PHENIX: THE IRRADIATION PROGRAM FOR TRANSMUTATION EXPERIMENTS

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Abstract

The French fast reactor prototype Phenix started commercial operation in July 1974. A major renovation, inspection and safety upgrade program was carried out at the plant from 1994 to 2003. The operating diagram for the 51st cycle is described in this paper.

The first part of this paper summarizes the experimental possibilities at the plant. As compared to the other experimental reactors, Phenix is distinguished by the very high neutron flux, more than ten times greater than the aforementioned reactors, with a relatively hard spectrum characteristic of fast breeder reactors. Other experimental conditions (temperature, measurements, loading, experiment follow-up....) are also described.

Then there is a brief description of the first transmutation test made at Phenix, referred to as SUPERFACT, irradiated in 1986-1987, which provided the first transmutation results in the field of homogenous and heterogeneous mode incineration of minor actinides.

Then follows the description of the current state of all the experiments introduced just before or during the 51st cycle, for incineration of Minor Actinides in heterogeneous mode (ECRIX-B , ECRIX-H ,MATINA ...) for incineration of Minor Actinides in homogeneous mode (METAPHIX 1,2 and 3), for transmutation of long-life fission products (ANTICORP 1 transmuting Technetium 99), and the measurement of the basic neutronic data to be used for transmutation calculations (PROFIL R).

Explanations are also given on new experiments which are currently being assembled such as CAMIX- COCHIX, MATINA 2 and 3, PROFIL M. All these experiments are also described in this paper.

In conclusion, the vast modernization program of Phenix was completed in 2003. The reactor restarted in July 2003. The specific experimental possibilities of a fast breeder reactor enable a wide scope of experiments in the field of fuel transmutation to be conducted. Significant knowledge will be gained from all these experiments during the remaining six operating cycles planned for the plant up to 2009, representing a total irradiation time of 720 EFPD equivalent to approximately 5.5 years of operation.

Introduction

An ambitious research program on separation and transmutation was started in response to axis 1 of the law of 30 December 1991 on waste management.

The principle of transmutation is to succeed in modifying the nuclei of the elements which make up long lived waste (minor actinides and some fission products) in order to convert them into bodies which are stable or which have much shorter lifetimes.

Under neutron flux, the minor actinides primarily undergo fission and neutron capture reactions. The goal is to promote fission for their destruction, for several reasons, firstly because fission generally leads to short-lived fission products and secondly because fission generates additional neutrons which can be used to destroy other waste or help sustain the chain reaction while producing energy. However capture only produces other actinide which simply perpetuates the problem.

Fission products only undergo neutron capture which converts them into stable bodies. The classic example is that of ^{99}Tc which neutron capture converts into very short life (16s) ^{100}Tc leading to stable Ruthenium-100 following the β disintegration.

Fast reactors are the best tool for this type of application due to their neutron advantages.

An experimental transmutation program is currently underway at the PHENIX fast reactor after it was started up again in July 2003.

This report describes the current status of the transmutation program as of 31 August 2004.

The Advantages of Fast Reactors for Transmutation

Flux Intensity

First of all, neutron flux in a fast reactor is 10 times higher than in a PWR (3-10^{15} n/cm^2/sec at Phénix), which increases test efficiency. Furthermore, neutron balance in a fast reactor is more favourable and provides the necessary excess neutrons.

Capture / fission ratio

Table N°1 gives the average effective fission and capture cross section (and their α ratio) in the neutronic spectrum of a PWR and a Super Phenix type FBR. The data in the table indicate that from the physical standpoint, the fast spectrum presents certain advantages for transmutation.

- The fast spectrum minimises the “parasite” capture reactions and promotes fission reactions for all the actinides, which makes it more “omnivorous”,

- In the thermalised spectrum, the minor actinides chiefly undergo neutron capture, except for Curium 243 and 245.

Table 1. **Average Cross-Section (barns)**

δ_f = fission, δ_c = capture, α = capture/fission

Isotope	**Thermal Reactor**			**Fast Reactor**		
	δ_f	δ_c	α	δ_f	δ_c	α
^{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
^{235}U	38.8	8.7	0.22	1.98	0.57	0.29
^{239}Pu	102	58.7	0.58	1.86	0.56	0.3
^{99}Tc	-	9	-	-	0.5	-

Neutronic Balance

An additional theoretical approach is to consider the number of neutrons required to convert an initial isotope (and all the isotopes produced by the successive reactions of this initial isotope) into stable isotopes or fission products. This number clearly depends on the effective cross-sections of the various reactions and thus on the neutron spectrum under consideration and on the neutron flux level.

We see that:

- In fast spectrum, we dispose of excess neutrons for all the actinides. Recourse to additional neutrons is only necessary to destroy all the long-lived fission products.
- In thermalised spectrum, the balance is negative for all the minor actinides (positive for Curium, negative for Np and Am). The Plutonium and minor actinides group also remains negative.

These theoretical considerations naturally lead to preferring the use of the fast spectrum for the incineration of the minor actinides.

Conclusion

Fast neutrons are the preferred tool for transmutation tests on long-lived waste.

Update on the PHENIX Reactor (as of 31 August 2004)

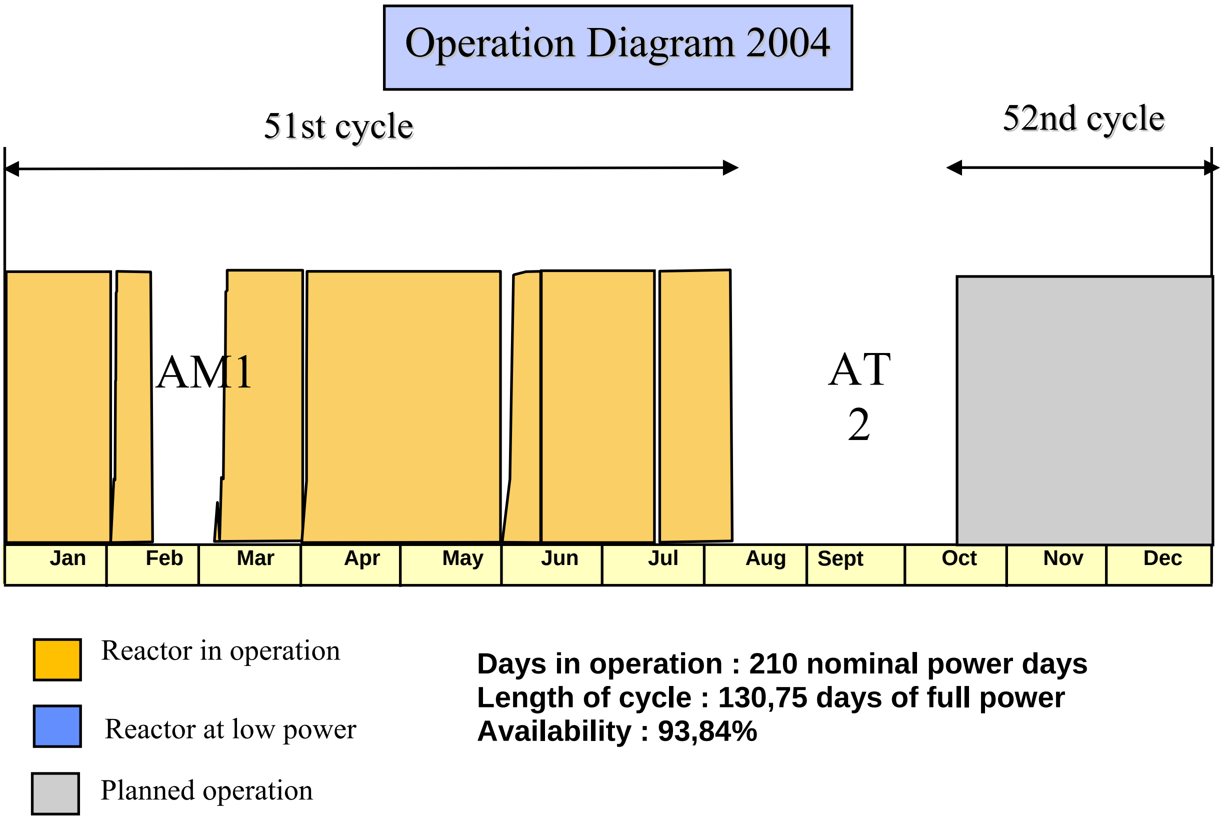
The reactor has a rated thermal power output of 563 MWth and uses sodium as coolant. Originally, six intermediate heat exchangers and three secondary sodium circuits carried the heat to the steam water circuits (3 steam generators), driving the operation of the 250-MW electric turbo-generator unit. This plant operated between 1974 and the second ten-year outage in 1989 with a load factor over 60%, which, for a prototype, was highly satisfactory and clear proof of the possibilities of this type of reactor system.

Recent years have been marked by successive safety re-evaluations which generated significant renovation work and in-depth inspections of the plant components and structures (ref. 1, 2 and 3).

This renovation work resulted in obtaining authorization to start up again in January 2003 for six cycles of 120 EFPD. During these cycles, two of the three available steam generators are in operation. The plant thus operates at 2/3 rated power, which is approximately 140 MWe.

Reactor start-up took place in July 2003, beginning the 51st cycle, which ended on 8 August 2004, after 131 EFPD operating time. The diagram below details reactor operation in 2004.

Figure 1. **Operation Diagram 2004**



Days in operation : 210 nominal power days
Length of cycle : 130,75 days of full power
Availability : 93,84%

We would like to point out that during the 1st mid-cycle shutdown, the Matina 1A experiment was removed from the reactor for non-destructive testing. During the shutdown at the end of the 51st cycle, the Métaphix 1 experiment was also removed for initial testing.

Background on the SUPERFACT experiment

The SUPERFACT experiment, irradiated at Phénix in 1986-1987, provided the first results on transmutation in the field of the incineration of minor actinides in the homogenous mode. (Although significant content was tested, since the pellet support was UO2, Superfact is considered more "representative" of the homogenous option.) Fuel containing 2 % Americium or Neptunium and incineration targets containing up to 45 % minor actinides were irradiated in fast spectrum to a transmutation rate of approximately 30 %. This experiment demonstrated the good behaviour of the fuel containing minor actinides and produced the first elements in the field of recycling Neptunium and Americium. [5]

Background on the experimental conditions at PHENIX

The Flux

Phénix stands out from the other experimental reactors (OSIRIS, HFR, BR2 type …) due to the very high neutron flux (more than ten times higher than the reactors mentioned above) with a relatively hard spectrum (high energy) characteristic of the fast neutron reactors.

The speed or dose rates obtained are thus much higher than those in other reactors. Furthermore, the timely use of moderating materials can locally slow down the neutrons, thus benefiting from the high flux inherent to FBR, while increasing the effective cross-section of the neutrons. The following table summarizes the possibilities:

Table 2. **Flux Possibilities**

		Fast flux **($n/cm^2/s$)**	**Thermal flux** **($n/cm^2/s$)**	**Dose rate**
PWR power reactor		$1.3.10^{14}$	$0.9.10^{14}$	2 dpa/year
OSIRIS, HFR, BR2 type research reactor …		2 to 4.10^{14}	4 à 7.10^{14}	3 dpa/year
Phénix reactor	Fast spectrum	$4.4.10^{15}$		18 dpa/6-month cycle
	Type 1 moderate flux*	$3.6.10^{15}$		12 dpa/cycle
	Type 2 moderate flux*	$1.5.10^{15}$	$3.7.10^{13}$	4 dpa/6-month cycle

** Types 1 and 2 correspond to the type of mode-rating carrier used*

Temperatures

The irradiation of the inert materials (containing no fissile material) generally takes place at sodium temperature, between 380°C at the entrance to the core and 550° C average temperature at core exit.

However, to cover the new needs shown by gas-cooled reactors, an innovative device is being developed which allows for irradiating specimens up to 1000°C. This device is based on the use of the power left in the materials (samples, sample-holders and heating boxes) by the radiation and the neutrons, and the dimensioning of the gas gaps providing the thermal insulation between the heating boxes and the sealed containers.

Measurements in the Reactor

The experiments are placed in the reactor core after passing through the sodium storage tank, referred to as the drum. In theory, there is no possible connection with the outside. This constraint sharply limits the possibilities of instrumentation and measurement.

The instrumentation commonly used includes measurements of sodium temperature at the entrance to and exit from the experimental set-up, radiological analysis of the sodium at the exit and mass spectrometry or gamma spectrometry analysis of the leakage gases.

Two temperature measurement devices will be used in the high temperature setups currently being developed. The first is based on phenomena of the SiC reconstitution which occurs when the SiC is heated, after irradiation, to a temperature above or equal to its irradiation temperature. The second determines the maximum temperature reached during radiation, using a post-irradiation examination. This measurement device uses a stack of materials with increasing fusion temperature, arranged so that their successive fusion causes the stack to gradually decrease in height.

Experimental Setups

The experimental sub-assemblies

These are sub-assemblies similar to the standard fuel assemblies in the Phénix core, to which different types of changes have been made:

- Nature of the structure materials (cladding, hexagonal wrapper, etc. …),
- Nature or composition of the fuel,
- Geometry of the pin or pellet,
- Production process for a component, etc.…

Most of these sub-assemblies are fissile; however, there can be fertile sub-assemblies and even control rods.

This type of setup provides irradiation conditions which are very close to actual operation, yet which do not allow for any significant deviation from the standard design. The experimental sub-assemblies have primarily been used for experiments related to the development of the Fast Neutron Reactor system.

The irradiation rigs:

The irradiation rig is complementary to the experimental sub-assembly. It is generally made up of a 40-mm diameter tube equipped with a sodium feed at the base and a locking and handling head which can contain a wide variety of experimental objects:

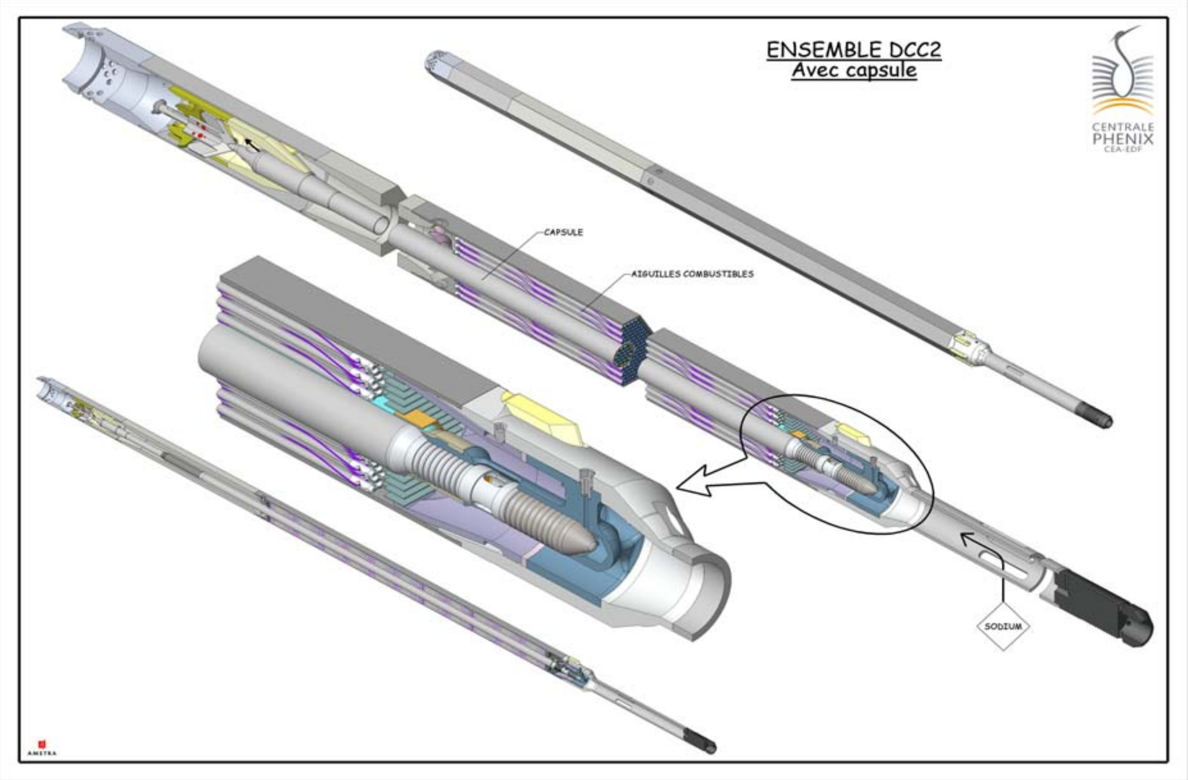
- Phénix or other geometry fuel pins, able to hold standard or experimental fuel,
- Absorber pins
- Specimens of materials intended for mechanical tests (tensile strength, resilience, toughness, fatigue, creep, etc…)
- Transmutation target, etc…

The capsules are irradiated inside special assemblies called “carriers".

The special feature of these sub-assemblies is their central channel into which the rig is inserted. There can be different types of carriers depending on the irradiation conditions being sought (fissile, fertile, steel, etc…). A new type of carrier has just been designed and made for the irradiation program. This carrier creates a zone of moderate neutrons around the rig, in the core or core periphery. The goal is to improve the transmutation performances of certain experiments.

Highly innovative experiments can be conducted with these irradiation rigs. They offer a wide range of irradiation conditions both with respect to the neutron flow and its spectrum, and the irradiation temperature range. In addition, since their assembly and dismantling are possible in cells at Phénix, pins which have already been examined after one irradiation can be irradiated again.

Figure 2. **Irradiation rig**

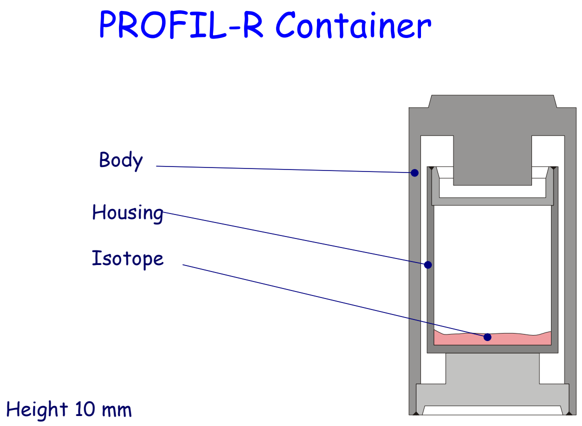


The Experimental Program at the Outset of the 51st cycle - Ref. [4]

PROFIL R basic neutron data acquisition

The objective of the PROFIL R experiment is to acquire basic neutron data to use for transmutation-related calculations. This experiment is conducted in a quasi-standard fuel sub-assembly in which two experimental pins have been placed among the bundle pins. The two experimental pins each contain 55 containers containing a few milligrams of separated minor actinide isotopes (from thorium to Curium) or fission products to be studied. This type of experiment is extremely productive to work out key information on the effective cross-sections of the various isotopes in the reactor's representative spectrum from the isotopic balances after irradiation.

Figure 3. **Diagram of a Profil-R container**



Transmutation of ANTICORP 1 fission products

The ANTICORP-1 experiment is designed to demonstrate the feasibility of transmutation of Technetium into stable Ruthenium. Three pins, each containing two bars of technetium 99 (accounting for a total mass equal to 3 x 12 g of Tc) in metallic form, are irradiated in moderate flux, with the objective of reaching a 25% rate of transmutation. ANTICORP-1 will provide us with further knowledge on the behaviour (in particular the swelling) of the metal bars during transmutation.

Transmutation of Americium in heterogeneous mode

ECRIX B and ECRIX H

The goal of the ECRIX B and H experiments is to test the feasibility of the incineration of americium in heterogeneous mode in the context of monorecycling in FBR. Two identical experimental pins are placed in the reactor under different irradiation conditions. Inside each pin is a 200-mm high column made of pellets holding americium oxide particles which are a few micrometers in diameter and are uniformly distributed in an inert magnesia matrix.

These experiments are conducted inside a device capable of locally moderating the neutron flux, in order to increase the effectiveness of the transmutation. The significant neutron flux available at Phénix can be used to advantage, in combining it with high cross-sections, accelerating the conversion of the Americium in Curium and promoting its disappearance, benefiting from the high fission cross-sections of the isotopes 243 and 245 of the Curium.

The three figures below show the evolution of the compositions of the Americium targets in a PWR, then in Phénix (but without moderator), then in Phénix with moderator (the case of ECRIX B and ECRIX H). We see that we therefore succeed in obtaining transmutation rates close to 100 %.

Figure 4. **Americium target in PWR**

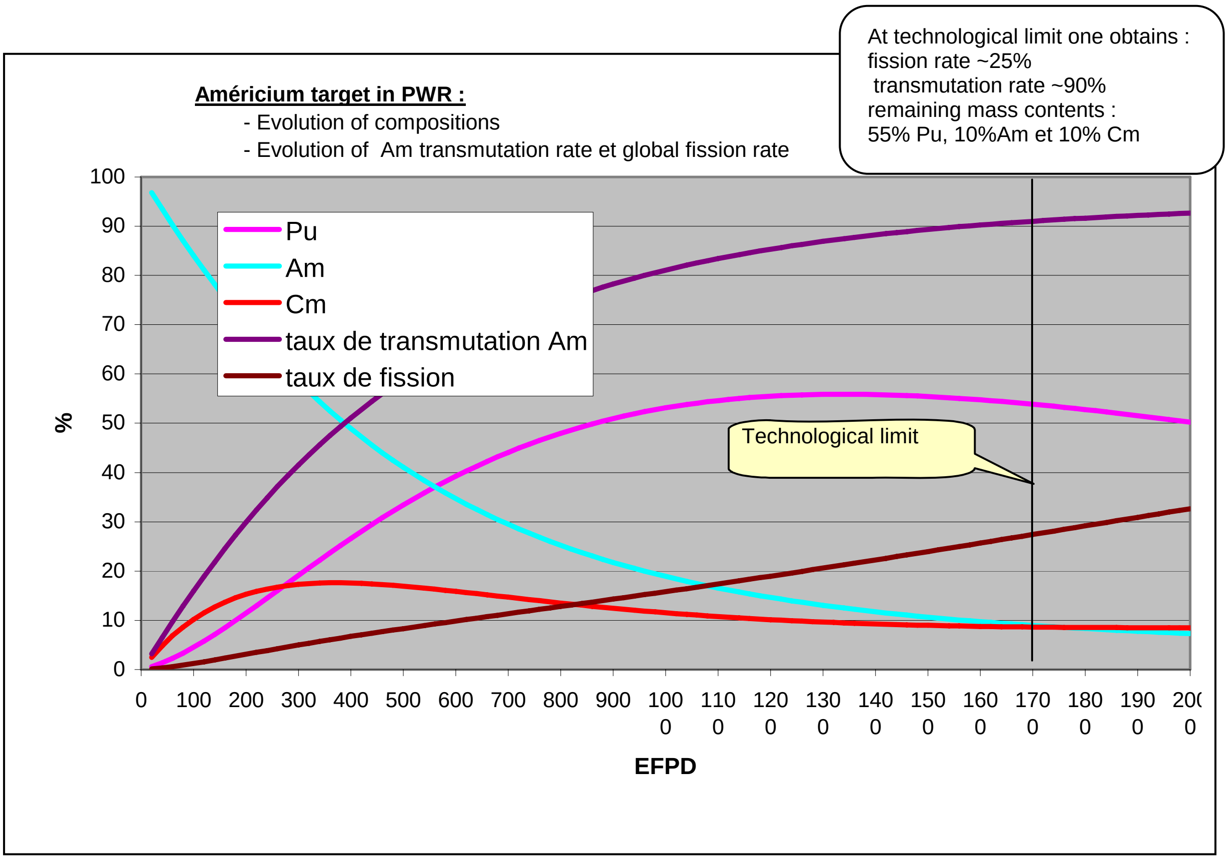


Figure 5. **Americium target in PHENIX fast neutron spectrum**

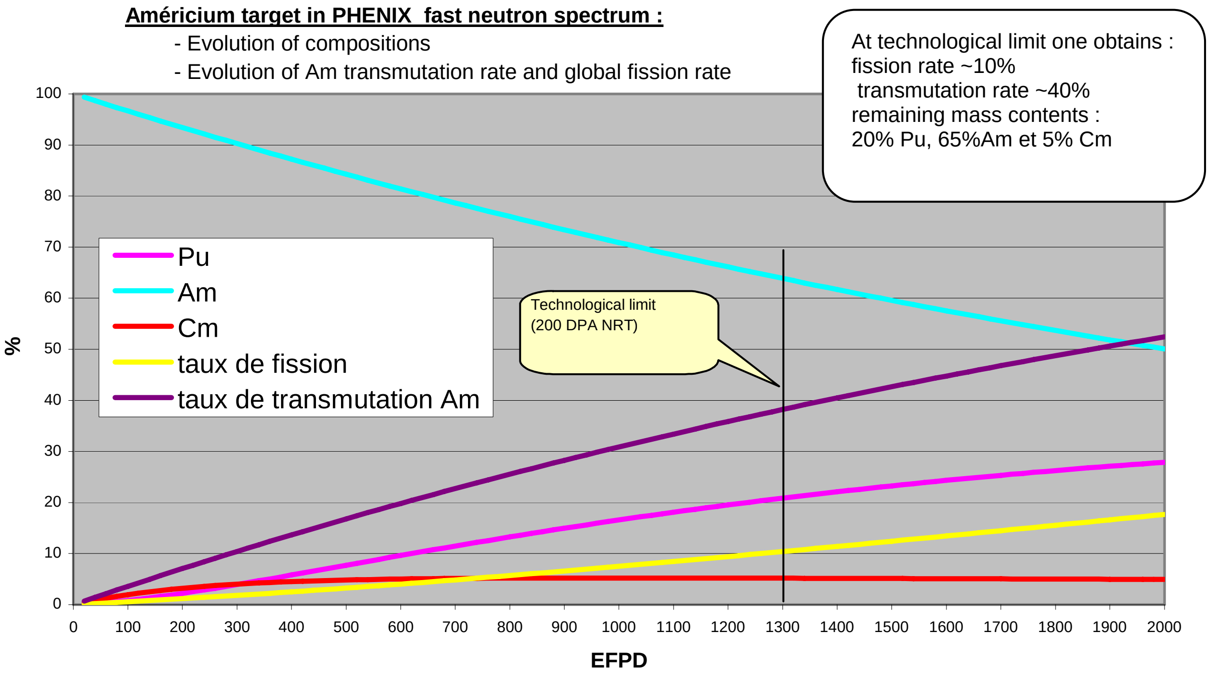
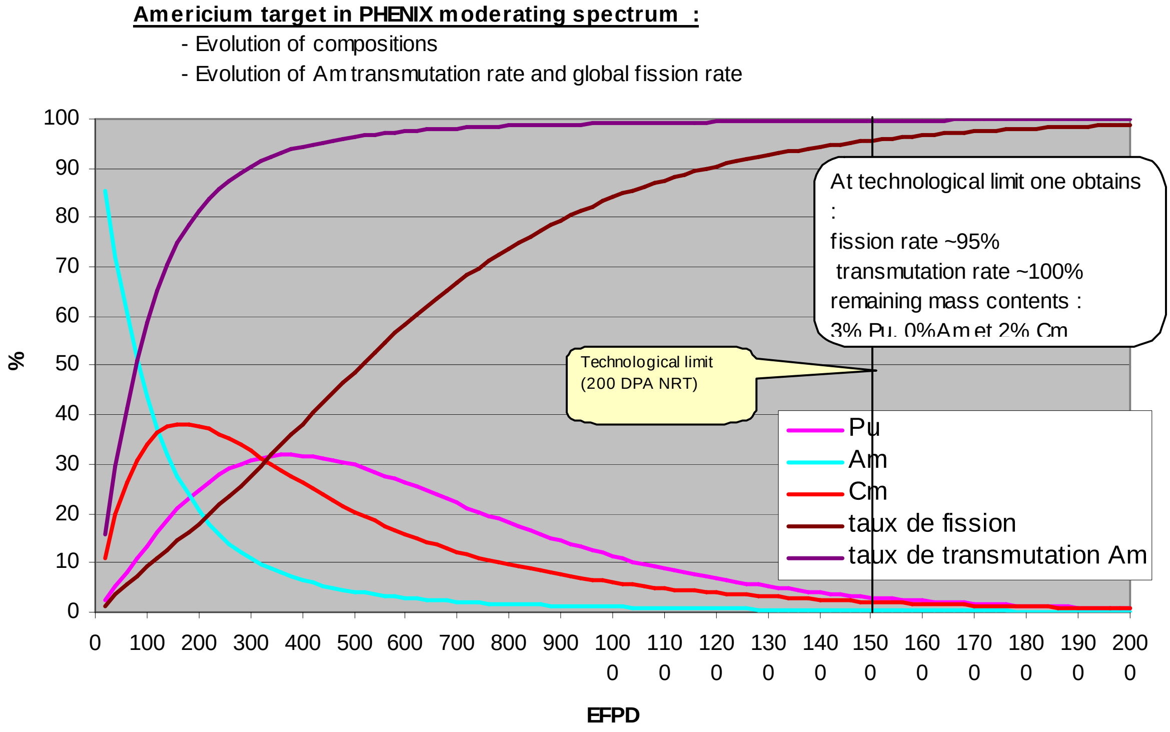


Figure 6. **Americium target in PHENIX moderating spectrum**



Transmutation of Americium in homogeneous mode

METAPHIX 1, 2, 3

METAPHIX is an experiment in the transmutation of minor actinides (Neptunium, Americium, Curium) and rare earths in homogeneous mode. Three rigs, each containing three experimental pins, are placed in the reactor for different periods of irradiation. This experiment is different because it uses the metal UpuZr alloy fuel similar to that used in the American fast neutron reactors.

Some section of fuel bars also contain rare earths in order to learn about their influence on the behaviour of the fuel during irradiation.

This experiment was conducted under contract to the CRIEPI.

Other experiments

MATINA 1A

The objective of the Matina 1A experiment is to study the behavior of new inert matrices for the incineration of actinides in heterogeneous mode. It was removed from the reactor during the mid-cycle shutdown for non-destructive testing conducted at Phenix in June and July 2004:

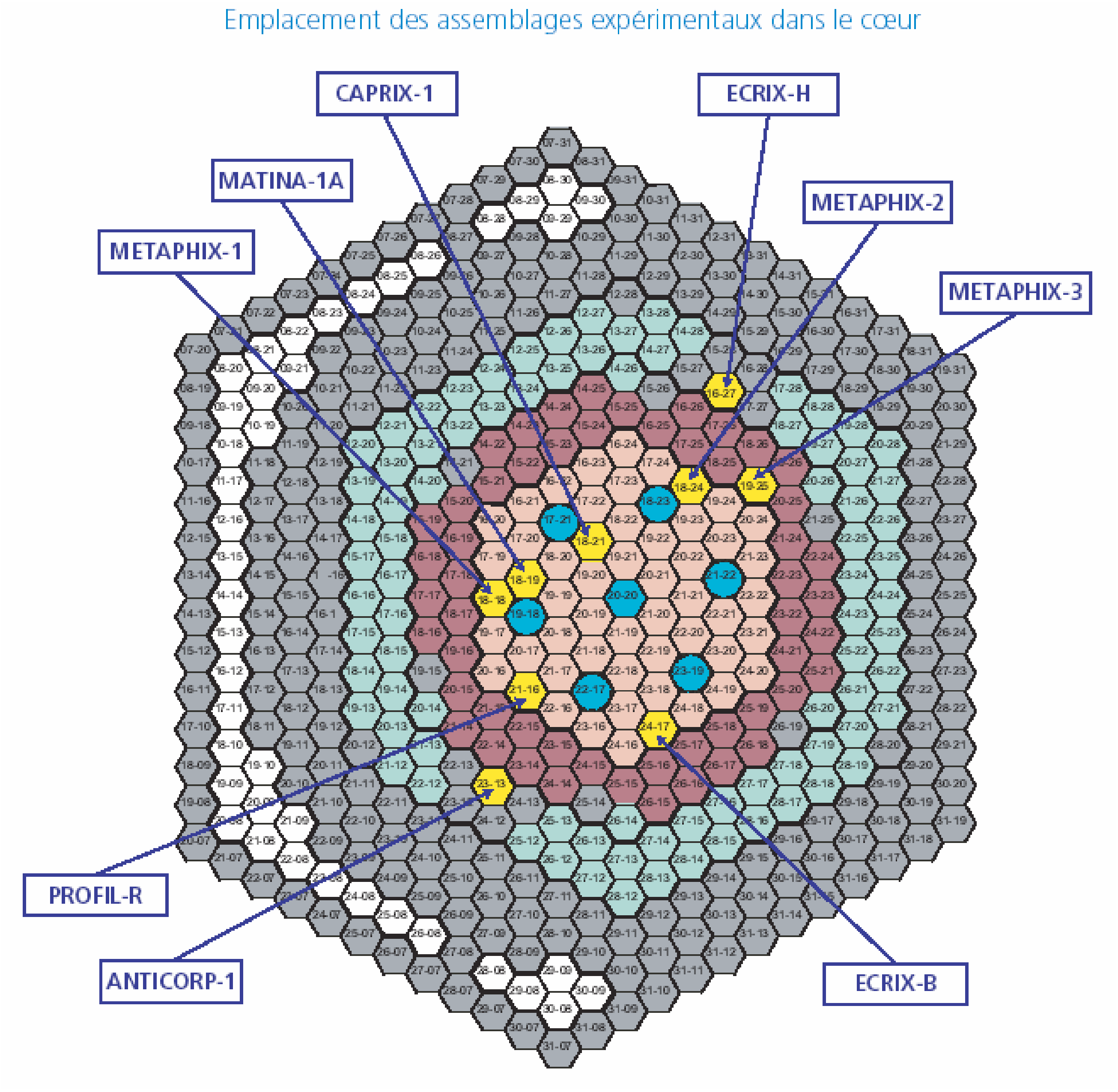
- Neutrongraphy
- Laser dimensional control,
- Spectrometry.

These measurements did not show any specific disturbances (swelling, fractures…) and will result in an initial validation of the setup.

CAPRIX

The Caprix experiment studied the behavior of very high plutonium content fuel (45 %) up to a burn-up rate of 10 at%. The experiment was placed back in the reactor for the 52nd cycle.

Figure 7. **Position of experimental assemblies in the core**



Conclusion

This diagram shows the position of all these experiments in the reactor core during the 51st cycle.

The Future Transmutation Programme

CAMIX COCHIX

The CAMIX-COCHIX experiment pursues the ECRIX experiment and will test the behaviour of optimized incineration targets. Evolutions in the experiment involve the mode of dispersion of the particles in the inert matrix and the stabilization of the crystalline structure containing the Americium.

This experiment will be irradiated in moderate spectrum in conditions similar to those in the ECRIX-H experiment.

PROFIL-M

PROFIL-M will add to the data obtained from the PROFIL-R experiment. Six pins, holding containers identical to those used in PROFIL-R, will be irradiated in moderate spectrum.

MATINA 2-3

MATINA 2 and 3 will add to the information obtained by the MATINA 1 and MATINA 1A experiments. The objective of the Matina 2-3 experiment is to study the behavior of new inert matrices and the new optimized concepts of targets to be used for the incineration of minor actinides in heterogeneous mode.

FUTURIX FTA

The FUTURIX/FTA experiment involves fuels dedicated to high minor actinide content incineration in sub-critical reactors. It will compare several types of fuel such as metallic fuel, nitrides and oxides, with and without uranium.

This experiment takes place in the framework of the international CEA, DOE, ITU and JAERI collaboration.

2 CEA pins (CECER)

$(Pu_{0.50},Am_{0.50})O_{2-x}$	In a MgO matrix
$(Pu_{0.20},Am_{0.80})O_{2-x}$	In a MgO matrix

4 DOE pins (Metallic and nitride)

$U_{0.24},Pu_{0.20},Am_{0.03},Np_{0.01},Zr_{0.52}$	Metallic
$Pu_{0.29},Am_{0.07},Zr_{0.64}$	Metallic
$(U_{0.50},Pu_{0.25},Am_{0.15},Np_{0.10})N$	Nitride
$(Pu_{0.21},Am_{0.21},Zr_{0.58})N$	Nitride

2 ITU pins (CERMET with macro masses)

$(Pu_{0.80},Am_{0.20})O_{2-x}$	Molybdenum matrix
$(Pu_{0.23},Am_{0.25},Zr_{0.52})O_{2-x}$	Molybdenum matrix

Conclusion

Following the extensive modernization program which was completed in 2003, the Phénix reactor was started back up in July 2003.

The 51st cycle ended on 8 August after very good operations throughout the year 2004.

The set of transmutation experiments in the reactor core will be providing the first validations of a wide spectrum of concepts.

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