SPENT FUEL CRITICALITY: how to make the most of burnup credit

In criticality safety files relating to situations involving spent fuels – storage, transportation, processing, etc. – studies are always highly conservative as they are built on the assumption that the fuel is fresh, meaning that it still contains the maximum amount of fissile material: uranium-235, plutonium-239 and plutonium-241. In reality, as the reactor is irradiated, the fuel undergoes radical changes to its composition that reduce its reactivity. By taking account of this phenomenon, known as burnup credit (BUC), criticality safety analyses can optimize the design of certain nuclear facilities in order to operate them more economically, while guaranteeing the necessary safety margins.

Spent fuel storage pit at the La Hague fuel reprocessing plant. By taking BUC into account, fuels with a higher initial enrichment in uranium-235 can be stored.

Harry Gruyaert/Magnum - Cogema

Negative reactivity nuclides

In a **pressurized water reactor (PWR)** (see box E, ***The main components of a PWR***), the fuel is composed of uranium oxide **enriched** with its ^{235}U **isotope** (**UOX**) or a mixture of uranium and plutonium oxides (**MOX**). The fuel undergoes **neutron** irradiation and **fission products** are formed. These result from the **fission** of uranium, plutonium, and heavy nuclei generated by repeated neutron **captures** or **(n,2n) reactions**, generally followed by β^- or α **disintegrations** (see box B, ***Interactions between neutrons and nuclei***). These heavy nuclei include **major actinides** (uranium and plutonium) and **minor actinides** (neptunium, americium and curium). The variations in composition reduce the **reactivity** of the fuel. Non **fissile** actinides and fission products contribute *negative reactivity*. During burnup in the reactor, fission products are also capable of absorbing neutrons. Most of them have very high neutron capture **cross sections**, leading to considerable fuel **poisoning** that increases with irradiation time.

Criticality safety computing tools developed in the 1970's did not take account of the consumption of fissile materials and the creation of negative reactivity **nuclides** in UOX fuel during burnup. The resulting safety margins calculated at that time now seem excessive in light of the optimization of plant design achieved since then.

In the 1980's, a certain burnup credit (BUC) was taken into account in optimizing the design of the new, higher-capacity UP3 unit at the Cogema-La Hague (Manche) spent fuel reprocessing plant in France. At that time, however, only data relating to the main actinides was deemed sufficiently qualified to be taken into account, which meant that the BUC could not be considered in full.

In the early 1990's, the BUC model was defined, taking into account changes in major actinides, the production of minor actinides and of the fifteen most negatively reactive stable or long-lived, non volatile fission products (see Table 1). This selection of nuclides was adopted by the group of experts in **criticality** calculations of the OECD (Organization for Economic Cooperation and Development) NEA (Nuclear Energy Agency). In 1995, a research and development program was initiated in France on the basis of these expert studies, jointly conducted by CEA, IPSN and Cogema. The aim of the program is to qualify the fifteen fission products adop-

actinides	negative reactivity (pcm)	fission products	negative reactivity (pcm)
uranium-235		samarium-149	- 1 030
uranium-238		rhodium-103	- 1 360
plutonium-239		neodymium-143	- 900
plutonium-240	- 8 370	cesium-133	- 750
plutonium-241		gadolinium-155	- 1 550
plutonium-242	- 710	samarium-151	- 500
major actinides	**- 9 080**	samarium-152	- 490
		technetium-99	- 440
uranium-236	- 910	neodymium-145	- 410
neptunium-237	- 620	europium-153	- 390
plutonium-238	- 310	molybdenum-95	- 290
americium-241	- 1 290	samarium-147	- 230
americium-243	- 280	samarium-150	- 270
minor actinides	**- 3 410**	silver-109	- 250
		ruthenium-101	- 220
total actinides	**- 12 490**	**total fission products**	**- 9 080**

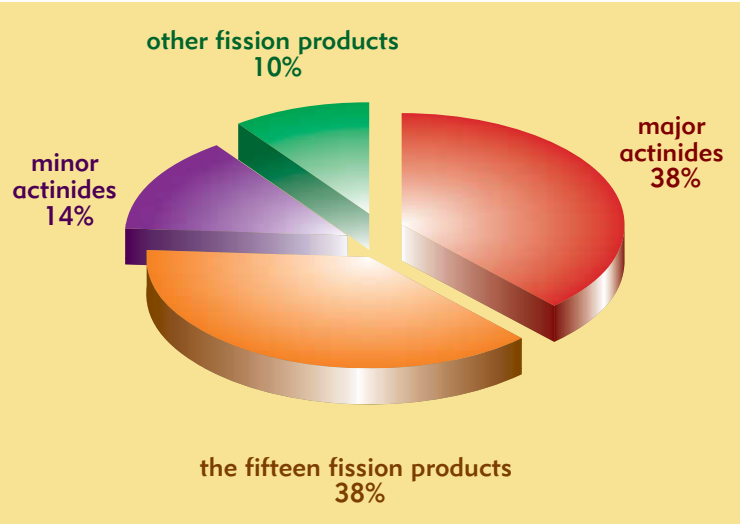


Table 1. Negative reactivity of nuclides selected in studying burnup credit for a uranium oxide fuel (UO_2), irradiated at a burnup of 40 000 MW·d/t and cooled for 5 years, and contribution of these nuclide categories to the total negative reactivity of the fuel (related BUC). Taken together, these nuclides account for 90% of total negative reactivity while the fifteen fission products represent 80% of the total poisoning of the fission products formed.

	consideration of BUC	related BUC	total percentage
today	major actinides	38%	38%
short term (2003)	main fission products	38%	76%
medium term	minor actinides	14%	90%

Table 2. Consideration of BUC stages.

ted by the expert group. It is scheduled to end in 2003, at which point it should be possible to integrate the main fission products in safety studies. A program focusing on minor actinides will be launched in the medium term (see Table 2).

Industrial requirements and expected advantages

For Cogema, consideration of BUC is of particular significance in three areas of activity: the *transportation* of spent fuel assemblies from nuclear reactors to the La Hague plant, implying in particular the *development of new improved transportation containers*; the *storage* of assemblies both at La Hague and in design work for storage facilities for different customers; the *processing* of spent fuel at La Hague.

Spent fuel transportation

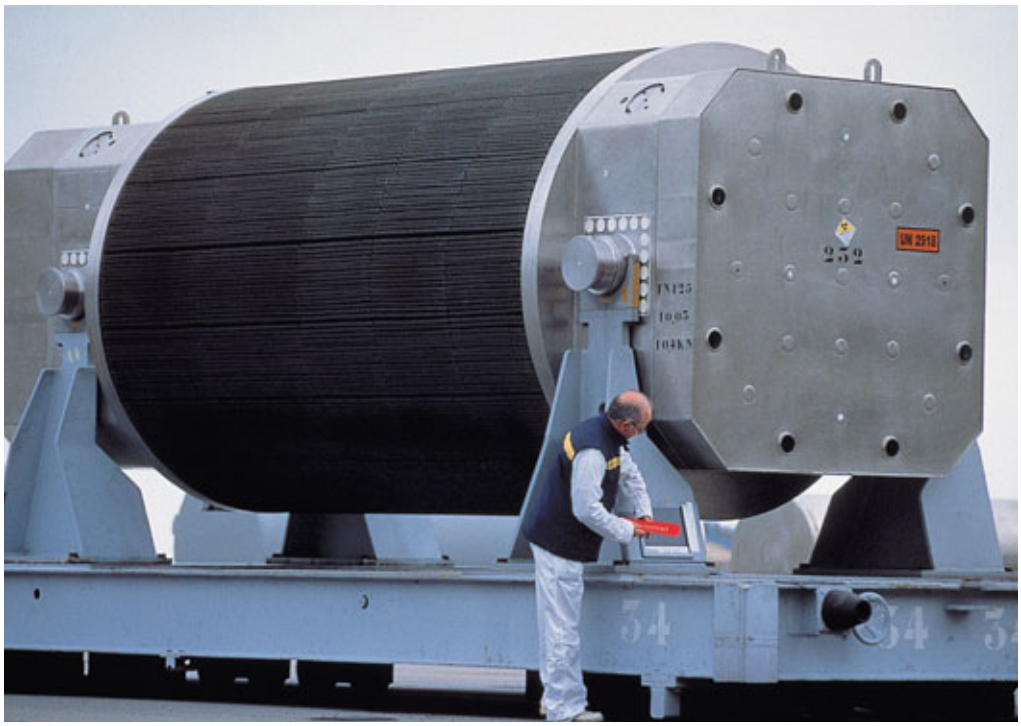
Consideration of the BUC means that more reactive fuel assemblies can be transported by a given type of container, more reactive fuels being those with a higher initial enrichment in ^{235}U at a fixed **burnup**. Integrating BUC into the design of new transportation containers makes it possible either to transport more assemblies per container, or to lighten the structure of the container by packing the assemblies closer together. Packing assemblies closer together increases the overall reactivity. However, the negative reactivity taken into account in the BUC offsets this increase, thus reducing spent fuel transportation costs.

Storage in spent fuel pits

Existing spent fuel storage pits were designed twenty years ago for one type of fuel assembly. Taking BUC into consideration means that these same pits can be used to store fuel assemblies which have higher initial enrichment, in order to be irradiated for longer periods, in keeping with the current trend. Thanks to BUC, new storage facilities will be able to offer more compact and, therefore, more economic storage concepts for a given type of fuel assembly.

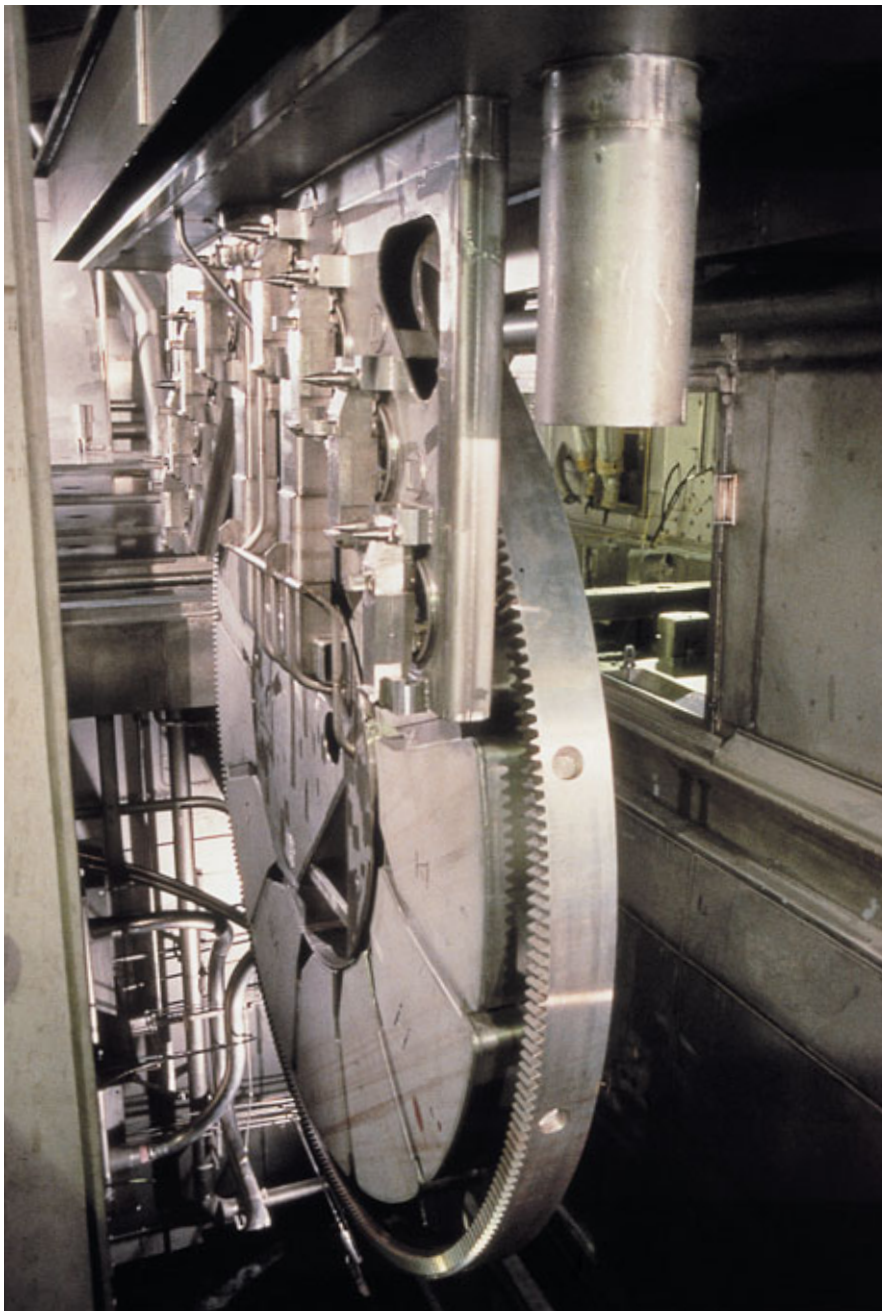
Spent fuel processing

In spite of the increased initial enrichment of the fuel handled at La Hague, BUC allows each of the two La Hague plants – UP3 and UP2-800 – to maintain the same processing rates of roughly 4 tonnes of fuel per day. This rate is imposed by the production shop at the start of the process, the spent fuel assembly shearing and dissolving shop where assemblies (bundles of rods about 20 cm in diameter and 4 m in height) are cut into segments 3 to 4 cm long. After shearing, these segments – called *hulls* – are dropped into a bucket feeding the wheel of a rotating dissolver. The wheel is immersed in a hot nitric acid solution that dissolves the nuclear material in the hulls.



Jean-Marie Taillat/Cogema

Spent fuel assembly transportation container. Consideration of BUC will make it possible to transport more assemblies per container or optimize container structure.



Sidney Jezequel/Cogema

One of the dissolver wheels in the spent fuel assembly shearing and dissolving shop at the La Hague spent fuel reprocessing plant. Taking BUC into consideration means that the plant will be capable of processing fuel with higher initial enrichment in uranium-235 at the same processing rate as before.

An interesting new application for BUC

This concerns a new type of container for storing spent fuel assemblies in the pit. The container can hold up to 9 assemblies and is more compact than containers currently in use at La Hague. The CEA Apollo **neutron calculation code** is used as part of a criticality safety study to determine the maximum initial enrichment permissible for **pressurized water reactor (PWR)** uranium oxide fuel, for a given burnup, in assemblies consisting of a 17 x 17 fuel rod lattice. The maximum permissible enrichment is obtained when the assemblies are considered to be off center in their cells (Figure 1). The result shows that without BUC – in other words, with zero burnup – maximum permissible enrichment is approximately **3.4%**. The fuel assemblies used in today's PWR cores have enrichment values between 3.7% and 4.1%. The French national electric utility, EDF, plans to increase these enrichment levels to 4.9% over the next ten years.

The only way to accommodate such levels is by taking BUC into consideration. For a burnup value of

10 000 MW·d/tU[1], for example, the maximum permissible enrichment rises from the present value of **4%** where only major actinides are considered, to more than **4.5%** if the first six fission products in Table 1 are integrated, and to more than **4.6%** when all fifteen fission products in the table are added. This example clearly illustrates the impact of BUC on the design of this new storage container.

The BUC calculation code package

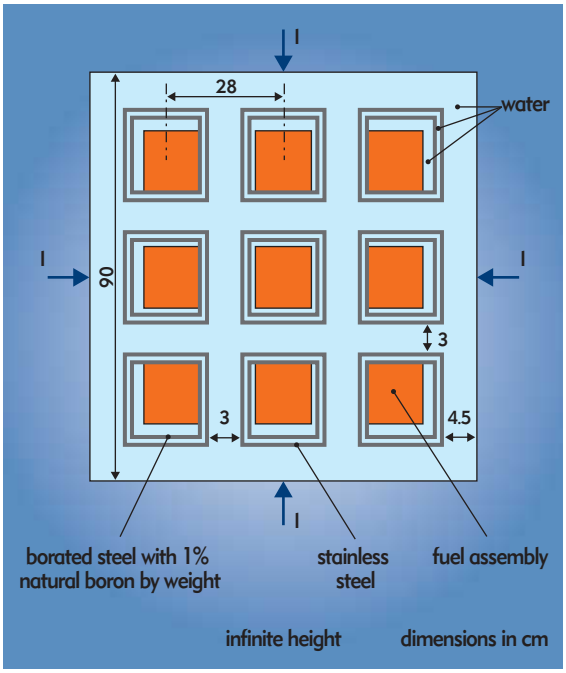
Acknowledging the impact of BUC in criticality safety studies calls for a qualified calculation code package to determine the abundances of nuclides, actinides and fission products contained in spent fuel, and the negative reactivity contributed by these isotopes in different configurations. For this purpose, a calculation code package coupling the Darwin and Cristal **packages** (see *Cristal: a new-generation criticality package*) is used. Darwin is used to plot changes in significant physical parameters – in particular the abundance of each nuclide – in the reactor and during the cooling cycle for fuel cycles in all types of reactors (**PWR pressurized water reactors**, **BWR boiling water reactors**, **FR fast neutron reactors**, etc.). It provides the opportunity to integrate the detailed history of the fuel assembly.

BUC calculations reveal specific physical problems such as the **resonance self-shielding** of fission products which prevents neutrons from disappearing in the reactor because of the gigantic capture cross sections of these nuclides. This problem is addressed by a finer breakdown into 172 energy groups in Apollo2 and by adding effective cross sections to the CEA93 library. Furthermore, studies have demonstrated that once a mean burnup of 30 000 MW·d/t per assembly has been reached, an axial burnup profile must be considered to obtain more accurate calculations. This is because the ends of the assembly are less irradiated and, therefore, more reactive. In this case, the Darwin package can be used to calculate axial variations in nuclide composition as a function of axial burnup.

A large number of experimental programs

A large number of experimental programs are under way to qualify BUC

(1) The customary unit for measuring fuel assembly burnup, i.e. the energy extracted from the assembly in the reactor, is the Megawatt·day. This burnup is expressed per unit mass of fuel, the tonne (metric ton) of initial uranium (MW·d/tU). 1 MW·d = $3\ 600 \cdot 24 \cdot 10^6 = 8.6 \cdot 10^{10}$ joules. The current irradiation level of fuel assemblies is in the region of 50 000 MW·d/t. Before irradiation, an assembly contains between 450 kg and 550 kg of uranium.



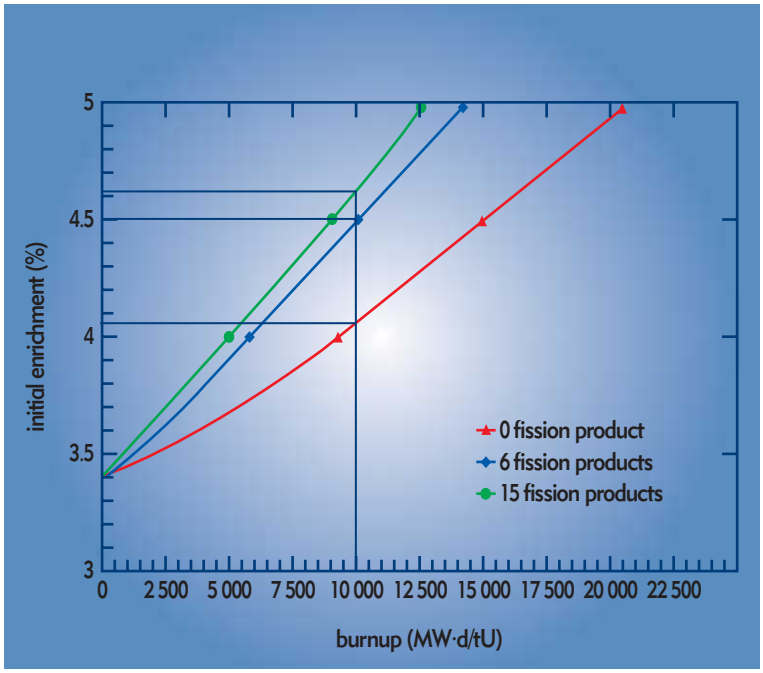


Figure 1. On the left, a new design of container for storing spent fuel assemblies in a pit, illustrating the penalizing case when the assemblies are not centered in their cells. This case represents the maximum permissible initial enrichment of the fuel. On the right, the maximum permissible initial enrichment for uranium oxide assemblies as a function of burnup. For a given burnup level, the maximum permissible initial enrichment increases when 6, then 15 fission products are considered in addition to major actinides.

CEA

The CEA/Cadarache Comir unit where the physical properties of irradiated fuel samples are measured.

calculations. The programs will help to obtain the experimental qualification of the Darwin package, used to calculate nuclide "material balances" by analyzing irradiated fuel samples, and the Cristal package, used to calculate the reactivity effect by taking measurements of the negative reactivity of each fission product and overall measurements integrating fission products and actinides.

Irradiated fuel analyses

As a complement to the actinide analysis programs carried out jointly by CEA, EDF and Framatome, analysis programs focusing on fission products contributing to BUC have been started as part of CEA and Cogema common interest programs. Segments of irradiated fuel are taken, then dissolved in the CEA/Cadarache Comir unit. Samples are then analyzed in CEA/Saclay laboratories. Analysis results for the nuclides mentioned in Table 1 cover the following ranges:

- PWR(UOX): burnup between 15 000 MW·d/t and 63 000 MW·d/t.
- PWR(MOX): burnup between 10 000 MW·d/t and 43 000 MW·d/t.
- BWR(UOX): burnup between 20 000 MW·d/t and 43 000 MW·d/t.

Experimental uncertainties are less than 1% for uranium and plutonium isotopes and between 2% and 8% for the other actinides and fission products.

Reactivity effect measurements

Common interest programs conducted by CEA and Cogema have been initiated in the Nuclear Energy Division's Minerve experimental reactor at CEA/Cadarache and in the IPSN *"appareillage B"* facility at CEA/Valduc.

In Minerve, the reactivity effect of a sample is measured by a series of oscillations(2) in an experimental lattice representative of industrial neutron conditions. This method provides a measurement with an overall precision of some 3%. Two types of samples are subjected to "oscillations": the fifteen fission products adopted for BUC purposes are considered separately to measure their individual neutron capture cross section, and segments of irradiated fuel are taken to measure the overall loss of reactivity (fission products + actinides) of different types of irradiated fuel (UOX and MOX). These measurements cover a burnup range of 15 000 MW·d/t to 63 000 MW·d/t. The experimental lattices studied are representative of the transportation and storage conditions of spent PWR(UOX), PWR(MOX) and BWR(UOX) fuels, and of UOX and MOX fuel dissolving conditions.

In the "*appareillage B*" facility, the integral experiments, taking into consideration the first six fission products

(2) The sample oscillation measurement method consists, in a critical reactor, to generate small periodic disturbances affecting certain neutron properties that cause a variation in the effective multiplication factor in order to obtain the reactivity variation Δρ resulting from the sample.

View of the Minerve experimental reactor core at CEA/Cadarache. This is used to measure the neutron capture cross sections of fission products and the overall reactivity loss of different types of irradiated fuel.

M. Faugère/CEA

Fuel rod lattice in the "appareillage B" facility at CEA/Valduc. A solution of fission products, separate or mixed, are made to interact with UOX rod lattices.

IPSN

contributing to BUC, measure the physical variable of interest (k_{eff}) in a geometry that is representative of industrial configurations. The experiments, carried out in a subcritical approach(3), require a gradual experimental approach, working from the simplest to the most complex configurations as follows: fission products in a casing in the center of a UOX fuel rod lattice immersed in water; UOX fuel rod lattice immersed in a uranium and fission products solution.

The many measurements carried out during these experimental programs will help to qualify the cross sections of the fifteen fission products and the Darwin-Cristal calculation code package for the consideration of BUC. The Osmose program currently being prepared at CEA/Cadarache will provide measurements of minor actinide cross sections in Minerve, thus completing this experimental base.

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(3) Adopting a subcritical approach entails causing a slow variation in a parameter characteristic of the reactivity of the medium or configuration. This parameter can be the depth of water in the vessel containing the fuel rod lattices. Water, which acts as a **moderator**, is gradually added to the vessel until the multiplication coefficient k_{eff} reaches a value just below 1 (subcritical system). The depth of water required to make the system critical is calculated by extrapolation.