Improving Actinides Recycling, a Key Step towards a More Sustainable Nuclear Energy

C. Poinssot, S. Grandjean, M. Masson, B. Boullis

Improving Actinides Recycling, a Key Step towards a More Sustainable Nuclear Energy – 15617

Christophe Poinssot *, Stéphane Grandjean*, Michel Masson*, Bernard Boullis **
* CEA, Nuclear Energy Division, RadioChemistry & Processes Department, CEA Marcoule, BP11, F-30207 Bagnols-sur-Cèze Cedex, France.
** CEA, Nuclear Energy Division, Nuclear Energy Division, Innovation and Nuclear Support Division, CEA Saclay, F-91191 Gif-sur-Yvette Cedex, France.

ABSTRACT

Nuclear energy is thought to be one of the energy sources that could help mitigating the global climate change together with the renewables, due to its low green-house-gases emissions, its reliability and its high load power. However, nuclear energy will only develop if future nuclear energy systems meet the criteria of sustainability in terms of durability, bearability and liveability. Nowadays, many countries chose the so-called once-through cycle which basically considers spent nuclear fuel as a waste, whereas others reprocess their spent fuel to recover the energetically-valuable material Pu (and partially U) to produce Mixed Oxide Fuel (MOX) to be irradiated in a second cycle (twice-through cycle). Both options can be considered relevant, fitting diverse criteria; but if we consider a long lasting prospective for nuclear options, the once-through cycle sustainability is clearly lower (low consumption of natural resource, no recycling of potentially valuable energetic material, larger volume of waste …). Reversely, twice-through cycle can be seen as a first step towards an improved sustainability with a partial recycling of U/Pu and the reduction of the waste volume and toxicity. However, it is likely not sufficient for the long-term and implementing more efficient recycling is a key-driver to improve the environmental friendship, the economics and the social acceptability of nuclear energy.

Future nuclear fuel cycles will have to better preserve the natural resource by improving the actinides recycling, to minimize the volume and toxicity of nuclear waste, to ensure their long-term confinement and to prevent any proliferation-risk. Such a gradual and progressive process calls for implementing appropriate technologies, either for reactors but also for fuel treatment/recycling facilities. This paper will describe the successive step that would allow improving the overall sustainability. Indeed, recycled Pu can be once recycled in MOX fuel feeding similar PWRs (Pressurized Water Reactor). Together with the recycling of reprocessed uranium, this first step already allows preserving ~17% of the natural U resource and to confine ultimate wastes without Pu in a tailored durable wasteform. Subsequent Pu multirecycling requires introducing Fast Reactors (FR) to efficiently use U-238 available in natural and depleted uranium stockpiles. In parallel, recycling processes would have to be adapted for treating spent LWR and SFR MOX spent fuels, which is currently under study thanks to an ambitious R&D program dedicated on MOX treatment and recycling. For the longer term, minor actinides recycling would allow decreasing the waste burden towards future generations: (i) Am-recycling leads to decrease the waste residual heat-power and increase the repository density and lifespan, (ii) Cm-recycling leads to decrease waste long-term toxicity. The stepwise implementation of these different processes defines a path towards sustainability for nuclear energy that are described in this paper from the fuel cycle viewpoint.

INTRODUCTION

Driven by the anticipated population increase as well as the economic development of new countries as Bresil, Russia, India and China, the energy demand is anticipated to significantly increase in the future, in the order of a factor two by 2050 according to the different energy scenario, *e.g.* International Energy Agency. If based on the current energy portfolio, such an increase would lead to a very significant increase

of the green-house gases (GHG) release in the atmosphere, yielding to a likely unacceptable temperature increase at the earth surface. Meeting such needs while mitigating the global climate change hence requires promoting an energy transition towards new energy models that release much lower GHG in the atmosphere, i.e. which are less dependent on fossil fuels (coals, gas or oil). Both renewables energies and nuclear energy should contribute to such new energy portfolio, based on their respective specificities and strengths. Finally, future energy modes will only develop if they succeed to be economically effective, to be environmental-friendly and to be accepted by a significant part of the population. These three pillars which define the sustainability are the key drivers and criteria for the selection of the most promising technology for the future. Future nuclear energy has therefore to be developed in such a perspective. This paper will describe how actinides recycling may be a key target to improve the sustainability of nuclear energy in the field of resource preservation and social acceptance [1].

IMPLEMENTING THE URANIUM AND PLUTONIUM RECYCLING, A MAJOR STEP TOWARDS RESOURCE PRESERVATION

Why Recycling Uranium And Plutonium?

Current nuclear fuel cycles, either once-through cycle that consider spent nuclear fuel (SNF) as a waste, or twice-through cycle that recycle once Pu in MOX fuels, do not meet the sustainability criteria. Indeed, both fuel cycles do not fully preserve the uranium natural resource for future generations, which is also referred to as inter-generational equity. After irradiation, spent nuclear fuel still contains ~96% of U and Pu that are potentially-valuable material to produce electricity, and should be recycled in view of sustainability.

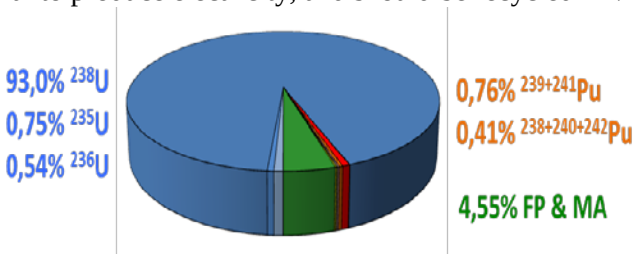

Fig.1: Relative composition of UOX fuel after 4y. irradiation (47.5 GWd/t) in PWRs. This clearly evidences that spent nuclear fuel still contains a large amount of energetic valuable material as uranium and plutonium [2].

This means that the total amount of uranium needed to feed reactors and produce electricity is much higher than the amount which is effectively consumed in reactor through fission or neutron capture. This defines the efficiency in the natural resource consumption. This figure is currently lower than 1% as evidenced by the global uranium balance in country like France for instance: From 8000t of natural uranium, only 1100t is used to manufacture nuclear fuel (due to enrichment) and feed the 58 reactors to produce 80% of the French electricity, among which only 60 to 70t is effectively transformed in fission products or minor actinides. Without any recycling, more than 99% of the initial natural uranium would be considered as waste, which is a non-sustainable way of using such a natural resource. This situation supports the research for an efficient recycling approach.

The Twice-Through Cycle, Or The Implementation Of The Plutonium Monoreycling In LWR MOX.

First step in this direction has been implemented for decades in France by developing and implementing at the industrial scale the Pu and U mono-recycling through their recovery in the AREVA La Hague plants

and their recycling in PWR reactors as MOX fuels.

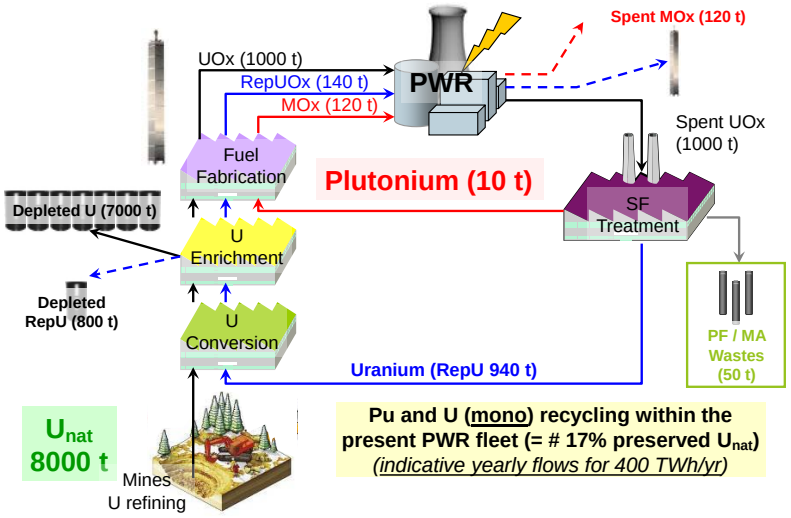

Fig. 2: Illustration of the French twice-through cycle in which Pu is recycled in MOX fuel [3,4].

More than 26,000 tHM of UOX spent fuels have been treated in the UP2 and UP3 plants at La Hague. The corresponding Pu inventory has been recycled as MOX fuels produced in the AREVA MELOX plant (Marcoule) and used in 22 PWR French reactors. Indeed, over 6,000 MOX fuel assemblies, *i.e.* 1800t, have been produced at MELOX.
Treatment and recycling is today mature, efficient, safe, clean and cost-effective technologies. It requires initial dissolution of the spent nuclear fuel to access the nuclear materials and subsequent partitioning the valuable elements, U and Pu, from the ultimate waste, mainly FP and MA, via the PUREX process. In the La Hague plants, both recovery yields and purification levels are very high (up to 99.9 % of uranium and plutonium recovered with decontamination factors of 10^6 or more). A final conversion step, involving precipitation of intermediate oxalate (Pu) or peroxide (U), produces oxide powders compatible with the fabrication specification of the fuel fabrication process.

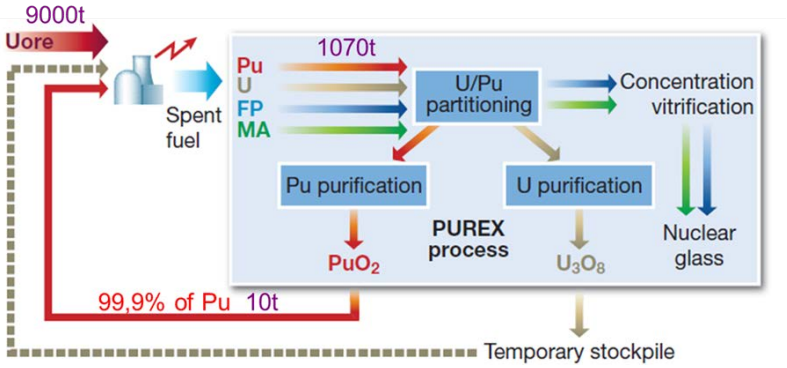

Fig.3: Schematic of the twice-through fuel-cycle with a focus on the separation steps. This fuel cycle allows recycling once plutonium in MOX fuels and is currently implemented in France [5].

By comparison to once-through open cycle, Pu mono-recycling in MOX already contributes in enhancing the sustainability of nuclear energy by:

- increasing the efficiency of the natural uranium resources of #17% due to the fission of $^{239+241}$Pu in MOX instead of ^{235}U in UOX.

- saving the corresponding enrichment energy cost, which is quite significant; in particular when using the first generation gaseous diffusion process.
- drastically decreasing the volume and long-term radiotoxicity of the final waste due to the recycling of Pu which is not in the waste anymore (Fig. 4). It hence contribute to decrease the Pu stockpile and limit the risk of diversion.
- Permitting an efficient conditioning of the ultimate waste, *i.e.* fission products and minor actinides, which are confined in a tailored borosilicate nuclear glass, the lifetime of which has been demonstrated to be in the range of 10^6 years in French future repository conditions.

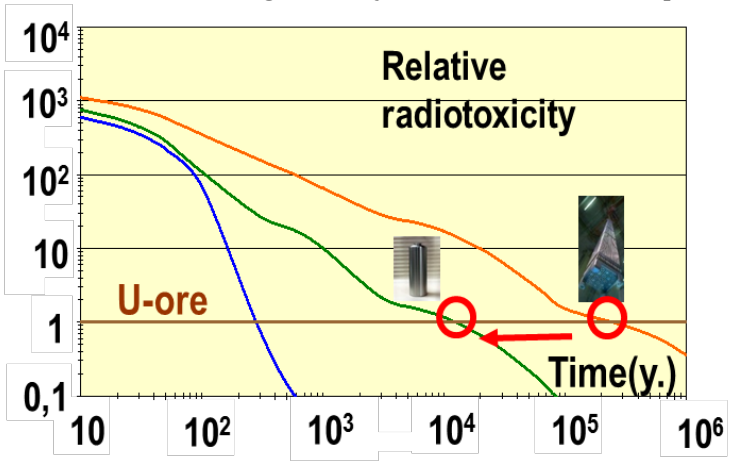

Fig.4: Evolution of the radiotoxicity of spent nuclear fuel (orange) and nuclear glass (green) relative to the radiotoxicity of natural uranium ore (brown). Mono-recycling Pu already allows decreasing the “lifespan” of the waste from some hundreds thousands years to some tens thousands of years [5].

The 4th Generation Fuel Cycle, Or The Implementation Of The Plutonium Multi-Recycling In FR-MOX.

Increasing further the efficiency of natural uranium resource consumption requires overcoming the impossibility of efficiently using U-238, representing 99.27% of natural uranium in the current PWR. Since U-238 is not fissile, the only way is to promote the "fertilization" of U-238 by neutron capture in order to produce Pu-239 which is fissile and thus implement Pu multi-recycling. Compared to thermal neutron spectrum, fast neutron spectrum increases the relative capture of neutrons by U-238, producing plutonium isotopes which are all fissile in fast neutron reactors. For example, the ratio of fission to capture cross sections of Pu-238, Pu-240 and Pu-242 are respectively increased in fast neutron spectrum by factors 22, 250 and 36 by comparison to the thermal neutron spectrum. Fast neutron reactors could hence potentially be able to use more than 80% of the uranium natural resource instead of 0.6 – 0.7% in the current PWR, which represents a very significant improvement towards sustainability. This would also significantly reduce the natural uranium resource consumed since the existing stockpiles of depleted uranium could be used, which could represent up to several thousand years of uranium lifespan.

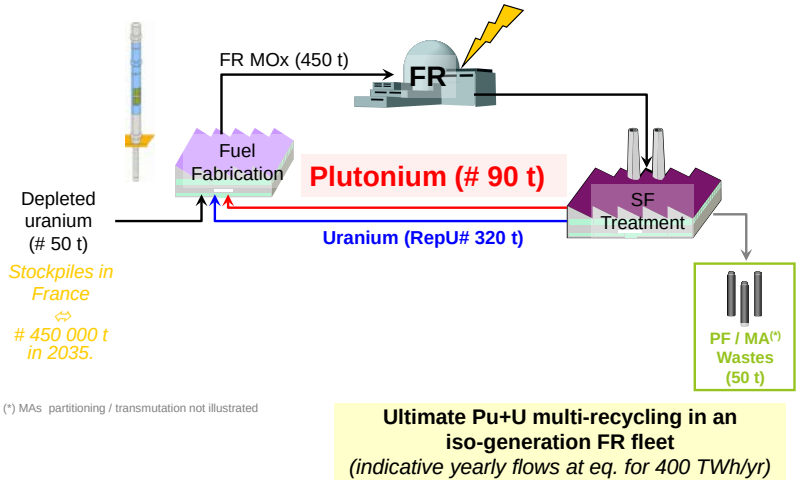

Fig. 5: Illustration of a potential multiple-through cycle in which Pu is multi-recycled in Fast Neutron Reactor MOX fuel.

In terms of fuel cycle, the feasibility has already been proven since treatment of FR MOX fuels and subsequent Pu multi-recycling has been demonstrated at the industrial scale in France where 25 tHM of FR spent MOX fuels have been reprocessed mainly in APM (Marcoule) and UP2-400 (La Hague) plants during the 1980's and 1990's as illustrated in Fig.6.

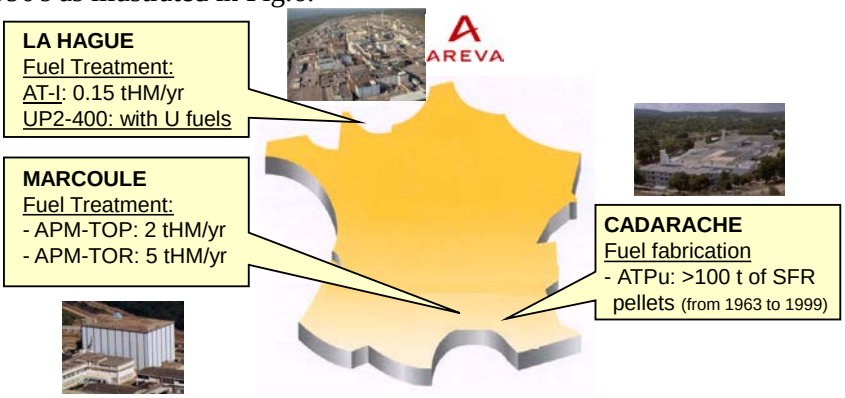

Fig.6: French experience in fast neutron reactors fuels fabrication and treatment.

In terms of fuel cycle, current treatment processes can be adapted to such types of fuel as described further below.

Key R&D Issues Associated To The Progressive Deployment Of Pu Multi-Recycling

If industrial campaigns of PWR MOX treatment were carried out at the AREVA La Hague plant without noticing any significant difference compared to UOX processing, optimizations could be required for higher treatment capacities. The main issue related to the treatment of PWR/FR MOx fuels is the higher Pu content by around one order of magnitude (cf. Fig.7) that impact all along the treatment process. Key R&D issues can be described as follow:

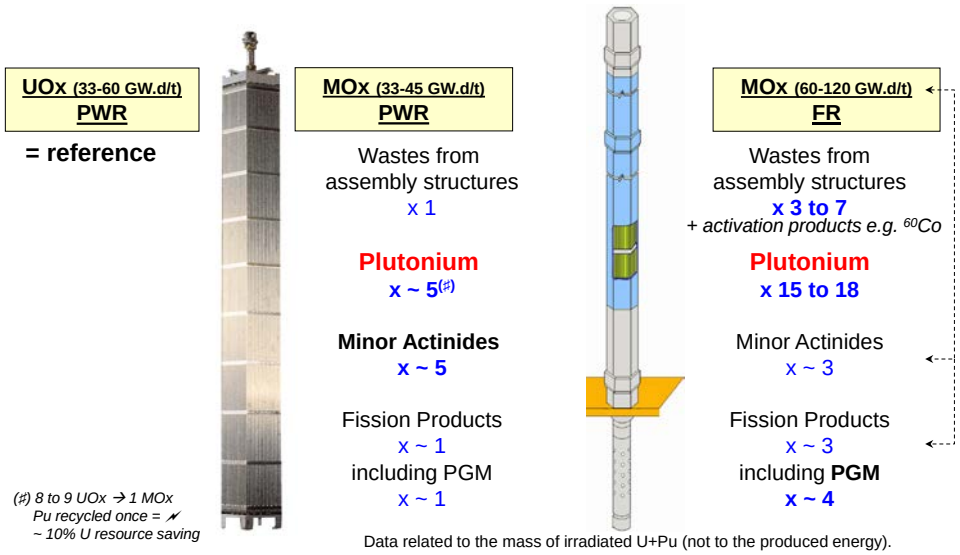

Fig.7: Main differences between spent LWR and SFR fuel (indicative data). 5 y cooling, no transmutation. Numbers in blue are the content increase for each element/family by comparison to the composition of a current UOX spent fuel (33-60 GWd/t).

- Regarding the head-end step (shearing and dissolution), main issues are related to the potential residual Pu in the solid wastes (cladding, insoluble particles) that could become a limitation depending on the chosen process and/or technologies. R&D is then devoted to limit these Pu losses at the required trace level. Dedicated digestion processes of insoluble fine particles associated to an advanced rinsing of the end caps/cladding is therefore under development.
- Regarding the partitioning step, main issues are related to the high Pu content that could lead to high consumption of chemical reagents with the PUREX process and a more complex mastering of criticality risk. Improvements are currently considering (i) the adaptation of the Pu and U co-extraction process to higher Pu/U ratio, (ii) simplifying the process by mitigating the consequences of extensive use of redox reagent due to higher Pu concentrations.
- Regarding the fuel fabrication, if powder metallurgy remains the reference process thanks to many decades of industrial feedback, R&D aims to develop an advanced and more integrated treatment/recycling scheme that could prevent the occurrence of fine dusts build-up. The use of pre-mixed Pu and U oxides produced by co-conversion and the recycling of scraps by hydrometallurgy in the fuel treatment facility could significantly simplify the fabrication of FR fuels at an industrial scale.
- Finally, as for the ultimate wastes, the main evolution is firstly the specificity of the structure material of FR MOx fuel assemblies: stainless steel vs Zircaloy for cladding, neutron protection material… Current process/technologies and management could be adapted or modified depending on the proportion of FR fuels to be treated with more conventional PWR fuels. Increased contents of PGM, potential corrosion products from stainless steel (FR cladding), neutron poison (if needed due to higher Pu flows, e.g. Gd) could motivate adapted or novel formulations of glass to confine these elements with the main stream of FP.

IMPLEMENTING THE AMERICIUM RECYCLING, A POTENTIAL STEP TOWARDS AN OPTIMISED WASTE REPOSITORY

Americium is the main contributor to the long-term waste decay heat power. Recycling Am would therefore allow significantly decreasing the waste residual heat which is the main parameter for defining the waste density within a repository. Indeed, host rock has to be maintained under a threshold temperature to ensure that it would not transform over time and modify its properties. Decreasing the residual heat would hence allow to design a denser repository. Calculations performed based on the French repository site and design demonstrated that one order of magnitude of gain could be reachable. It would therefore allow a very significant preservation of the repository resource [2].
Recycling Americium requires developing separation process to partition Am from the other actinides and lanthanides. Based on the long-lasting skill developed in France, CEA has developed in 2008 a new separation scheme, referred to as EXAm process, to selectively strip Am from the light lanthanides

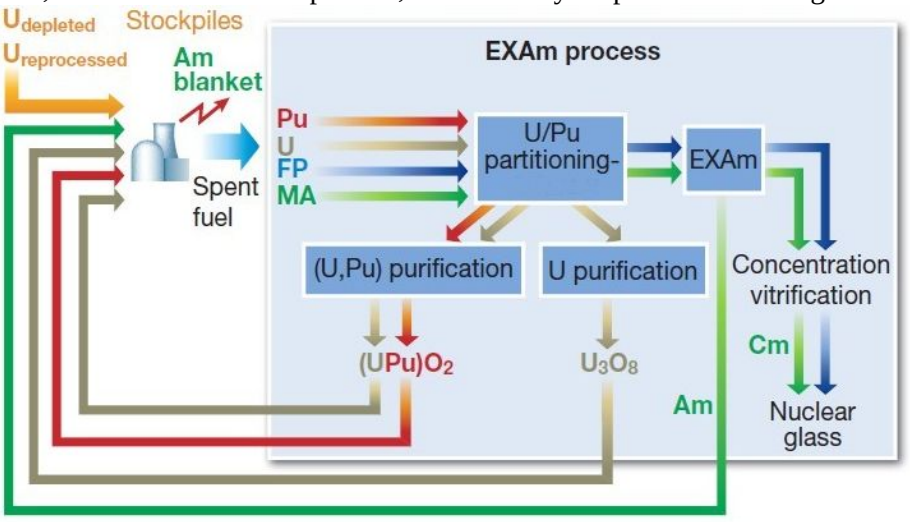

Fig.8: Schematic of the different actinides fluxes within a fuel cycle in which the EXAm separation process would be implemented (adapted from [5]).

This process has been demonstrated to be quite efficient based on an experiment performed in the CBP in ATALANTE on a few kilograms of spent nuclear fuel: 98.5% of Am has been recovered and decontaminated over Cm with a factor of 500.

IMPLEMENTING THE MINOR ACTINIDES RECYCLING, A CONTRIBUTION TO AN INCREASED ACCEPTABILITY

Opinion polls evidenced in many countries that the main reasons for which people fear or even reject nuclear energy are (i) safety and preservation of major accidents leading to land contamination and health impact, and (ii) nuclear waste that are seen as a strong pollution source for the future. As for safety, reinforcing the robustness of the facilities and processes, and deepening the understanding of the physical and chemical processes are all along the nuclear fuel cycle key contributions. Safety is a very general research issue that any development has to consider.
Regarding waste management, one of the main reason for the bad perception of public opinion lies in the very long lifetime of some of the nuclides which are within the waste. Dealing with millions years is all but common sense, and is not understandable for most of the people since it goes well beyond the Human history. Therefore, any process that could decrease waste lifetime could lead to a significant gain in nuclear energy acceptance. The main contributors to long-term toxicity of the waste are successively plutonium, which is already recycled and not in the waste anymore, minor actinides (americium and curium) that

dominate the long-term radiotoxicity (Fig.8).

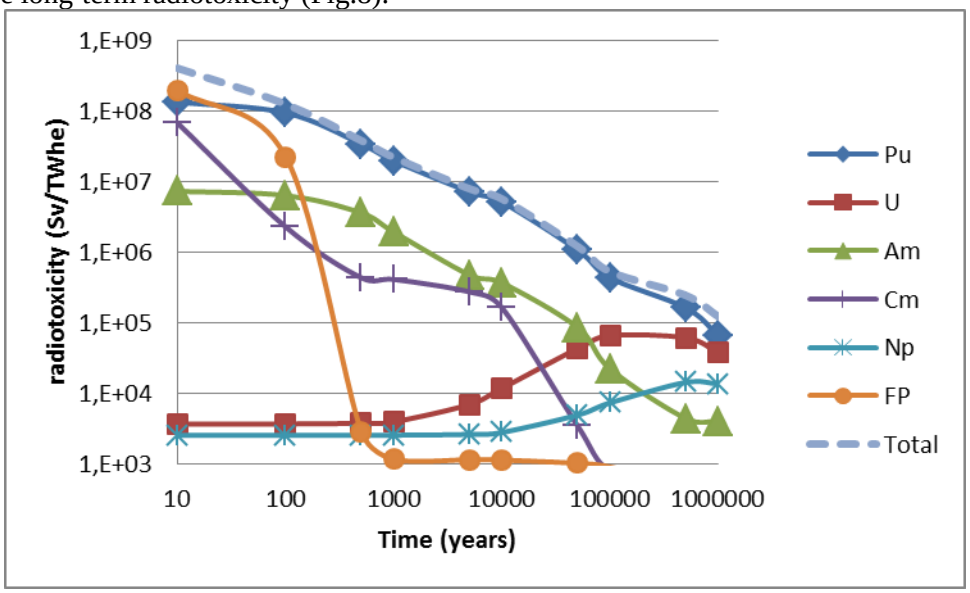

Fig.8: Evolution of the radiotoxicity of spent nuclear fuel (Sv/TWh electric) as a function of time, with the contribution of the different radionuclides. This figures clearly illustrates that long-term toxicity (>300y.) is dominated by the minor actinides, Americium (Am) then Curium (Cm) [1].

Recycling these elements and transforming them in lower decay time elements by transmutation would severely decrease the waste lifetime and improve the public perception.
Several options can be envisaged for the recycling of all the minor actinides: the homogeneous recycling of MA in the whole fuel core at low concentration, or the heterogeneous recycling of MA in dedicated target or blanket at higher concentrations. Among others, France has supported an extensive R&D program to develop specific and efficient partitioning processes to recover the minor actinides, either altogether in the so-called homogeneous recycling thanks to the GANEX separation process, or in a dedicated flux in the heterogeneous recycling thanks to the DIAMEX/SANEX separation process. The efficiency of both GANEX and DIAMEX/SANEX has been demonstrated on genuine spent nuclear fuel dissolution solution in the ATALANTE hot laboratory (see for instance [7], this conference).

CONCLUSION

Improving the sustainability of nuclear energy will require improving its economical effectiveness, its environmental-friendship and its social acceptance. In this context, recycling the actinides is the cornerstone of any sustainable nuclear energy system, as it allows to much better use and preserve the uranium natural resource for the future generations, it improves the overall environmental footprint and it may help to increase the social acceptability by decreasing the waste volumes, lifetime and toxicity and reducing the significance of the final repository.
However, recycling the actinides to increase nuclear energy sustainability is a long-standing path that requires a stepwise approach to allow a progressive industrial deployment to reduce any industrial risk as described in Fig.9:

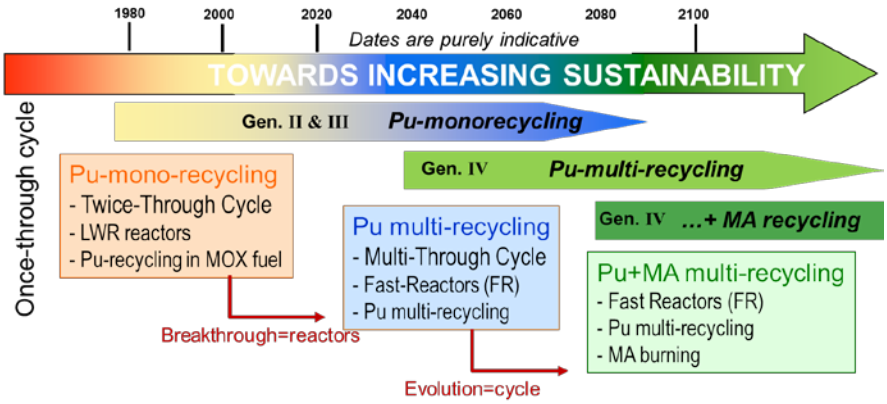

Fig.9: Roadmap towards sustainable nuclear energy by improving the actinides recycling [1].

- First of all, optimize the consumption of natural resource to preserve them for future generations and hence guarantee the energetic independence of the countries (no uranium ore is needed anymore). The current twice-through cycle of Pu implemented by France, UK, Japan and soon China is a first step in this direction and already allows the development and optimization of the relevant industrial recycling processes. It also allows a major improvement regarding the conditioning of the ultimate waste in a durable and robust tailored long-term matrix, *i.e.* nuclear glass.
- Second, the multi-recycling of Pu thanks to the deployment of FR is a major step towards a better use of uranium natural resource and a strong improvement of the environmental footprint linked to the reduction (or suppression) of mining activities.
- Third, recycling americium which is feasible with FR would lead decreasing the ultimate waste residual heatpower and allow designing a much denser repository (waste packages could be disposed closer to each other), saving hence the scarce repository resource. This would mean limit the burden towards the future generations and allow a single repository to accommodate waste produced by several centuries of nuclear reactors.
- Finally, recycling the whole minor actinides inventory could be an ultimate option for the far-future which would lead to a strong decrease of the waste long-term toxicity, allowing having it down to that of uranium ore within a few centuries. It would bring the waste issue back within the human history, which should promote its acceptance by the social opinion.

This stepwise approach defines a clear roadmap towards sustainable nuclear energy which would allow a very efficient use of natural resource and its preservation for future generations, a strongly-reduced volume of less radiotoxic waste to be disposed of in a reduced repository. Actinides recycling is hence one of the main pillars together with fast reactors for developing future sustainable nuclear energy systems.

REFERENCES

1. C. POINSSOT, S. GRANDJEAN, M. MASSON, B. BOULLIS, D. WARIN, , « Improving the actinides recycling in closed fuel cycles, a major step towards nuclear energy sustainability", GLOBAL 2013, Salt Lake City, Utah, USA, Sept. 30- Oct. 4, 2013.
2. C. POINSSOT, C. ROSTAING, S. GRANDJEAN, B. BOULLIS, "Recycling the actinides, the cornerstone of any sustainable nuclear fuel cycles", *Procedia Chemistry*, **7**, 349 – 357, (2012).

3. S. GRANDJEAN, M. MASSON, C. POINSSOT, L. PARET, D. WARIN, B. BOULLIS, « Towards the multi-recycling of plutonium and uranium: CEA's R&D program on advanced fuel treatment/recycling », GLOBAL 2013, Salt Lake City, Utah, USA, Sept. 30 - Oct. 4, 2013.
4. S. GRANDJEAN, C. POINSSOT, M. MASSON, D. WARIN, B. BOULLIS, « French R&D Program for Multi-Recycling of Plutonium », International Conference on Fast Reactors and Related Fuel Cycles: Safe Technologies and Sustainable Scenarios (FR13) Paris, France March 4-7, 2013.
5. C. POINSSOT, B.BOULLIS, Actinide recycling within the closed fuel cycles, *Nuclear Engineering International*, Jan.12, 17-21 (2012).
6. S. GRANDJEAN, N. REYNIER-TRONCHE, A. SALVATORES, N. HERLET, X. HERES, J-PH. DANCAUSSE, M. MASSON, C. POINSSOT, L. PARET, D. WARIN, B. BOULLIS, "Overview of the CEA's R&D dedicated to the treatment/recycling of Pu-based fuels (towards Pu multi-recycling)", Plutonium Futures the Science 2014 conference, Las Vegas, Nevada, USA, September 7-12, 2014.
7. C.POINSSOT, JM.ADNET, M.MASSON, C.ROSTAING, B.BOULLIS, " Main Results of the French R&D on Minor Actinides Partitioning, a Significant Improvement towards Nuclear Waste Reduction", Waste Management symposia 2015, March 15-19, 2015, Phoenix, Arizona, USA.