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Advanced fuels for plutonium management in pressurized water reactors

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Abstract

Several fuel concepts are under investigation at CEA with the aim of manage plutonium inventories in pressurized water reactors. This options range from the use of mature technologies like MOX adapted in the case of MOX-EUS (enriched uranium support) and *COmbustible Recyclage A ILot* (CORAIL) assemblies to more innovative technologies using IMF like DUPLEX and advanced plutonium assembly (APA). The plutonium burning performances reported to the electrical production go from 7 to 60 kg $(\mathrm{TW\,h})^{-1}$. More detailed analysis covering economic, sustainability, reliability and safety aspects and their integration in the whole fuel cycle would allow identifying the best candidate.

1. Introduction

Light water reactors should dominate the production of electricity by nuclear systems during most of the current century. The future development of fast reactors needs for this period a flexible and economically acceptable plutonium management strategy.

The first generation of French reactors (900 MW), initially licensed to use enriched UO_2 fuel, were slightly adapted to accept plutonium. For more efficient use of plutonium in pressurized water reactors (PWRs) several fuel concepts are currently being examined. The objective of these innovative fuel concepts is to facilitate core management in a Pu multi-recycling strategy and to increase fuel burn up performances, keeping safety margins the same or better as for current UO_2-fuelled PWRs.

This paper summarizes the technical progress of the work on the fuel concepts like advanced plutonium assembly (APA), based on the use of inert matrix fuel (IMF), and *COmbustible Recyclage A ILot* (CORAIL) an heterogeneous assembly using $((Pu,U)O_2)$ MOX and (UO_2) UOX fuel rods, and MOX-enriched uranium support (EUS) an homogeneous assembly using MOX with EUS fuel rods. The MOX-EUS concept uses a homogeneous mixture of oxides (UO_2 and PuO_2) in each fuel rod. The CORAIL concept uses a heterogeneous arrangement of UO_2 rods and MOX rods, and the APA concept uses a heterogeneous arrangement of UO_2 rods and rods with PuO_2 in an inert matrix.

A comparative analysis based on a tentative set of criteria and the impact of the introduction of each type of fuel in the reactor park are presented.

2. Concept description

Several issues must be considered when planning to recycle Pu and minor actinides (MAs) in PWRs and in the future European pressurized reactor (EPR).

- The changes of reactivity must be accommodated. This is carried out by adjusting the quantity of Pu in the MOX fuel or by adjusting the ^{235}U enrichment. As the quantity of even-numbered Pu isotopes with

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high thermal capture cross-sections increases, the total quantity of Pu must be increased.

- The presence of Pu hardens the neutron spectrum, which reduces the worth of control rods and soluble boron. The control problem may be alleviated by limiting the Pu content, by redesigning the control rod assemblies, or by improved neutron moderation. In 900 MW electrical french PWRs, the boron concentration of the refuelling storage tank was increased from 2000 to 2400 ppm and the boron concentration of the boric acid makeup tanks were increased from 7000 to 7500 ppm. Four rod cluster control assemblies were added without significant economic penalties so that cores loaded up to 30% in MOX could be accepted. For the EPR, these control system improvements were included from the design phase, allowing the loading of 100% slightly moderated MOX assemblies.
- The safety margins change. Accidental voiding of coolant system also hardens the neutron spectrum. At high neutron energies all the isotopes of Pu can undergo fission, which increases the reactivity. From a study of six widely different mixes of Pu isotopes, it was found that the limiting total plutonium fractions (above which the void coefficient becomes positive) vary from 12.5% to 15% [1]. ^{239}Pu also has a much lower delayed neutron fraction (0.0021) than ^{235}U (0.0065), which makes kinetics control more challenging for cores with high Pu content.

Neutron thermalization in MOX or IMF with Pu fuelled PWRs may be improved by limiting the Pu mass in the assembly or by increasing the moderation ratio.

2.1. Options offered for Pu management

2.1.1. Near term technologies: assemblies with standard MOX fuel rods

Plutonium mass loading during multiple recycling is limited for safety reasons, so fissile materials needed for targeted burn-up is completed by LEU allowing keeping safety parameters within an acceptable range, two main options are considered. The first one consists of using only MOX fuel rods in the assembly with LEU. This is the MOX EUS concept [2], the second one is mixing in each assembly standard LEU UOX rods and MOX rods. This is the CORAIL concept [3].

2.1.1.1. MOX-EUS. With homogeneous fuel the Pu content in all assemblies is limited for safety reasons and to comply with fuel management constraints ^{235}U enriched UOX support is used This is the *MOX-EUS* concept [2]. The *MOX-EUS* concept uses all MOX rods (12% Pu), in a standard PWR fuel assembly configuration, maintaining safety margins similar to those of the all-uranium core and maintaining criticality by adjustment of the ^{235}U enrichment (2.5%). This option allows higher Pu masses loading in each core so lower number of reactors involved for the same Pu burning performance.

2.1.1.2. CORAIL. In heterogeneous assemblies with separate UO_2 and MOX rods, the Pu content in the MOX rods can be limited. This is the CORAIL concept [3–5]. The *CORAIL* concept uses a heterogeneous arrangement of MOX rods (PuO_2 in a depleted UO_2 matrix) and UO_2 rods in a fuel assembly. This reduces the neutron spectrum hardening and the required enrichment relative to the MOX UES concept. There are several ways to distribute the two types of rods in the assembly. Fig. 1 shows a cross-section in an example of the CORAIL concept with UO_2 rods surrounded by MOX rods. The MOX rods are placed in the periphery of the assembly so the control rods are inserted in less hardened neutron spectrum and keep their efficiency.

2.1.2. Future technologies: inert matrices and overmoderation

The use of inert matrices (U free fuels) improves Pu burning by avoiding their production by neutron captures in ^{238}U. Furthermore, modified rod geometries locally improving neutron moderation compensates the spectrum hardening induced by Pu.

2.1.2.1. DUPLEX. The DUPLEX assembly (17×17) consists of a heterogeneous arrangement of PuO_2 in an inert matrix surrounded by UO_2 rods. In the DUPLEX case, the standard geometry of the assembly is maintained, the IMF rods are deployed in the UOX assembly like in the CORAIL case (see Fig. 1).

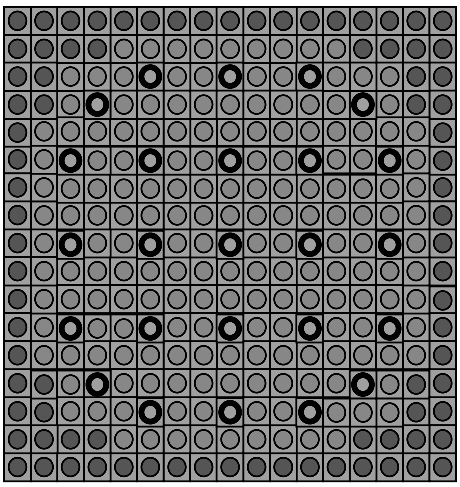

Fig. 1. Cross-section of a CORAIL (17×17) assembly. MOX fuel rods in the periphery, UOX fuel rods in the centre, and guide tubes.

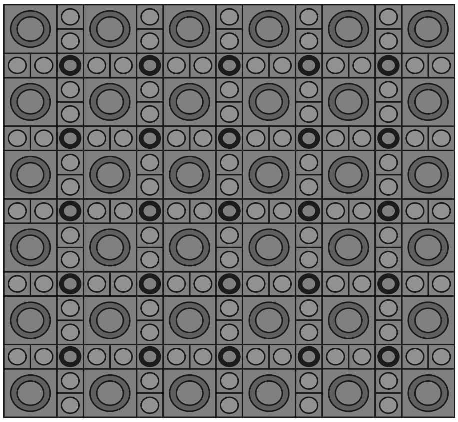

Fig. 2. Cross-section of an APA: annular Pu fuel rods version ●, UOX fuel rods in the centre ●, and guide tubes ●.

2.1.2.2. APA. The APA assembly consists also of a heterogeneous arrangement of PuO_2 in an inert matrix surrounded by UO_2 rods [6–8]. In the case of APA, the geometry of the assembly is changed in order to obtain a local over-moderation. Several materials and geometries are under investigation. In Fig. 2, four standard rods are replaced by an annular inert matrix Pu rod. In Fig. 3, about one third of standard rods are replaced by cross-inert matrix Pu rods. The annular fuel design facilitates enhanced spectrum thermalization, with a local moderation ratio ~8. The other APA rod designs, such as standard cylindrical or cross-shaped PuO_2-IMF rods, are being studied for various inert matrix candidates. The main physics results are very close for the different materials depending mostly on the moderation ratio.

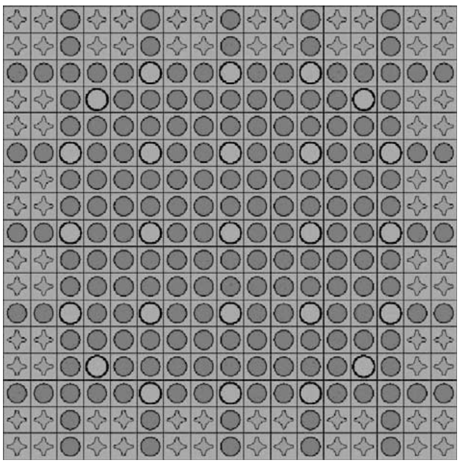

Fig. 3. Cross-section of an APA: crosses Pu fuel rods version ✧, UOX fuel rods in the centre ●, and guide tubes ●.

As IMF material, a cercer such as PuO_2–CeO_2 or a solid solution such as $(Pu,Ce)O_2$ was firstly considered due to their good behaviour in contact with hot water and the easy reprocessing. Other cercers like $MgAl_2O_4$–$(Pu,Zr)O_2$ could be used but they are still in progress of assessment towards fabrication, radiation resistance and reprocessing. A cermet fuel was also envisaged. For this option, the PuO_2 particles are included in a zircalloy metal matrix or in intermetallic material such as AlSi.

2.2. Comparison of advanced fuels for plutonium management

The main plutonium management characteristics calculated with the APOLLO 2 cell code [10] for various Pu recycling concepts in EPR are summarized in Table 1. The different concepts currently studied for plutonium management have a wide range of performances and technological maturity. Most of the parameters characterizing their performances are expressed per units of electrical energy produced. Their impact on a whole fuel cycle is obviously depending on their respective fraction introduced.

2.2.1. Plutonium and minor actinides mass balance

A comparison of Pu burning performances and associated MAs production is presented in Fig. 4. The Pu balance is a good indicator of the flexibility offered by the assembly to adjust Pu stockpiles to a desired value.

The APA concept has the best performances in terms of Pu burning due to the use of IMF and local over-moderation. This explains why less then 30% of the reactor fleet is enough to burn the plutonium produced by the other standard UOX cores and obtain the Pu inventory stabilization. The MAs masses produced by the MOX-EUS, CORAIL and APA assemblies are in the same range even if MOX-EUS and APA produce the maximum values for curium.

Clearly the IMFs offer the best performances with a reasonable MAs production. Pu consumption in a single APA IMF rods may reach 90%. Even if the standard MOX has similar performances to APA to burn Pu, it must be noted that this is obtained during the first cycle. For this fuel, multi-recycling would be much more difficult than for the other concepts.

2.2.2. Adapting the fuel cycle

After multiple recycling (several decades), residual plutonium is supposed to be used for fast reactor fuel fabrication. The percentage of fissile isotopes is a good indicator of the quality of this residual plutonium for further utilisation in nuclear fuels. Plutonium quality data (%Pu fissile) after multiple recycling are given in Table 1.

Fig. 5 shows that percentage issued from different assemblies in a theoretical equilibrium state. The initial

Table 1
Characteristics of different assemblies

Assembly		UO_2	MOX	MOX-EUS	CORAIL	DUPLEX	APA
Number of cycle	–	0: open	One[a]	Equilibrium	Equilibrium	Equilibrium	Equilibrium
Number of batches	–	4	4	4	4	4	4
Burn up	$GW\,d\,t^{-1}$	60	60	60	60	60[b]	60[b]
^{235}U in UO_2 rods[c]	%	4.7	–	–	5.2	5.0	5.0
^{235}U in Pu rods[c]	%	–	0.25	3.9	0.25	–	–
Pu in Pu rods [c]	%	–	12	12	11.3	100	100
Number of UO_2 rods	–	–			180	180	120
Number of MOX rods	–	0	264	264	84	–	–
Number of IMF rods	–	0	–	–	–	84	36
U assembly weight	kg	518	456	456	499	353	235
Pu assembly weight	kg	–	62	62	19	19	23
Pu mass balance	$kg\,TW^{-1}\,h^{-1}$	26	−70	−58	−7	−25	−60
Np mass balance	$kg\,TW^{-1}\,h^{-1}$	1.8	0.4	1.1	1.6	1.5	0.4
Am mass balance	$kg\,TW^{-1}\,h^{-1}$	1.6	14.1	17.5	6.1	5.1	6.9
Cm mass balance	$kg\,TW^{-1}\,h^{-1}$	0.3	2.9	3.9	1.7	2.9	4.8
Total MA mass balance	$kg\,TW^{-1}\,h^{-1}$	3.7	17.4	22.5	9.4	9.5	12.1
Pu quality after multiple recycling							
Fraction of fissile isotopes	%	–	–	49	47	40	30
Natural U needs	$t\,TW^{-1}\,h^{-1}$	19.7	0	14.2	15.0	14.4	9.9
U enrichment needs	$SWU\,TW^{-1}\,h^{-1}$	14 900	0	10 100	11 900	11 300	7900
Fraction of the reactor park to be loaded to stabilize Pu inventory	%	–	–	31	79	51	30
Number Pu fuel rods (MOX or IMF) to be fabricated per year to stabilize the Pu inventory	–	–	–	128×10^3	104×10^3	66×10^3	17×10^3

[a] Mono-recycling.
[b] Equivalent UO_2.
[c] At beginning of cycle.

plutonium is an 'average' plutonium issued from the French inventory by 2016 having 64% fissile isotopes. Neutronics studies are planned to identify the corresponding penalties on fast reactor core designs. Nevertheless, Pu to be used in future fast reactor cores will result from a mixture of various plutonium qualities.

Concerning uranium and enrichment needs, the CORAIL concept, with respect to UOX assemblies, reduces the natural uranium by about 24%. The APA assemblies allow more the 50% savings (Fig. 6). The uranium enrichment needs savings range from 20% (CORAIL) to 47% (APA) compared to the reference UOX fuel cycle.

From fabrication and reprocessing needs point of view, the APA concept in the annular rod version requires much lower number of Pu fuel rods to be manufactured and reprocessed.

2.2.3. Core management

All the concepts considered are compatible with standard PWRs *core/fuel management schemes*. The APA assemblies require particular care on core loading patterns due to the very high heterogeneities during irradiation between standard and Pu fuel rods. Due to the power decrease at the end of each irradiation cycle, and the low temperature of the fuel, the APA concept minimizes gas fission products release compared with standard MOX fuels.

Concerning the behaviour under incidental/accidental conditions, the following may be stated. EPR design

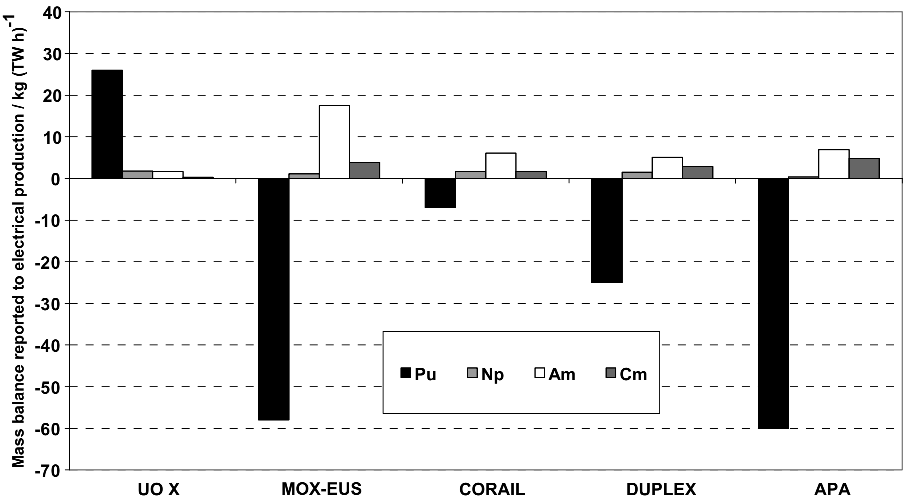

Fig. 4. Pu and MAs balance.

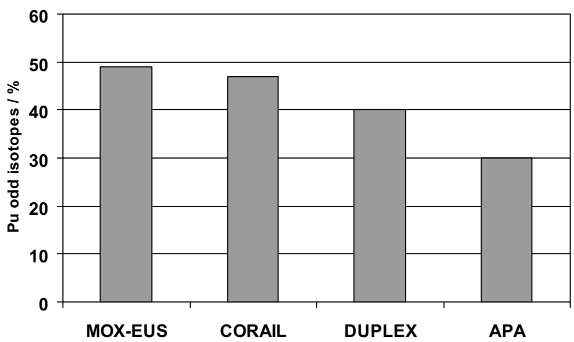

Fig. 5. Pu quality in theoretical equilibrium state.

can easily accommodate to plutonium loadings. Nevertheless, the whole safety analysis must be carried out especially for the APA concept where non-standard materials and geometries are considered.

Nevertheless, some difficulties could arise during transition fuel cycles (heterogeneous cores) from a 100% UOX core to a 100% advanced fuel core like unacceptable power distribution or undesirable hydraulic interactions between standard and advanced assemblies. There is a need to adapt the heterogeneous core loads during transition cycles. Obviously, MOX-UES, CORAIL and DUPLEX assemblies could more easily accommodate to this transitions phases than APA.

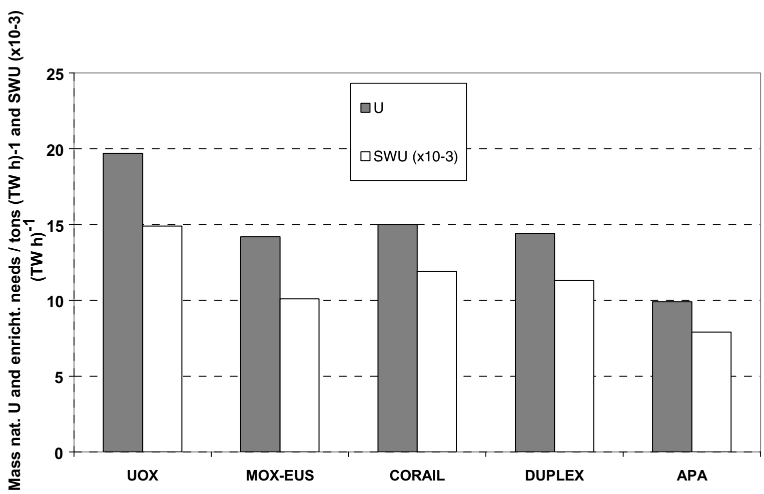

Fig. 6. Natural uranium need (t) and enrichment requirements (SWU $\times 10^{-3}$).

3. Park scenario and potential for meeting Generation-IV goals

3.1. Park scenario

The fuel concepts discussed in the previous section were assessed in a simplified nuclear fleet scenario starting from the current situation up to pseudo steady state [9]. Assuming a nuclear park with 60 GW electrical producing 400 TW h per year, Fig. 7 shows the evolution of the Pu inventory in the cycle (reactors and facilities) for the following PWR scenarios: open cycle, Pu once through cycling, and Pu multi-recycling. In 2050, the open cycle has about 630 tons of Pu, and mono-recycling has about 520 tons. For multiple recycling the Pu inventory varies between 230 tons (APA) and 430 (*MOX-EUS*) tons according to the fuel assembly concept selected. High temperature reactor (HTR) performances are added for comparison with PWR options.

3.2. Potential for meeting Generation-IV goals

The advanced fuels for Pu multi-recycling in PWRs were assessed against the Generation IV goals:

- Sustainability
- Safety and reliability
- Economics

The benefits or liabilities are evaluated relative to a typical Generation III reactor once-through uranium fuel cycle.

3.2.1. Sustainability

Advanced fuels for Pu recycling have the advantage of better using the nuclear resources in recovering the Pu energetic potential rather than managing it like a waste. The savings in natural uranium and SWU on the whole reactor park due to the use of advanced fuels in PWR are 15–25% in comparison with the UO_2 open cycle. Recent studies demonstrated the potential for MA incineration (americium and curium). For example, using APA fuel in 40% of the PWR park could stabilize the (Pu + Am + Cm) inventory in the cycle. Build-up of actinides (such as ^{238}Pu) with high self-heating and neutron emission reduces the attractiveness of Pu diversion for weapons use and makes detection of stolen materials easier, even if the use of reprocessing facilities provides another pathway where diversion could occur.

3.2.2. Safety and reliability

The Pu fraction in the core is limited by safety constraints to about 30% in present PWRs, due to the lower delayed neutron fraction, harder neutron energy spectrum, reduced effectiveness of control rods and boron, and design modifications needed to maintain a negative void coefficient. These issues could affect reactivity control reliability, but they can be accommodated by proper design.

In high multi-recycling regimes the buildup of higher actinides could increase the dose to workers during refueling or during an accident.

The core would have more TRU present than the once-through fuel cycle core. This would increase the radiotoxicity source term available for potential release during a severe accident, but the increase would prob-

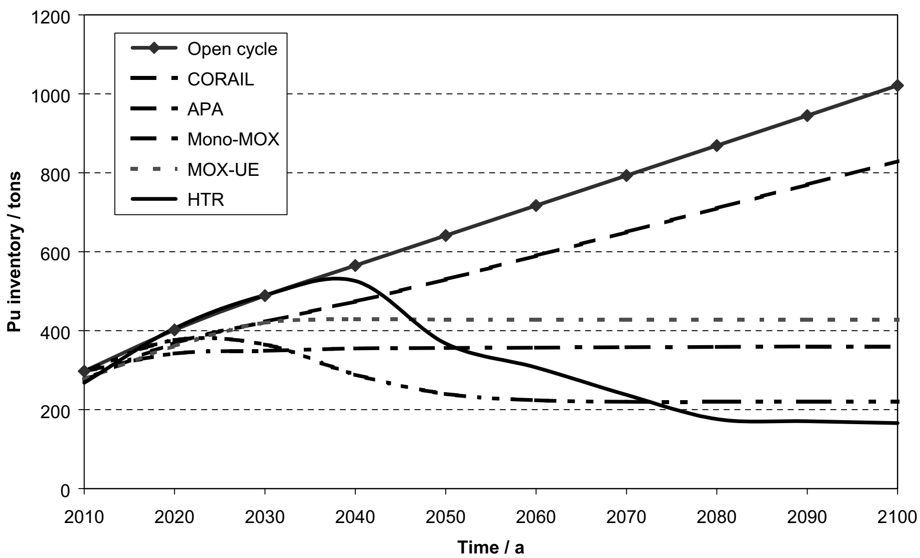

Fig. 7. Pu inventory for various 400 TW h electrical reactor parks.

ably not change the degree of offsite emergency response required for present LWRs.

Studies of the offsite consequences of severe accidents are incomplete. With similar fuel forms, cladding, and power density, the release of volatile fission fragments would probably not differ greatly from that in present LWRs.

3.2.3. Economics

These fuel cycles result in lower costs for uranium and enrichment. The costs for actinide waste disposal may be reduced if the actinides are multiply recycled and high decontamination factors can be attained during reprocessing.

The reprocessing plant and remote fabrication plant operations would increase the fuel cycle costs. With the APA concept, however, only about 1/4 of the total number of rods would contain recycled Pu, and the rest could be ordinary UO_2 rods.

The fuel fabrication and reprocessing could be more expensive than current LWR once-through fuel fabrication, especially if actinides with high self-heating and neutron emission rates were present in the recycle fuel.

3.2.4. Strengths and weaknesses identified in the Generation IV assessment

The strengths of Pu recycle in PWRs are:

- Enhanced uranium utilization.
- Reduced uranium enrichment requirements facilitated by recycle of fissile Pu.
- Reduced waste especially if actinides are recycled with high decontamination factors.
- Possibility to burn surplus Pu.

The APA concept could produce Pu with a high content of actinides with self-heating and neutron emission, making that Pu unattractive for diversion.

The weaknesses of Pu recycle in PWRs are:

- Reprocessing facilities open another pathway for diversion of nuclear materials.
- Added fuel-cycle cost of reprocessing facilities.
- More expensive fuel fabrication facilities.

4. Conclusions

This range of advanced assembly concepts shows that, from the reactor core physics aspect, solutions for multi-recycling of plutonium in PWRs should be possible. Options range from a concentration of Pu in a limited number of rods (CORAIL, DUPLEX, APA), with or without recourse to an inert matrix, to total dispersion of Pu throughout the assembly (MOX-EUS), with various consequences on manufacturing, Pu consumption and MA production.

For all these concepts, studies are underway in order to make a decision concerning their feasibility. All of these solutions require technological feasibility and qualification for the manufacturing, from studies of their behaviour under irradiation, etc. before a decision can be made concerning their technical and economic utilisation in the park. It is expected that MOX based fuels could be ready for deployment by 2015 and IMFs by 2025.

Multiple recycle of Pu in PWRs could enhance nuclear power by appreciably increasing the energy available from uranium resources. It could also reduce the amount of the high level waste to be disposed.

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