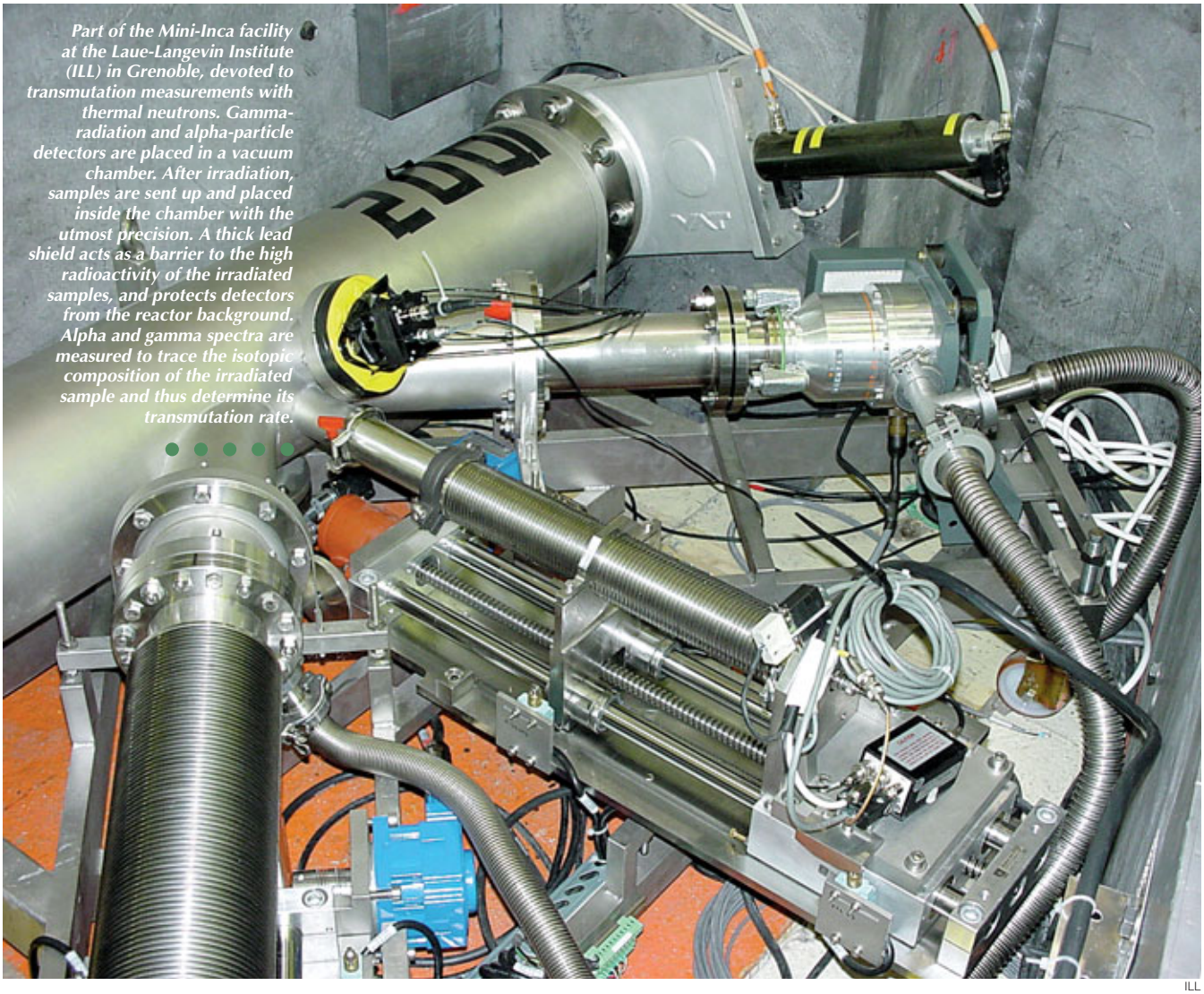
# TRANSMUTATION IS TECHNICALLY FEASIBLE

*After sorting the waste, and extracting from it the long-lived elements making up most of the radiotoxic inventory, or presenting the greatest chemical mobility, the most appealing solution would be to eliminate them by transmutation, recycling them in nuclear reactors. Although the theory of this operation had been known for a long time, there was no proof that the operation was technically feasible in existing or new, purpose-designed facilities. This proof now exists in some areas, while experiments are still in progress in other areas at the European level.*



*Part of the Mini-Inca facility at the Laue-Langevin Institute (ILL) in Grenoble, devoted to transmutation measurements with thermal neutrons. Gamma-radiation and alpha-particle detectors are placed in a vacuum chamber. After irradiation, samples are sent up and placed inside the chamber with the utmost precision. A thick lead shield acts as a barrier to the high radioactivity of the irradiated samples, and protects detectors from the reactor background. Alpha and gamma spectra are measured to trace the isotopic composition of the irradiated sample and thus determine its transmutation rate.*

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## Transmutation: the basic physics

The principle of **transmutation** as applied to nuclear waste reprocessing consists in modifying the nuclei of **long-lived** elements to transform the **isotopes** concerned into products that are stable, or with a much shorter lifetime, or with lower **radiotoxicity** (see box C, ***Radiotoxicity of spent fuel***).

This operation has the potential to achieve in the long term significant reductions in the quantity and toxicity of the final waste generated by nuclear energy, and represents one of the key areas of research in the management of high-level, long-lived waste from existing reactors (see *Radioactive waste management research*, box 1). But how can this "philosophers' stone" be implemented in practice? By bombarding "target" nuclei with

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*Concept of a hybrid reactor capable of transmuting nuclear waste (the inset shows a close-up of the core).*

elementary particles. Several different methods have been explored, based on the use, respectively, of photons, protons, and neutrons.

## The photon method: lacking in efficiency

By bombarding a target with an electron beam from an accelerator, photons can be obtained by *Bremsstrahlung*(1) that are capable of triggering (γ,*x*n) reactions, meaning that due to the effect of incident **gamma** radiation, *x* neutrons are ejected from the nucleus. When applied to materials that are too neutron-rich and, for this reason, unstable, such as certain **fission products** (strontium-90, cesium-137, etc.), these reactions generally lead to stable nuclei. Nevertheless, in view of their very poor efficiency and the very high intensity electron stream required – currently impossible to achieve – the photon method has not been adopted.

## One possible solution: protons and spallation

The interaction between the nucleus and proton induces a complex reaction known as **spallation** (see box D, ***Spallation***), which causes the nucleus to fragment, releasing a number of particles, including high-energy neutrons. Transmutation based on direct interaction with protons is not cost-effective, as it requires very high-energy photons (1–2 GeV) to overcome the Coulomb barrier(2), and the production energy required is greater

(1) German term adopted in English, literally meaning "braking radiation": High-energy photon radiation generated by accelerated (or decelerated) particles (electrons) describing a circular trajectory, tangentially emitting braking photons; those with the highest energy level are emitted in the axis of the electron beam.
(2) Force of repulsion that tends to prevent electrical charges of the same sign from approaching each other.

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*Titanium sample-holder (7 x 7 cm approx.), used as part of the Mini-Inca experiment to irradiate americium contained in two quartz vials, placed inside two lateral graphite tubes (shown in black) in the ILL's high-flux reactor in Grenoble. A natural iron sample is placed inside the central tube to measure the neutron flux integrated during irradiation.*

than that recovered during the process generating the waste. Indirect transmutation, however, using high-energy neutrons (of which about thirty are produced, depending on the type of target and the energy of the incident proton), can significantly improve performance. This is the process adopted for "hybrid reactor" concepts combining a **subcritical** core and a high-intensity proton accelerator (see box E, ***What is a hybrid system?***).

## Neutrons: the best choice

As they have no electric charge, neutrons are by far the most suitable particles given the required criteria. Furthermore, they are "naturally" present and available in large quantities in nuclear reactors where physicists use them to induce **fission** reactions and thereby produce energy. In this respect, it is interesting to observe that the best way to **recycle** waste is to reinject it into the facility that created it. In order to obtain successful nuclear fission, it is important to know the probabilities of interaction between the neutron and the material. These probabilities are referred to as neutron **cross-sections**. Their values vary enormously from one nucleus to another and even more so from one isotope to another within the same nucleus. Large-scale effort on the international level has led to the creation of extremely comprehensive libraries using measurements performed in many experimental facilities. Nuclear data must now be added to and refined to optimize the performance of systems intended for transmutation, especially for the most innovative among them (box 1).

## Capture and fission

The interaction between neutron and nucleus leads to two mains types of reaction: **capture** of the neutron by the target nucleus, and nuclear fission. For fission products, the former type of reaction usually leads to the generation of a stable product after a succession of transformations. The typical example of this is technetium-99 (**half-life** of 210,000 years) that, following neutron capture, is transformed into technetium-100 with a very short half-life (15.8 seconds), leading on to stable ruthenium-100 following $\beta^-$ **disintegration** (box 2). This reaction must be minimized in

## Basic data requirements for transmutation

**1**

As from the 1980s, as more and more data came to light regarding the most important nuclei for civilian and military nuclear applications, the related experimental facilities were closed and nuclear data-evaluation activities reduced.

New projects are now under way in France and other countries to obtain nuclear data to enhance the precision of computer simulations of innovative systems for nuclear-waste **transmutation**. The neutronic properties of such systems are significantly influenced by the massive quantities of **isotopes** – whether contained in fresh **fuel** or during irradiation – that play only a minor role in the conventional fuel cycle. The available nuclear data for rare isotopes are usually obtained from theoretical evaluations, which are often arrived at without experimental data. Even if they are reliable enough to ensure simulations of the behavior of existing **thermal** and **fast** reactors, the same cannot be said for innovative systems devoted to transmutation.

Two types of **cross-section** measurements are available for experiments: differential and integral(1). For the first type, which takes several weeks and requires large quantities (between 1 mg and several tens of grams) of isotopically pure samples, CEA uses the best facility in the world at the European Union Joint Research Center in Geel, Belgium. The neutron cross-section of technetium-99 (box 2) and neptunium-237 have been measured there, and the same will soon be true for iodine-129. A new European project, n-TOF, currently under way at the CERN in Geneva, Switzerland, will complement these studies using a beam with a higher intensity, which will simplify measurements of **radioactive** isotopes. Several French laboratories, including CEA, were expecting initial measurements by the end of 2001.

As for integral measurements, which are the most widely used and required when the sample is not available in adequate quantities, two projects are in progress at CEA: Profil-2, an experiment in the Phénix "fast" reactor at Marcoule, and Mini-Inca, in the high-flux reactor at the Laue-Langevin Institute (ILL) in Grenoble. In the context of the second project, it will be possible to modulate the neutron spectrum, by positioning the sample at various distances from the fuel element. In this way, the ideal transmutation conditions can be determined for the isotopes that contribute the most to waste radiotoxicity.

Many uranium, plutonium, thorium, americium, curium, and neptunium isotopes, together with several fission products, must be studied in fast, **epithermal**, and thermal neutron fluxes. These experiments will not only provide cross-section values, but also transmutation rates in spectra similar to those of transmutation systems. These measurements can then be compared with calculations based on data libraries to evaluate their validity for transmutation studies.

(1) See "Measurement of Cross Sections" in *Clefs CEA* No. 45, p. 16.

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# Technetium-99, a fission product particularly suited to transmutation

Of the six elements at the top of the list for transmutation there are three fission products, including the technetium-99 isotope ($^{99}$Tc), of which large quantities are generated in nuclear reactors. This **beta-minus ($\beta^-$)** radiation emitter also has a long **half-life** (210,000 years). Following the **capture** of a neutron by $^{99}$Tc, yielding $^{100}$Tc, and the instantaneous deexcitation of the compound nucleus by **gamma ($\gamma$)** radiation emission, the $^{100}$Tc decays through $\beta^-$ emission with a half-life of 15.8 s to become ruthenium-100 ($^{100}$Ru), which is stable. This makes technetium well suited to **transmutation** (figure A). At the time the research work provided for by the 1991 Act began, not enough was known about $^{99}$Tc **cross-sections** to build a reliable model of its behavior in a future nuclear waste **incinerator** system. For this reason, a measurement program was launched to determine its cross-sections in the **resonance** range.

At the Gelina neutron-source facility in Geel, Belgium, a CEA team carried out capture and transmission measurements on several samples of $^{99}$Tc. The electron beam from the accelerator strikes a uranium target and the gamma braking radiation (or *Bremsstrahlung*) creates neutrons through ($\gamma$,n) and ($\gamma$,f) reactions. These neutrons are then **moderated** by water in two containers located below and above the uranium. More than 600 resonances have been analyzed at values of up to 10,000 **electronvolts** (eV) and mean cross-sections measured up to a value of 100 keV (figure B). These measurements have considerably improved our knowledge of this isotope.

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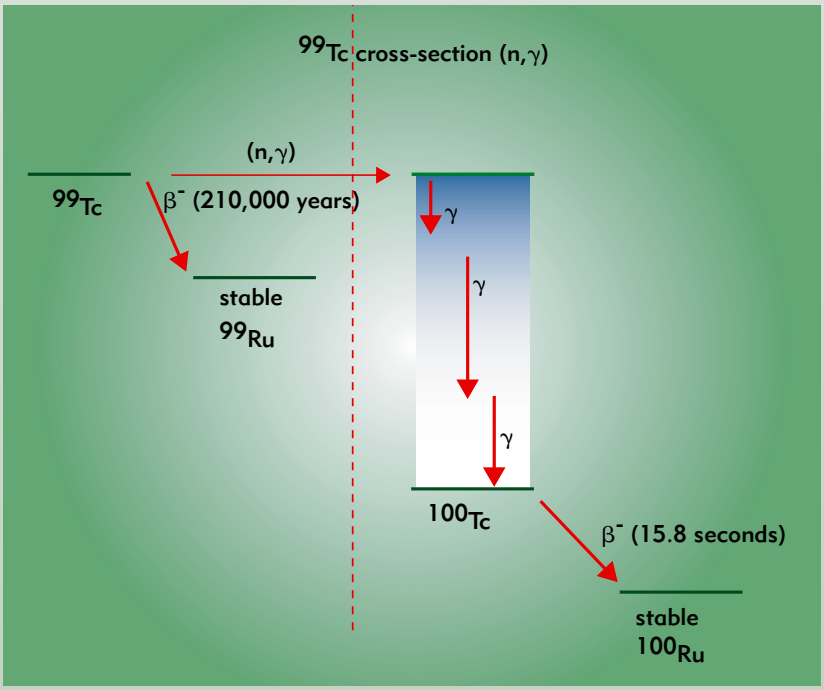


*Figure A. Technetium-99 transmutation principle.*

*The Gelina linear electron accelerator at the European Union Joint Research Center in Geel, Belgium, one of the most powerful neutron sources in the world. This facility is used to determine cross-sections, a vital factor in transmutation studies.*

IRMM-Geel

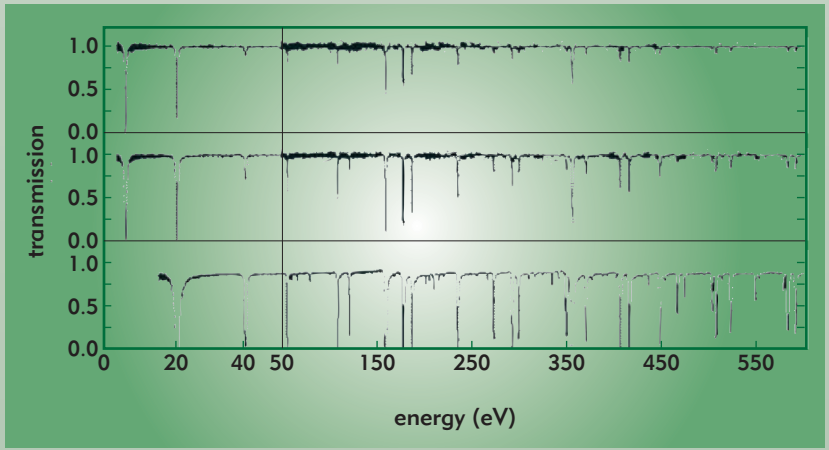


*Figure B. Neutron transmission through $^{99}$Tc targets of three different thicknesses. Each dip represents an eigenstate of the $^{100}$Tc nucleus located above its ground state at an energy level of 6.8 MeV (binding energy of the neutron in the compound nucleus), plus the kinetic energy of the neutron shown on the X-axis. The shape of the dip is thus described by the resonance parameters and the widening due to the Doppler effect and resolution.*

## Can the minor actinide, americium-241, be transmuted by thermal neutrons?

As part of their research on the **transmutation** of **long-lived** waste, teams from the Physical SCIENCES and Nuclear ENERGY divisions of CEA and the Laue-Langevin Institute (ILL) in Grenoble have joined forces to find a solution to a major problem in the chain of nuclear reactions involved in the transmutation of americium-241 ($^{241}$Am), induced by **thermal neutrons**. Amongst the **minor actinides**, it is the most significant in terms of quantity and the most **radiotoxic** for a period of 2,000 years (it has a half-life of 432 years), at which point, the neptunium generated by its **decay** takes over in the long term, after 200,000 years. So far, it has been impossible to determine whether transmutation was feasible with thermal neutrons. In its transmutation chain, the **capture cross-section** of the ground state of americium-242 ($^{242gs}$Am, gs standing for "ground state") varies by a factor of 20 in the most commonly used nuclear databases. The US database, ENDF-B/VI, gives a value of 253 **barns**, while its European counterpart, JEF-2.2, quotes 5,500 barns. These values lead to totally contradictory results in thermal-neutron transmutation simulations. In the former case, transmutation is theoretically possible, as $^{242gs}$Am **fission** has the upper hand over capture, leading to $^{243}$Am, a non-**fissile isotope** that would therefore require an additional capture to be transmuted through fission. In the second case, it is not feasible because it demands an excessive input of outside neutrons. It should nevertheless be stressed that this difference, which has a crucial impact on the neutronic properties of an **incineration** system with a high thermal-neutron flux (approx. $2 \times 10^{15}$ n/s/cm$^2$), has virtually no effect in a conventional reactor, where the concentration of $^{242gs}$Am remains very low because of the low neutron flux and the absence of $^{241}$Am in the initial fuel.

The discrepancy between databases is explained by conflicting theoretical evaluations, due to the lack of experimental values. The very short **half-life** of $^{242gs}$Am, 16.02 hours, rules out the use of conventional experimental methods. A solution has been set up at the ILL, which owns the reactor with the highest thermal-neutron flux in the world. Using a $^{241}$Am target, the high flux forms enough $^{242gs}$Am to have a strong impact on the transmutation chain. Furthermore, the perfect neutron **thermalization** of the ILL reactor implies that experimenters have no need to make corrections that would decrease the measurement accuracy.

At the start of the transmutation chain (figure), it can be seen that the $^{242gs}$Am capture cross-section determines the amount of $^{243}$Am formed during the irradiation sequence. During irradiation, $^{241}$Am transmutes through successive neutron captures and **radioactive** decay into other, possibly fissile, actinides.

The aim of the experiment was to observe changes in a set of 13 samples, each containing 30 µg (or $10^{-6}$ g) of $^{241}$Am, irradiated in two neutron fluxes of different intensities ($5 \times 10^{14}$ n/s/cm$^2$ and $3 \times 10^{13}$ n/s/cm$^2$) for periods from 30 minutes to 24 days. In each sample, the quantity and isotopic composition of americium, curium, and plutonium nuclei, and **fission products**, greatly depends not only on the $^{242gs}$Am capture cross-section value, but also on that of other cross-sections. By measuring the isotopic composition of each irradiated sample, the transmutation of $^{241}$Am in a high-intensity thermal-neutron flux can thus be determined experimentally.

Unfortunately, it is not easy to determine the isotopic composition. After irradiation, the samples are highly radioactive, making

(1) Branching ratio: the branching ratio of an isotope A to an isotope B is the fraction of isotope B formed for a decay of isotope A.

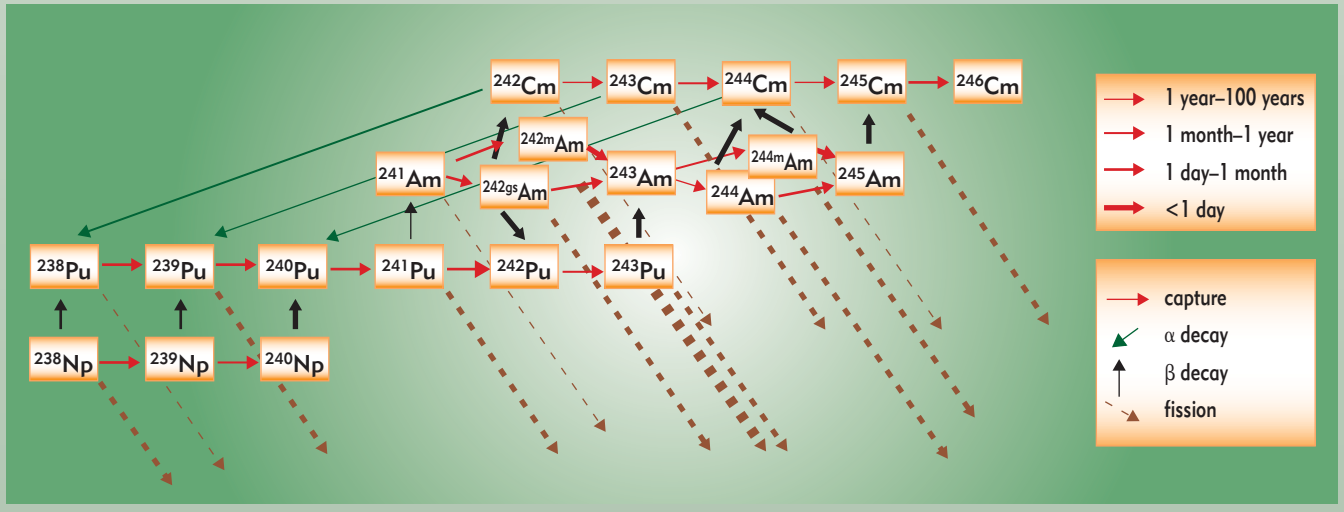


*Figure. Isotopes formed during $^{241}$Am irradiation in a thermal neutron flux. The dotted arrows in bold indicate at what point incineration through fission becomes possible. The transmutation process is more effective when fission occurs after the fewest successive neutron captures.*

the case of minor actinides, as it leads to the generation of other **actinides** and simply shifts the problem elsewhere.

In the second reaction (for **heavy nuclei** only), the impact of the incident neutron causes the nucleus to split into two fragments, fission products with a generally short half-life. This is obviously the better choice for destroying actinides (box 3), firstly because fission generally leads to short-lived residues that are less toxic in the long term than the destroyed actinide, and secondly because fission generates additional neutrons that can be used to destroy other waste or help to sus-

it impossible to handle them or take direct measurements by gamma (γ) spectrometry. The solution is to combine an X- and γ-ray measurement of one sample after a very brief irradiation period (30 minutes), with that of another sample irradiated for 24 hours but cooled for 8 months. The spectra of the two samples can be compared to determine the precise $^{241}$Am capture cross-section value, another critical and uncertain parameter in the transmutation chain. Most analyses were carried out by thermal-ionization mass spectrometry (TIMS) or inductive-coupled plasma mass spectrometry (ICP-MS) after six to nine months of cooling at CEA/Saclay. This method gave the isotopic composition of the americium, curium, and plutonium nuclei and fission products of the different samples. This method offers sensitivity of several tens of nanograms ($10^{-9}$ g) and very high precision (between 0.5 and a few per cent).

Results were compared with the predictions obtained for various cross-section values of $^{241}$Am and $^{242gs}$Am. This comparison confirmed the low theoretical prediction for the $^{242gs}$Am cross-section, with a value of (330 ± 50) barns and, for $^{241}$Am, a value of (696 ± 48) barns, with a branching ratio(1) (between the formation of the ground state and the metastable state of $^{242}$Am) of (0.914 ± 0.007). In addition, after 19 days' irradiation in a thermal neutron flux of $5.6 \times 10^{14}$ n/s/cm$^2$, (46 ± 5)% of the initial $^{241}$Am was transmuted through neutron capture, of which (22 ± 8)% was incinerated through fission. Consequently, it remains theoretically possible to transmute $^{241}$Am using a high-intensity thermal-neutron flux.

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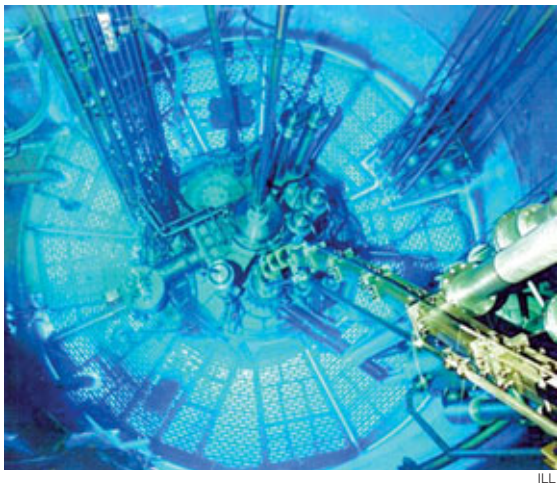

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*The thermal neutron, high-flux reactor at the Laue-Langevin Institute (ILL) in Grenoble, where the feasibility of transmuting the minor actinide americium-241, using thermal neutrons has been demonstrated.*

| isotope | PWR "thermal" reactor | | | FNR "fast" reactor | | |
|---|---|---|---|---|---|---|
| | $\sigma_f$ | $\sigma_C$ | $\alpha$ | $\sigma_f$ | $\sigma_C$ | $\alpha$ |
| $^{237}$Np | 0.52 | 33 | 63 | 0.32 | 1.7 | 5.3 |
| $^{241}$Am | 1.1 | 110 | 100 | 0.27 | 2.0 | 7.4 |
| $^{243}$Am | 0.44 | 49 | 111 | 0.21 | 1.8 | 8.6 |
| $^{242}$Cm | 1.14 | 4.5 | 3.9 | 0.58 | 1.0 | 1.7 |
| $^{243}$Cm | 88 | 14 | 0.16 | 7.2 | 1.0 | 0.14 |
| $^{244}$Cm | 1.0 | 16 | 16 | 0.42 | 0.6 | 1.4 |
| $^{245}$Cm | 116 | 17 | 0.15 | 5.1 | 0.9 | 0.18 |

*Table. Average fission $\sigma_f$ and neutron capture $\sigma_C$ cross sections, expressed in barns, and their $\alpha = \sigma_C/\sigma_f$ ratio.*

tain the **chain reaction** while producing energy.

The different kinds of fission reactors (critical or, as will be seen later, subcritical) are characterized by different **spectra**: **thermalized neutrons** (e.g. **pressurized water reactors** or **PWRs**) or **fast neutrons** (**FNRs**): see box F, ***PWR, FNR, and GCR***). The probability of actinide nucleus fission varies with neutron energy and is always in competition with other reactions, especially neutron capture. The cross-sections of the various isotopes can be averaged in the various types of neutron spectra to obtain an overview of their respective qualities. This is particularly helpful in deciding whether or not to favor fission rather than neutron capture.

The table shows the mean fission and capture cross-sections (and their ratio α) in the neutron spectrum of a standard PWR and a Superphénix-type FNR. The values show that FNRs offer definite advantages for transmutation from the physical point of view. They promote fission, which explains their "omnivorous" character. In the thermalized spectrum, the minor actinides chiefly undergo neutron capture, except for curium-243 and -245. This particular feature has been put to use in one of the processes currently studied, aimed at promoting transmutation by neutron capture to transform americium into curium, and then **incinerate** it to exploit its high nuclear-fission probabilities. These characteristics are fundamentally linked to the neutron spectrum in the reactor, regardless of whether the reactor is operated in critical or subcritical mode.

## More thorough analysis required

The analysis discussed in this article is still at a qualitative and approximate stage. More detailed answers must now be found to several questions. Is the addition of minor actinides or fission products to be transmuted compatible with the neutron balance of the reactor core involved? Will the production of other isotopes have an impact on fuel-cycle operations for example, owing to an increase in neutron and gamma emission sources? Does adding "outside" isotopes to the core affect the physical parameters of the core? The answer to these and other questions is discussed in the following articles.

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