Internship Report

Master of Science in Nuclear Engineering - Fuel Cycle
Academic Year 2019/2020

Performance assessment of nuclear fuel cycles dedicated to plutonium and uranium multi-recycling

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Abstract

In light of the recent postponement of the deployment of commercial Sodium Fast Reactors in France to at least the second half of the 21^{st} century, multi-recycling of plutonium and uranium in PWRs has become an attractive option for better fissile material and natural resource management. The implications for cycle facilities and waste management, that these advanced material management options entail, need to be carefully studied and can be used to compare the different options.

The newly developed equilibrium scenario code SEPAR ("**S**imulateur d'**E**quilibres de **P**arcs **A**vancés de **R**éacteurs") is used to simulate equilibrium PWR fleets dedicated to multi-recycling of uranium and plutonium. The functionality of the code was tested on mono-recycling cycles, after which partial multi-recycling and full multi-recycling cycles, using the MIX fuel concept, were modelled. Post-processing routines were developed to extract the results from SEPAR and calculate values of interest such as isotopic compositions, fuel fabrication needs and natural uranium consumption. A method to calculate the production of plutonium and minor actinides, associated to the modelled cycle options, was devised and is described.

It was found that recycling fissile material allows to reduce natural uranium consumption, but generally increases the minor actinide production. Reactors fuelled by plutonium-based fuels are shown to increase the production of americium and curium, while using fuels based on reprocessed and re-enriched uranium increases the neptunium production of the fleet. If plutonium is recycled, the fleet composition and the production of plutonium and minor actinides are strongly dependent on cooling and fabrication times of the fuels. An increase of the minor actinide production with longer cooling and fabrication times is found for these fleets, with the effect being largest for full multi-recycling fleets. The isotopic compositions of fuels found for the full multi-recycling scenarios confirm that new cycle facilities, such as reprocessing and plutonium-based fuel fabrication facilities, are needed to achieve the advanced scenarios.

The code was found to perform well and obtained results are promising, although not validated yet. Future work will include validating the results and including the post-processing methods directly in the SEPAR code. Sensitivity studies on the advanced cycles as well as simulating other advanced cycles, including different systems, such as fast reactors or transmutation systems, can be performed. Some minor improvements to the code, such as extending the list of considered elements and isotopes, are also possible.

Acknowledgements

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Secondly, I would like to thank InnoEnergy for the financial support during my Master's studies, for the opportunity to study at two excellent universities, with professors and experts that are at the top of their field, and for the numerous field visits and external conferences that have allowed me to stimulate my interests greatly these last two years. Thank you also for providing us with a one-of-a-kind international environment that has allowed me to forge friendships with people from all over the world.

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Last, but not least, thank you, Emma, for making my time in Paris truly special, for your unconditional love and support, and for helping me to grow as a person.

Disclaimer

This report is the result of my internship at CEA Cadarache (DER/SPRC/LE2C). Due to the SARS-CoV-2 pandemic and the resulting confinement measures, the initially agreed start date was reported from 30th March 2020 to 22nd June 2020, while the end date was kept on 30th August 2020, thus shortening the official internship duration from 22 weeks down to 10 weeks.

Since the Master in Nuclear Engineering officially requires an internship with a minimum duration of 20 weeks, it was agreed, together with the internship supervisors and the academic responsibles at Chimie ParisTech, that work on the internship topic could be started before the official internship start date, and that the resulting internship modalities are deemed in accordance with the requirements in light of the extraordinary situation. This work began on 30th March 2020 (the initial start date) and continued up until the official start date with only short interruptions and consisted in a thorough literature review of the topic of nuclear fuel cycle modelling.

As a result of this situation, this report should be seen as a report in two parts: Chapter 1 (Introduction and Background) has emerged from the work done previous to the official internship start date, and should be considered as substantiation for the work done during this period. Work for all other chapters was performed during the official internship period. For the sake of readability, the report is however kept as a whole, as opposed to splitting it up in two distinct fragments, that do not make much sense if read separately.

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- **ASTRID:** Advanced Sodium Technological Reactor for Industrial Demonstration
- **CEA:** "Commissariat à l'énergie atomique et aux énergies alternatives", French Nuclear and Alternative Energies Commission
- **CIGEO:** "Centre industriel de stockage géologique", French deep geological repository project
- **DNB:** Departure from Nucleate Boiling
- **EPR:** European Pressurized Reactor
- **ERU:** Enriched Reprocessed Uranium
- **FIFO:** First in First Out
- **FLM:** Fuel Loading Model
- **FP:** Fission Product
- **FR:** Fast Reactor
- **HLW:** High Level Waste
- **LEU:** Low Enriched Uranium (<5%)
- **LHS:** Latin Hypercube Sampling
- **LIFO:** Last in First Out
- **LOCA:** Loss of Coolant Accident
- **LWR:** Light Water Reactor
- **MA:** Minor Actinide
- **MIX/MOXEUS:** Mixed Oxide Fuel with Enriched Uranium Support
- **MOx:** Mixed (Uranium & Plutonium) Oxide (fuel)
- **natU:** Natural Uranium
- **PUREX:** Plutonium Uranium Reduction Extraction
- **PWR:** Pressurized Water Reactor
- **repU:** Reprocessed Uranium
- **SF:** Spent Fuel
- **SFR:** Sodium Fast Reactor
- **SWU:** Separative Work Unit
- **TRU:** Transuranic Elements
- **UOx:** Uranium Oxide (fuel)
- **XML:** Extensible Markup Language
- **XSP** Cross Section Predictor

Chapter 1: Introduction & Background

1.1 The Current French Nuclear Fuel Cycle

For many decades, nuclear energy has been playing a large role in the French electricity mix. Facing an insecure energy transition due to the oil crises in the 1970's, France chose to tap into the potential of nuclear energy for electricity production to reduce its reliance on fossil fuels. It embarked on an ambitious journey to develop a large civil nuclear program and quickly proceeded to build a large number of Pressurized Water Reactors (PWR). As of today, 56 reactors of different electrical output, ranging from 900, 1300 over to 1450 MW and soon 1650 MW for the currently developed European Pressurized Reactor (EPR), are generating more than 70% of the country's electricity (as of 2019) [1].

As a part of the energy strategy, the choice in France was to recycle Pu from spent fuel, through the PUREX process at the La Hague reprocessing facility, and to re-use it once in mixed uranium-plutonium oxide (MOx) fuels. Depending on the future fuel cycle option chosen, MOx spent fuel can be considered as a valuable resource, since the plutonium still contains large amounts of potentially usable energy, or as a waste product. A schematic representation of the current French fuel cycle is shown in Fig. 1.1.

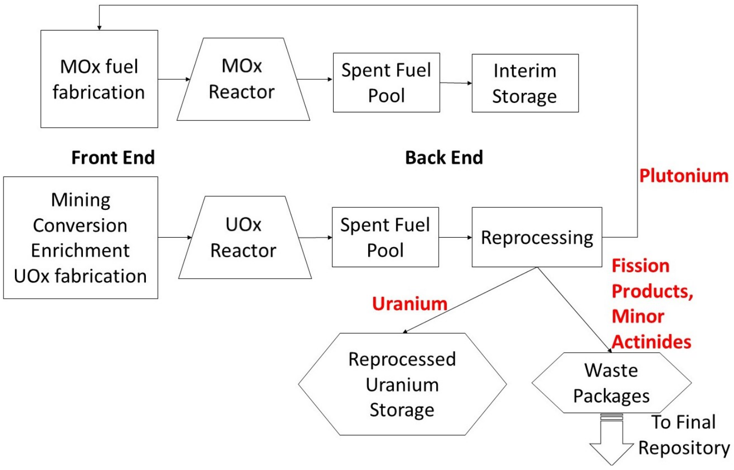


Figure 1.1: Schematic representation of the current French nuclear fuel cycle

The intention of the early developers of the French nuclear industry was to close the plutonium cycle by complete plutonium recycling. To achieve a fully closed cycle, physical reasons make it favourable for reactors to operate in the highly energetic, fast neutron energy spectrum (see section 1.2.1). All French reactors operating today are water-cooled, prohibiting the use of a fast neutron spectrum since water has moderating properties on the neutrons, slowing them down and causing a thermal, low-energy neutron spectrum in the reactor core, with neutron kinetic energies at thermal equilibrium with their surroundings.

Several types of Fast Reactors (FR) exist today, either operating, shut-down or conceptually. In France, engineering efforts have mainly been put into the Sodium Fast Reactor (SFR), yielding 3 SFRs operating in the past: Rhapsodie (1967-1983)[2], Phénix (1973-2010) [3] and Superphénix (1985-1998)[4]. The deployment of SFRs in France had rekindled in the 21st century with the Advanced Sodium Technological Reactor for Industrial Demonstration (ASTRID) program. The ASTRID program was intended to prove the technological readiness and commercial feasibility of the SFR and pave the way toward a commercial 1500 MWe SFR design and an associated closed fuel cycle. The project was dropped in August of 2019 and SFR deployment was delayed to the second half of the 21st century, a political decision reflecting an ambiguous stance toward nuclear energy, but also economic factors, as SFR are more expensive than PWRs, especially in light of very low uranium prices [5].

Given that French nuclear waste management laws mandate a sustainable nuclear material management [6] and FRs having disappeared from the short and medium term planning, alternative fuel cycle scenarios have to be developed. Recently, research has focussed on the possibility of multi-recycling fissile material in PWRs. The scope of this work is to examine such scenarios at equilibrium using the SEPAR code and determine the consequences on fuel and fissile material management, reprocessing and fuel fabrication capabilities, as well as waste management and natural uranium (natU) consumption issues.

1.2 Nuclear Material Management in the Fuel Cycle

Nuclear material management plays a key role for the nuclear energy sector in terms of waste management and sustainability, but also for fuel cycle economics and energy independence. Since spent low-enriched uranium (LEU) fuel still contains a large fraction of fissile and fissionable material, typically about 96% of uranium and 1% of plutonium [7], which can be re-used, strategies for material recovery and reduced natural resource use have been devised.

Using these resources can reduce the need for natural uranium but requires highly technological reprocessing solutions. From an economic point of view, the once-through option used in many countries and the French fuel cycle are considered to have similar cost within the margins of uncertainties [8]. In general, lower natural uranium prices make the once-through option more economically attractive but costs are less volatile in a reprocessing strategy which reduces the dependence on foreign-imported natural uranium resources. The cost differences for different cycle options can be important, the main challenge is therefore to find a good balance between technological feasibility, sustainability, economic competitiveness and energy-strategic considerations.

1.2.1 Multi-recycling of plutonium and constraints

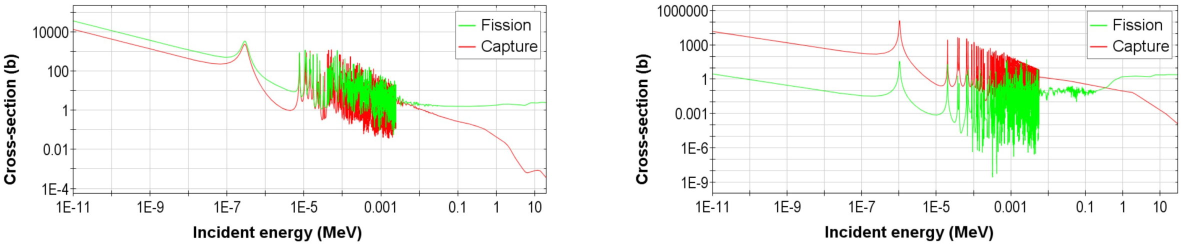
Technological feasibility of plutonium multi-recycling has been demonstrated for SFRs in the Phénix and Superphénix reactors [9]. In the case of conventional Light Water Reactors (LWR) there are physical barriers to multi-recycling. This is due, on one hand, to the shape of the fission and capture cross sections of the main fissile and fertile isotopes present in the fuel, and on the other hand to the shape of the neutron spectrum which is also affected by the isotopic compositions and geometric arrangement of the fuel.

In a thermal neutron spectrum, it is a general trend that plutonium grade decreases with increasing burnup of the fuel. Plutonium fissile grade is defined as the ratio of fissile isotopes over all isotopes, where the non-fissile decay product of ^{241}Pu, ^{241}Am, is included in the ratio. Although all isotopes of plutonium are included here, the main non-fissile isotopes of plutonium in spent fuel are ^{238}Pu, ^{240}Pu, ^{242}Pu.

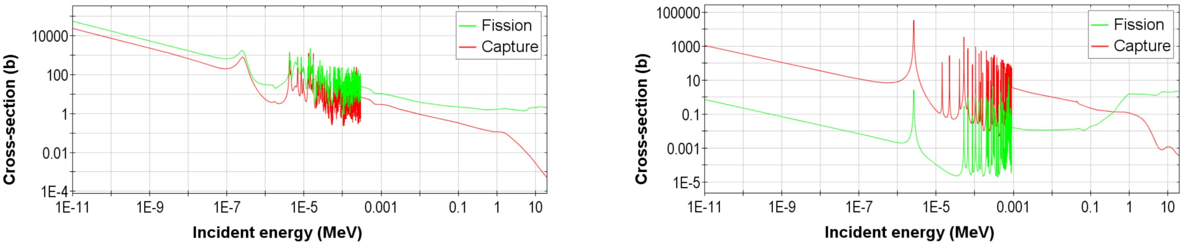
$$g = \frac{^{239}Pu + {}^{241}Pu}{\sum_{A=236}^{244} {}^{A}Pu + {}^{241}Am} \tag{1.1}$$

Fig. 1.2 shows that the capture/fission cross section ratio $\alpha = \sigma_c/\sigma_f$ is much higher for the fertile isotopes in the thermal region than in the fast region. This explains the accumulation of fertile isotopes in LWRs and thus the decrease of plutonium grade with increasing burnup in this type of reactor. Conversely, the fast spectrum is much better suited to use plutonium as a fuel since the α ratio decreases for all plutonium isotopes with increasing neutron energy: even isotopes become fissile with high enough neutron energy. The atoms thus fission much more efficiently in a fast spectrum and the accumulation of fertile elements is reduced.

Decreasing plutonium grade is not a problem per se: the decrease in macroscopic fission cross section with increasing burnup could theoretically be compensated by increasing the plutonium content in fresh MOx fuel. Increasing the plutonium content of the fuel bears a different risk relating to the safety of operation: the strongly decreasing capture cross sections of ^{240}Pu and ^{242}Pu (see Figs. 1.2b and 1.2d) towards a harder (more energetic) neutron spectrum could potentially create a positive void coefficient in the reactor core. In the case of creation of steam (void) in the reactor core, the neutrons are less efficiently moderated, hardening the spectrum and leading to a decreased absorption by ^{240}Pu and ^{242}Pu. This increases the number of neutrons and the generated power, generating more void and starting a positive feedback loop with potentially catastrophic consequences.



(a) Pu239 (b) Pu240



(c) Pu241 (d) Pu242

Figure 1.2: Main plutonium isotope fission and capture cross-sections [10]

Although the void fraction is very small for a PWR during normal operation (only some small bubbles due to nucleate boiling are possible), several accident scenarios with depressurization of the primary system, most notably loss of coolant (LOCA) scenarios, could lead to large void fractions in the core and therefore prohibit high contents of plutonium in the assemblies. It is generally accepted that a plutonium content of 12% should not be exceeded in PWR MOx assemblies to keep operation within safe limits [11]. SFR fuels, as used in Phénix for example, can have plutonium contents well above 20%. SFRs can also exhibit high moderator void coefficients, which can be handled either by including large negative reactivity reserves (control rods) or by having specific core designs that significantly lower the moderator void coefficient and even make them negative [12].

Outside of the reactor, constraints also appear when reprocessing plutonium based fuels. With current processes, fuels with high plutonium contents are more difficult to reprocess than uranium-based fuels, partly because plutonium oxide is less easily dissolvable than uranium oxide. Although MOx reprocessing has been demonstrated in existing facilities, by the reprocessing of small amounts of both SFR and LWR spent MOx in the past [13], research is still required to study the dissolution kinetics and mechanisms that are involved when plutonium oxide is dissolved [14]. Using data from [15], we can estimate that the La Hague facility could be able to reprocess about 50 t/y of spent MOx fuel to extract the plutonium and reintroduce it into fresh MOx fuel.

High ^{238}Pu content can also pose severe problems at the reprocessing step because of intense α radiation that can lead to high heat loads and radiolytic alteration of solvent and extraction molecules used in the liquid-liquid extraction process. At the same time, the heat load can be a problem when the material is handled for transport or fuel production. Today, the ^{238}Pu content limit for the MELOX MOx fuel fabrication facility is approximately 3.4% [16]. The problem with ^{238}Pu is heightened if spent ERU fuels are reprocessed since this fuel typically contains the highest amount of ^{238}Pu, but spent MOx fuel is also problematic. UOx and ERU/MOx spent fuel (SF) assemblies could be reprocessed together to dilute the ^{238}Pu in solution, but this has obvious limits.

In order for the current La Hague plant to reprocess ERU or MOx fuels, expensive operational and safety studies and possibly even more expensive additional equipment would be necessary. It is not sure that safe operation with limited negative impact on the process is technologically feasible, but even if it were, the regulatory process to license the facility for such isotopies would be a major hurdle. In the scope of this work, we therefore consider that the reprocessing of ERU SF requires new facilities, whereas when MOx is recycled, we assume an annual limit on MOx SF reprocessing.

1.2.2 Fuel Concepts for plutonium multi-recycling in PWRs

Plutonium management in a PWR core loaded with MOx is not straight-forward and needs to be carefully considered. The presence of large amounts of plutonium in the core changes the behaviour of the reactor compared to standard UOx fuel. Besides the smaller delayed neutron fraction of plutonium compared to uranium, which changes the dynamic behaviour of the reactor, the most notable effect is a different neutron energy spectrum. Indeed, larger absorption cross sections in the thermal domain for plutonium decrease the thermal flux in the case of MOx fuel, leading to a neutron spectrum that is shifted to higher energies and missing a peak in the thermal region (Fig. 1.3), an effect called hardening of the spectrum.

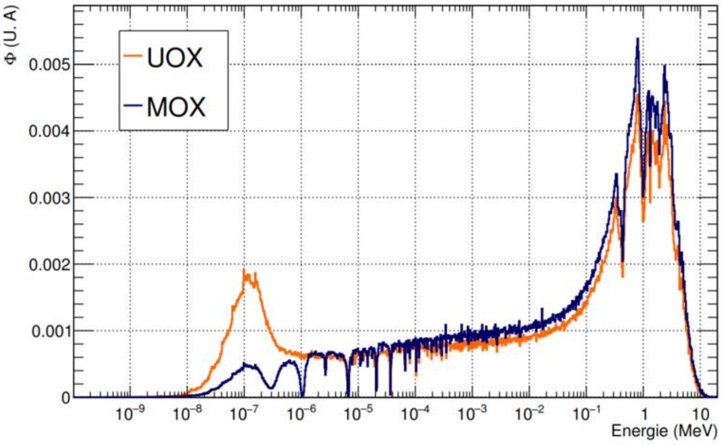


Figure 1.3: Normalised neutron energy spectra for UOx (3.25% enrichment) and MOx (Pu content of 5.3%) assemblies [17]

The hardened spectrum has an effect on the control rod worth. Since the rods are designed to have high absorption in the thermal region, a decreased thermal part in the case of MOx fuel decreases the effectiveness of the rods. For this reason, in a reactor loaded with MOx fuel, more control rods are necessary to guarantee the available negative reactivity insertion is sufficient at all times.

The plutonium distribution in the core has to be carefully studied. On the core level, MOx assemblies are generally located in the core periphery. This is done to limit the discharge burnup of the MOx fuels since, with high burnup, fission gases are released from cracking MOx pellets to the free space in the rod. The pressure in the rod is increased, an effect that should be limited to keep the fission gas leakage minimal. The presence of MOx fuel in the core periphery also reduces the temperatures of the fuel since the periphery of the core generates less power. This is important because MOx fuels present a slightly lower thermal conductivity than UOx fuels.

Although the EPR is designed to be able to operate with a full MOx core, the current fleet of PWRs in France is only licensed to operate with up to 30% MOx assemblies [18]. The necessary presence of two different types of fuel in the core makes appear boundary effects between UOx and MOx assemblies. Leaking thermal neutrons from UOx assemblies are absorbed at the periphery of a neighbouring MOx assembly and the increased thermal flux provokes an increased thermal power in the peripheral pins. Generally, large thermal peaking factors are unwanted phenomena since they can reduce the margin towards Departure from Nucleate Boiling (DNB). In order to avoid large power peaks at MOx/UOx interfaces, MOx assemblies have pin arrangements such that the plutonium content decreases towards the periphery, thus shielding the innermost, high-Pu, pins from the thermal flux of the UOx assembly.

Innovative fuel concepts for plutonium multi-recycling in PWRs exist. Two main concepts have been developed, namely the **MIX** (or MOXEUS) and **CORAIL** assemblies.

In the **MIX** concept, all pins in the assembly are identical with a fixed and homogeneously distributed plutonium content. Plutonium contents of 8%, 9.54% and 12% are often used as references [19]. Contrary to usual PWR MOx assemblies, an enriched uranium support is used, instead of depleted uranium, to compensate the plutonium degradation over several recycling cycles, and the uranium enrichment needs to be tuned to the specific plutonium composition of the fuel.

The **CORAIL** concept consists of an assembly with both UOx and MOx pins, where the reactivity is adjusted with the UOx pin enrichment and MOx pin Pu content simultaneously. The MOx pins are arranged in the periphery of the assembly to expose them to the lowest possible thermal fluxes and reduce power peaks due to UOx/MOx or water-filled guide tubes/MOx interfaces [20].

1.2.3 Uranium Management in PWRs

In today's fuel cycle, UOx assemblies are produced using an input of natural uranium (natU) and separative work units (SWUs) to yield LEU, with typical ^{235}U enrichment between 3% and 5%, and depleted uranium as a by-product with residual enrichment of around 0.2%. The depleted uranium stream is mainly considered without use, except some small amounts that are used in MOx production.

As spent UOx fuel is discharged from the reactor and reprocessed, three main streams of materials arise: a waste stream containing virtually all fission products and minor actinides (MAs), a reprocessed uranium (repU) stream and a plutonium stream. Plutonium recycling was already discussed in section 1.2.2, but the uranium stream also contains a considerable amount of fissile material that can be re-used to fuel a reactor.

Besides the total mass, the isotopic composition of uranium has changed considerably during irradiation in the reactor. Natural isotopes of uranium are ^{238}U with 99.275 wt%, ^{235}U with 0.711 wt% and traces of ^{234}U with 0.0054 wt% [21]. In spent fuel, all uranium isotopes with mass numbers ranging from 232 to 238 are present in varying proportions. Generally, for given initial enrichment, ^{235}U content decreases with burnup. The ^{236}U concentration increases with initial fuel enrichment and burnup since it is the product of neutron capture of ^{235}U. ^{236}U is considered to be a neutron poison due to its thermal absorption cross section. Although only very small amounts of ^{232}U are produced due to long formation routes, the isotope presents the largest contributor to the radiological impact of all uranium isotopes because some of its daughter nuclei are high-energy gamma and beta emitters [22]. A typical isotopic composition of high-burnup spent UOx fuel can be found in Table 1.1.

Table 1.1: Uranium isotopic composition (%) of natural uranium and uranium from irradiated UOx fuel for different burnups and initial enrichments (in brackets)

Isotope	**Natural Uranium**	**50 GWd/t** (4.19%)	**55 GWd/t** (4.58%)	**60 GWd/t** (4.99%)
^{232}U	/	4.95×10^{-8}	6.54×10^{-8}	8.40×10^{-8}
^{234}U	0.0054	0.016	0.017	0.018
^{235}U	0.711	0.786	0.808	0.833
^{236}U	/	0.609	0.680	0.754
^{238}U	99.275	98.590	98.495	98.395

The modified isotopic composition of reprocessed uranium poses two major hurdles for its re-use in a reactor: the high radiological impact caused by the presence of ^{232}U prohibits the easy handling of the material, making more radiological protection measures necessary, whereas the presence of ^{236}U, in quantities similar to ^{235}U, reduces the macroscopic fission cross section of the fuel, thus creating the need for over-enriching in the ^{235}U isotope.

Re-enrichment can happen either directly, in a gas centrifuge plant for example, but another possibility is to mix the reprocessed uranium with already enriched uranium to obtain the desired final enrichment. Alternatively, reprocessed uranium can also be used as a support for MOx fuel, decreasing the needed concentration of plutonium in the fuel because of the higher enrichment than the depleted uranium that is normally used.

Although several countries have experience with separating and re-enriching reprocessed uranium and using the fuel in their reactors, it is a rather uncommon practice today and most reprocessed uranium is just stored in oxide form. In France, four reactors (Cruas 1 to 4) have been operated with enriched reprocessed uranium (ERU) fuel reloads from 1994 to 2013 [22]. Political tensions between France and Russia, where the reprocessed uranium was enriched at the Seversk enrichment facility, halted the use of ERU fuel in French reactors [23]. Recently, French utility EDF has concluded a contract for reprocessed uranium enrichment with Urenco and fuel design and fabrication with Framatome, to start the supply of ERU fuel starting from 2023 until 2032 [24].

For uranium multi-recycling, reprocessing of ERU spent fuels is necessary. Although reprocessed uranium must be over-enriched in ^{235}U, there are no direct limitations to recycling the uranium stream multiple times in the reactor, since over-enriching doesn't pose an immediate safety problem in the reactor. The multi-recycling of uranium is limited by the current reprocessing process because of the high ^{238}Pu contents in the plutonium from spent ERU fuels (see section 1.2.1).

1.2.4 Impact on Waste Management

The fissile material management in the nuclear fuel cycle not only has an impact on in-reactor behaviour of the fuel and economics, but also on the form, composition and radiotoxicity of the final waste form. In France, high level nuclear waste (HLW) is reprocessed whereby the three streams, plutonium, reprocessed uranium and a solution containing fission products and minor actinides, are produced. The fission product/minor actinide stream is considered as final waste, the solution containing these elements is calcinated to a powder, mixed into a nuclear glass matrix and cast into stainless steel containers. These containers are the final waste packages that are stored in interim storage until the final repository project CIGEO is operational. Reprocessed uranium is not considered a final waste and is generally stored in oxide form, awaiting possible re-enrichment.

Radiotoxicity is a measure of the harmfulness, due to biological effects of ionizing radiation, of a substance on a human being, expressed as a hypothetical dose (in Sv) [25]. This harmfulness is dependent on the type of radiation, and therefore on the radionuclide contained in the substance, and on the type of contact.

The main contributor to radiotoxicity in high level waste are the transuranic (TRU) elements, especially on the long term, while for the initial hundreds of years, the fission product (FP) contribution is similar to the TRU contribution. Out of the TRU elements, plutonium dominates the radiotixicity over most of the time [25].

Total radiotoxicity of MOx fuel is considerably higher than that of UOx fuel [25] and consequently, it takes a longer time for MOx fuel to fall under the radiotoxicity value of natural uranium, which is often taken as a reference. This is due to the fact that spent MOx fuels contain more TRU elements than UOx fuels, which are mainly created by neutron capture on heavy elements such as uranium and plutonium, and higher plutonium content. With plutonium multi-recycling, this effect is exacerbated due to the increasing plutonium content in fresh and spent fuel.

Nevertheless, plutonium recycling has an important advantage over the once-through strategy: even though a higher minor actinide content will be present in the waste, only traces of plutonium will be present in the final waste form. This has the effect that the radiotoxicity of the waste is reduced drastically. However, even in a fully closed cycle strategy, the plutonium present in the reactor fleet and reprocessing facilities must eventually be considered as waste if nuclear fission energy is suddenly phased out. Plutonium multi-recycling has the advantage that a global equilibrium plutonium inventory can be reached using only a PWR fleet [19], meaning that the total amount of plutonium is kept constant.

Important limits for the waste packages are the heat load and self-irradiation from the decay of fission products and minor actinides as well as the chemical solubility limit of the different elements in the nuclear glass. Waste streams with high Am (^{241}Am especially) contents for example, release large amounts of decay heat and α irradiation, meaning that the heat evacuation capacity in interim storage and in the final repository must be adjusted to ensure that the packages are not altered faster than intended over time. Some products such as noble metals or Cm for example, are also not easily soluble in the glass matrix, and their increased production in MOx reactors might reach the limits on the waste content in the packages.

1.3 Scenario simulations

Scenario simulations are regarded as the main tool to investigate the advantages and drawbacks, technological constraints, cost and overall desirability of different fuel cycle and reactor fleet developments. The goal of scenario simulations is to take into account as many parameters as possible that influence the performance of the system, while allowing the simulation of a large number of scenarios to find the optimal one, given a set of criteria. From scenario studies, informed policy decisions can be derived to assure the best outcome for the population.

1.3.1 Main Parameters of a Nuclear Energy System

The modelling of a complex system such as a nuclear energy system requires physical modelling of all major processes occurring in the system. A nuclear energy scenario is generally defined by the components of the system, i.e. enrichment, fuel fabrication, reprocessing facilities and reactors and the scenario aims to forecast the evolution or equilibrium state of the system depending on input parameters. The evolution is mostly captured in the material fluxes between the different components of the system.

At the front end of the fuel cycle, **natural uranium mining, conversion, enrichment and fuel fabrication** capacities need to be calculated in order to allow for a smoothly functioning fuel supply for the system. Fuel fabrication refers not only to LEU fuel from natural uranium but fuel fabrication facilities must also be able to produce the required ERU and plutonium-based fuels for LWR (and possibly FR), depending on the chosen fuel cycle strategy.

The heart of the nuclear energy system is the reactor fleet. For the simulation, a **target electricity generation capacity** needs to be defined, as well as the **reactor types and numbers composing the fleet and according fuel types and core arrangements** that are used. These parameters directly influence the aforementioned front-end, but also back-end facilities. They define the required fuel enrichment, plutonium content according to the grade and total quantity of assemblies needed. The relation between the reactor physics and isotopic evolution of the fuels is a core issue in long-term scenario simulations and is described below (Section 1.3.2).

The back-end of the fuel cycle is mainly described by the **reprocessing facility throughput and interim storage requirements for spent fuel**. They are influenced by the cooling time of spent fuel between discharge from the reactor and the reprocessing step and the order in which the spent assemblies are sent into reprocessing. Two main strategies are possible, the first being the First In First Out (FIFO) strategy, where the oldest assembly in storage is sent to reprocessing. The second strategy is the Last in First Out (LIFO) strategy, where the most recent spent assembly is sent for reprocessing first. Both approaches have advantages and disadvantages. While in the FIFO strategy a considerable portion of fissile ^{241}Pu has decayed over the years into non-fissile ^{241}Am, the LIFO strategy presents problems with radiological hazards and heat evacuation, since the shorter-lived fission products have not decayed yet and are producing considerable amounts of heat. Currently, the FIFO strategy is carried out in the French mono-recycling strategy.

Finally, the waste composition and volumes, required vitrification facility capacity and final repository sizing can be determined.

Since the use of industrial scenarios is ultimately to make strategic decisions, the simulated scenarios should be defined in a realistic frame. Specifically, it has to be taken into account that some technologies might not be mature for deployment on the industrial scale now. Another real world restriction on the scenarios is that construction times for new facilities should be respected and that all facilities are designed for a given capacity at which they will operate for several decades. Quick changes in such time- and capital-intensive infrastructure projects are difficult to manage.

1.3.2 Calculating the isotopic evolution in the system

The heart of the scenario modelling problem resides in predicting the isotopic composition of the fuel assemblies at different locations in the cycle. The output of the reactor is defined by the reactor type, core arrangement and by the fresh fuel that was initially introduced in the reactor.

General Principle

To be able to predict the evolution of the nuclear fuel, the first thing that needs to be known is the fresh fuel composition. Although the fresh fuel obviously needs to be built from available material, creating the fresh fuel is not a trivial task since the fuel should contain enough fissile material to reach the desired burnup. The first modelling step in the scenario simulation therefore needs to be a prediction on the fissile content (uranium enrichment or plutonium content), needed for a given burnup at the isotopic composition of the available material. The fresh fuel composition can be calculated by iteratively varying the fissile content in the fuel until the desired burnup is reached.

The evolution during irradiation could be calculated using validated neutronic codes. From the fresh fuel composition, the isotopic composition, if necessary with resolution in time, can be calculated accurately. An example of a deterministic code that solves the Boltzmann equation for the neutron transport is APOLLO2 [26], whereas a stochastic or Monte Carlo code, like TRIPOLI [27], coupled to a depletion module, could be used with the same result. The issue with either of the two setups is that these codes generally require powerful computers (or long computing times), making it impractical to calculate through many scenarios.

Neural Network Approach

Instead of using resource-intensive neutron transport codes, neural networks can be used to predict the fresh fuel composition and the fuel evolution in the reactor. The idea here is that, instead of performing neutron transport calculations each time, a database with a large number of neutronic simulations can be created previously and the results can be used to train neural networks, which then in turn can be used to do the predictions. The advantage of this approach is that the neutronic calculations and the neural network training, which are both time-intensive, only need to be performed once, after which the neural network can perform the predictions very quickly.

The neural network approach does not change the general principle but only the calculation method. First, the fresh fuel composition is predicted using the isotopic composition of the input material as an input into the neural network. Then, from the fresh fuel isotopic composition, another neural network is able to predict the isotopic composition of the fuel after irradiation.

Dynamic Scenario Codes

Dynamic scenario modelling codes, like COSI [28] or CLASS [29], are able to predict the state of the nuclear energy system for any given timestep. The methodology that is explained here is the methodology used in the CLASS code but even though other codes might use slightly different approaches, the general idea is the same.

The method relies on two main modules that are trained on the results from a coupled transport-depletion code. The first module is a Mean Cross Section Predictor (XSP), needed to do the depletion calculation, while the second module is a fuel loading model (FLM), needed to determine the required plutonium content for a given plutonium composition, uranium enrichment and target burnup.

Since the XSP needs the isotopic composition of the raw materials for the fuel as an input, the FLM is used to build the fuel composition from the available resources at the time of fuel production. The available stock of nuclear material that can be used to produce the fuel is calculated from previously discharged fuel elements and cooling and reprocessing times are modelled to respect the isotopic changes during these periods. The FLM needs to calculate the necessary amounts of plutonium at the current grade, with known uranium support enrichment to reach a desired burnup.

Once both modules are implemented, the calculation scheme is rather straight-forward: the FLM takes today's reprocessed fuel, for which characteristics are exactly known, builds a fuel according to the required burnup and the used uranium enrichment and sends this information to the depletion module that is made of the XSP and the Bateman solver. The depleted material is sent back to the stock after the cooling and reprocessing time and by propagating this method into the future, the behaviour of the fuel in the reactor for changing composition over time can be modelled.

Equilibrium Scenarios Codes

If the goal of the simulation is to explore final equilibrium states of the nuclear energy system, the transitory behaviour of the system must not necessary be modelled. A dynamic scenario simulation can obviously be used to find a possible equilibrium of the system, but a dedicated code to explore equilibria can be more effective in terms of computing time. Since the equilibrium solution will not have any temporal resolution, the equilibrium code can directly predict the output isotopic composition from the initial composition and the burnup.

Training Data for the Neural Networks

For the model to work, the neural networks need to be trained with results from a validated transport-depletion code. For simplicity and speed, modelling is often done on a fuel assembly with infinite geometry (mirroring boundary conditions) and the corresponding reactivity of a whole core of these assemblies is derived from the calculation, which is a standard procedure for modelling LWR cores.

For this, an average k_∞ method is used. This simplified approach considers a whole core infinite multiplication factor that is averaged over the time spent by an assembly in the reactor [30]. The expression for k_∞ can be found in equation 1.2.

$$< k_\infty(t) >= \frac{1}{N} \sum_{i=0}^{N-1} k_\infty \left(t + \frac{iT}{N} \right) \tag{1.2}$$

The time spent in the reactor is divided into N refuelling cycles and the total time spent in the reactor is denoted T. The assembly infinite multiplication factor in the sum is denoted k_∞. The criticality condition for the core in the infinite reactor approximation would be $< k_\infty(t) >= 1$. Taking into account the inevitable neutron leakage and absorption in structural materials that are not included in the simulation, the real criticality condition is then $< k_\infty(t) >= 1+P = k_{threshold}$, where the P factor includes leakage and parasitic absorption [31]. For simplicity, the boric acid concentration is kept constant during irradiation, meaning that the condition must only hold at the end of the cycles: $< k_\infty(\frac{iT}{N}) >= k_{threshold}$. The leakage and absorption factor P is defined by user input and is generally between 0.03 and 0.04 [30]. To determine the maximum achievable burnup, the depletion calculation is run until time T is reached. From the final irradiation time T the burnup of the fuel can easily be determined.

To build a simulation database for a fuel, a wide range of input parameters, spanning all the parameter space that can be encountered in the nuclear power industry, needs to be used as input for the calculations. For the simulation, isotopic composition of uranium and plutonium (for MOx fuel), final burnup and specific power are needed as inputs. The enveloping intervals that delimit the ranges for these parameters are determined by using "usual" isotopic compositions for spent fuels and extending the isotopic fractions by margins so as to be sure that, even with more advanced scenarios, the isotopic compositions will be contained within the determined ranges [31].

Once the enveloping ranges for all the isotope contents have been defined, a large number of simulations can be run by sampling isotopic compositions of plutonium, the enrichment of the uranium support and the total plutonium content in the fuel. In practice, the content for the plutonium isotopes 238, 239, 241 and 242 as well as for ^{241}Am are sampled in their range and the content of ^{240}Pu acts as a buffer, meaning that it is adjusted such that the sum of isotope contents is 1.

As a sampling method, Latin Hypercube Sampling (LHS) is often used for this type of application. The method consists in dividing each parameter range into M intervals with M+1 interval edges and by randomly combining one interval edge from each parameter range into an input vector. The result are M+1 randomly selected input vectors spanning the whole parameter space in a more or less uniform manner.

Once a large number of input vectors have been chosen, the neutronic calculations can be performed and the maximum achievable burnup is saved for the given input vectors. In case of a dynamic code, the evolution of the mean cross section of the isotopes as a function of depletion time are saved and constitute a fuel library. In the case of equilibrium scenarios, the fuel evolution is predicted directly as an output isotopic composition and there is no temporal resolution to the results. The neural networks for both the XSP (or irradiation model) and the FLM are trained on the fuel library, after which the neural network code is ready to make its own predictions.

Chapter 2: Calculation Methods

2.1 The SEPAR code

The SEPAR (**S**imulateur d'**E**quilibres de **P**arcs **A**vancés de **R**éacteurs) code was developed at CEA by the DER/SPRC/LE2C and is able to model a nuclear energy system at equilibrium with the possibility to model advanced fuel cycles. It relies on a neural network approach to estimate the equilibrium material composition and fluxes for each isotope to and from reactors, enrichment, reprocessing and final storage facilities.

The code takes into account the mass balance equation for each isotope i of element E including the decay of E_i in equilibrium with simultaneous accumulation due to the decay of possible precursor or parent isotopes denoted P_{E_i}. The decay equilibrium for elements P_{E_i} and E_i are considered over cooling periods t_{sR} (where s denotes the fuel management type and R denotes the reprocessing facility) and fuel fabrication period t_r. Partial fluxes are treated using a fraction of flux denoted ϵ_{sR} for the partial transfer of the flux from reactor s to reprocessing facility R and ϵ_R from the reprocessing facility R to the fuel management system with index k and batch fraction x_k. The material balance is done at the supply point of the fuel management system k (Fig. 2.1).

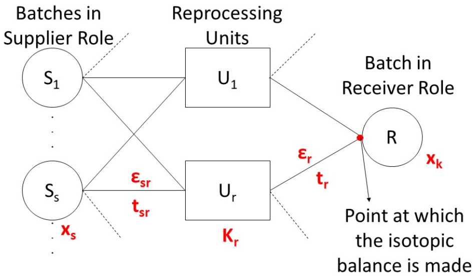


Figure 2.1: Schematic representation of fluxes between different components of the nuclear cycle in SEPAR

The decay and accumulation behaviour derives from a simple decay equilibrium of parent and daughter isotopes. The system for two isotopes is described by the differential equations 2.1.

$$\begin{cases} \frac{dm_p}{dt} &= -\lambda_p m_p \\ \frac{dm_i}{dt} &= -\lambda_i m_i + B_{pi}\lambda_p m_p \end{cases} \tag{2.1}$$

Here, B_{pi} is the branching factor for the decay of p to i. The solution to this equilibrium problem is well known and the solution for the evolution of the daughter nucleus is:

$$m_i(t) = B_{pi}\frac{\lambda_p}{\lambda_i - \lambda_p} m_p^0 (e^{-\lambda_p t} - e^{-\lambda_i t}) + m_i^0 e^{-\lambda_i t} \tag{2.2}$$

Here, m_i^0 and m_p^0 denote the initial mass of daughter and parent nuclei respectively. In the model, this initial mass is the mass at the output of the fuel management system s. In the final master equation, this initial mass is expressed with the batch fraction x of the material flux φ coming from the different fuel management types and reprocessing facilities and the total isotopic fraction f_i: $m_i^0 = \sum_R \epsilon_R \sum_S \epsilon_{sR} \cdot x_s \cdot \varphi_s \cdot f_i$. An analogous expression exists for the parent nucleus.

With the described approach, the equations only take into account decay chains of two elements. For longer decay chains, like the $^{241}Pu \rightarrow {}^{241}Am \rightarrow {}^{237}Np$ chain for example, the results are therefore not complete in the current version of the SEPAR code, although this approximation should not influence the overall results dramatically. During the course of this work, a way to incorporate decay chains of arbitrary lengths into the SEPAR code was elaborated by solving the Bateman equations analytically with a method described in [32]. Since modifying the code is beyond the scope of this work, a short description of the method can be found in Appendix A.

Since the evolution of spent fuel from the reactor to another use in the reactor is not a simple decay but other changes, like reprocessing, are executed, the expression becomes slightly more complicated. We can divide the process into two steps: from the reactor to the reprocessing step and from the reprocessing step back to a reactor. As we have the reprocessing and fuel fabrication times t_{sR} and t_R, we can write the evolution for both periods:

$$\begin{aligned} reactor \rightarrow reprocessing : m_i(t_{sR}) &= B_{pi}\frac{\lambda_p}{\lambda_i - \lambda_p} m_p^0 (e^{-\lambda_p t_{sR}} - e^{-\lambda_i t_{sR}}) + m_i^0 e^{-\lambda_i t_{sR}} \\ reprocessing \rightarrow reactor : m_i(t_R) &= B_{pi}\frac{\lambda_p}{\lambda_i - \lambda_p} m_p^{0'} (e^{-\lambda_p t_R} - e^{-\lambda_i t_R}) + m_i^{0'} e^{-\lambda_i t_R} \end{aligned} \tag{2.3}$$

Note here that the initial masses for the isotopes are not the same in both equations. This is due to the fact that during reprocessing, both parent and daughter nuclei have decayed and/or accumulated, but also have been separated completely or partially during reprocessing. For the mass of isotope i having been produced due to the decay of p during the reprocessing time, we have to adjust the quantity by the part of this mass that is going to decay during fuel fabrication and by the amount that will be lost in reprocessing, so we introduce a decay factor $exp(-\lambda_i t_R)$ on the produced quantity as well as a reprocessing transfer function $K_R(E_i)$. Similarly, for the period of fuel production, we need to take into account that in the reprocessing period, the parent isotope has been depleted due to decay and it might also have been removed during reprocessing, so we introduce a decay factor $exp(-\lambda_p t_{sR})$ and a reprocessing transfer function $K_R(P_{E_i})$ to account for this.

For the reprocessing, the reprocessing function of an element i is a linear function of the isotope content E_i: $K_R(E_i) = \rho \cdot E_i$, where ρ is a constant. As the PUREX process is very efficient with very small losses, ρ is generally close to 1 for wanted elements (plutonium and/or uranium) and close to 0 for unwanted elements (fission products, minor actinides). Taking all these considerations into account, SEPAR is able to find the equilibrium conditions for the nuclear energy system.

2.2 Batch Fractions and Equilibrium Input Isotopic Compositions

The equilibrium conditions of the fleet are calculated using the SEPAR code and complemented with calculations from python routines developed by the author, which use results from the code. The inputs into SEPAR are the reactor types and description of the fuel management (final burnup, reloading fraction, core mass, equivalent full power days, load factor, thermal efficiency) in the respective cores as well as the links between fuel batches and reprocessing facilities and the times that the fluxes need to pass from one facility to the other. The fuel batches, the links and respective times are provided to SEPAR in files in the XML format. The fuel library data, necessary to train the neural networks, are provided in the DAT format and generated from APOLLO2 simulation results. The outputs of SEPAR are the fractions of each fuel batch (batch fraction) and the input isotopic composition of the respective batches. SEPAR also provides the neural network files containing the architecture and parameters of the neural networks used to calculate the equilibrium conditions.

2.3 Fuel Needs and Natural Uranium Consumption

To calculate the fuel needs for each fuel management, a target installed capacity of the fleet is chosen, and from the batch fractions calculated by SEPAR and the fuel management characteristics, the yearly fuel fluxes for each management can be calculated. For all results, an installed capacity of 61.2 GW is assumed, which is representative of the current French fleet. The natural uranium consumption is calculated using the UOx fuel needs and enrichment with an assumed tails assay of 0.2%.

2.4 Output Isotopic Compositions

For the minor actinide and plutonium production calculation, the output isotopic compositions of the fuels in the cycle need to be known. SEPAR has the capability to predict the isotopic composition of irradiated fuel from the input isotopic composition using neural networks, but, at the current version, it does not give these values as an output. Nevertheless, SEPAR can be "tricked" into generating neural networks that predict the output content of each isotope, even if it does not necessarily need the information to perform the equilibrium calculation. From these neural network files, using a self-developed python method, all the necessary output isotopic compositions of the fuels can be predicted using the known input compositions.

2.5 Enrichment and Enrichment Factors

Enrichment factors refer to the ratios of enrichment for different isotopes of an element compared to the reference isotope (in our case ^{235}U). Since centrifugation relies on the mass difference of isotopes to enrich material, isotopes of different masses are affected to different degrees by this process. This means that in isotopic mixtures containing more than two isotopes, it is necessary to assign enrichment factors to each isotope to take into account how efficiently this isotope is enriched in the process. In the case of reprocessed uranium, this is extremely important, since not only ^{235}U is enriched, but significant quantities of ^{236}U and ^{234}U are also enriched at the same time. The isotopic composition of the uranium plays a major role for the neutronic properties of the fuel, it is therefore important to be able to calculate the isotopic composition of reprocessed uranium after enrichment accurately.

Here, we use the fact that, for small mass differences between isotopes in the mixture, the enrichment factors obey a linear law [33]:

$$EF_i = \frac{M_{^{238}U} - M_i}{M_{^{238}U} - M_{^{235}U}} \tag{2.4}$$

where M denote the atomic masses of the respective isotopes and i denotes the isotope for which the enrichment factor is calculated. Using Eq. 2.4 on the isotopes ^{234}U and ^{236}U, we find enrichment factors of 1.33 and 0.67 respectively.

The required enrichment for ERU fuel is calculated using a neural network trained on data from APOLLO2 equivalence calculations. In APOLLO2, the required enrichment to reach the desired burnup is calculated iteratively and the fractions of other uranium isotopes are adjusted using an enrichment factor.

2.6 Minor Actinide and Plutonium Production

Calculating the amounts of MAs or Pu that are produced in an equilibrium fleet is not straightforward. This is due to the fact that, even if a fleet is in prefect equilibrium, the MA and Pu masses in each part of the cycle are never really at equilibrium. Their decay always induces a notable change on different time scales, according to their half-lives. **The challenge at hand is therefore to assign a representative value to an inherently non-equilibrium quantity for a system that is supposed at equilibrium**.

To achieve this, a pseudo-dynamic approach to the problem has been devised by the author: the production of isotopes is calculated in every part of the cycle at a given point in time. For the calculation, the isotopic evolution due to decay of materials in different places of the cycle is explicitly taken into account, using the appropriate form of equation 2.2. **The origin of time is defined as the moment at which the fleet first discharges SF into a SF pool whose inventory is never stabilized, and the fleet is supposed to operate at equilibrium from t=0**. The definition of t=0 is dependent on the fleet: for a once-through UOx cycle for example, the UOx SF inventory is increasing steadily, while in an UOx-MOx fleet, the UOx SF inventory is stabilized. With this approach, the notion of an equilibrium fleet is kept, while at the same time, we are able to introduce some notion of time to take account the decay of certain elements.

Since MAs and Pu have a constant in-reactor production term at equilibrium, it is the time-dependent decay/accumulation outside of the reactor and the initial quantities of each of the isotopes that determines the production term as a function of time. **For every isotope, an equilibrium between in-reactor production and decay/accumulation will be reached according to the half-life of the isotope (and the parent isotope's, if there is one) and the equilibrium will be reached in order of increasing half-lives.** In reality, an equilibrium is always reached, but might be so far in the future that the times do not make sense in the realm of scenario simulations. SEPAR only considers isotopes to be radioactive if they have half-lives smaller than 1000 years, which is a relevant approximation for the times considered in the scenario simulations.

The temporal evolution of Cm is characterised by fast decay (without source term from parents) since ^{242}Cm, ^{243}Cm and ^{244}Cm have half-lives of 0.44, 18 and 30 years respectively and Cm will be the first element to reach an equilibrium production. Am production shows an initial fast increase due to the short half-life of the ^{241}Pu parent isotope (14y), followed by a decrease according to the ^{241}Am half-life (432 years). Np production is driven by the decay of ^{241}Am to ^{237}Np, and since ^{237}Np has a half-life on the order of a million years, it is considered stable in SEPAR.

The production of Pu is slightly more complicated due to the multiple accumulations and decays possible: ^{242}Cm, ^{243}Cm and ^{244}Cm produce ^{238}Pu, ^{239}Pu and ^{240}Pu respectively and ^{238}Pu and ^{241}Pu are considered as radioactive by SEPAR. The half-lives make it such that, initially the production declines rapidly due to the ^{241}Pu decay (the accumulation due to Cm is negligible since only small quantities of Cm are present). Once ^{241}Pu production has reached equilibrium, the production of ^{238}Pu increases (since the decay term is decreasing), meaning that the Pu production reaches a minimum and then reaches equilibrium at a higher value.

Fig. 2.2 shows this temporal evolution, and we can see that **total MA production has already nearly reached its equilibrium value at t=1000. Similarly, the Pu production has already passed through its maximum and minimum value at t=1000. Therefore, the total MA and Pu productions will be reported as ranges.** The figure is drawn until a time of t=10000, but it should be noted that only half-lives smaller than 1000 years were considered, so the real evolution would be different. The solid black line at t=1000 shows the limit, past which no conclusions should be drawn.

Since Np and Am productions have not reached their equilibrium at t=1000, their production would depend on the moment of time chosen as a reference. **Therefore, it is chosen to calculate the in-reactor production of Np, Am, Cm and give their relative fractions.** This allows to compare different fleets to each other, but the values are distorted to low Am and Np, but high Cm values, since the former isotopes would build up outside of the reactor over time. This limits the ability to compare the results obtained for Np, Am and Cm to results from the literature, since those values generally take into account the out-of-reactor evolution as well.

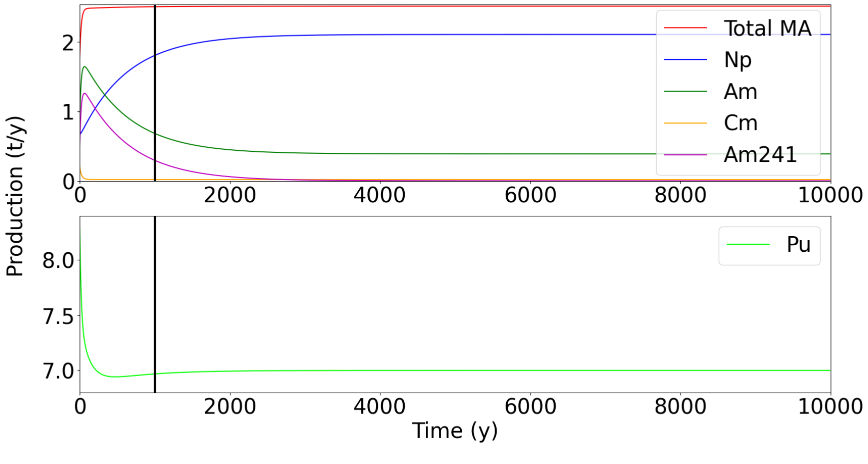


Figure 2.2: Evolution in time of minor actinide and plutonium production on the example of a UOx-MOx fleet. Note that the y-axis for the Pu plot does not start at 0 so that the curve minimum is visible.

Chapter 3: Results

All fuels used in the simulations are assumed to reach a final burnup of 55 GWd/t, all cores are supposed to have a mass of 129 t, $4592 MW_{th}$ of thermal and $1653 MW_e$ of electrical power. The initial UOx enrichment is 4.58%. The load factor of the reactors is assumed to be 83% [19] and the refuelling fraction is 1/3. Reactor data, as well as geometric data used for APOLLO2 simulations are representative of a modern GENIII PWR design and are taken from [34–40]. With the required 61.2 GW installed capacity, the fleet comprises a total of 37 reactors.

The fleets are all modelled for several combinations of cooling times (=time a fuel assembly spends in the spent fuel pool before reprocessing) and fabrication times (= time between reprocessing and reintroduction of material into reactor). The chosen cooling times are 1, 2, 5, 10, 20, 50, 100, 200 years whereas the fabrication times are chosen among 1, 2, 5, 10, 20 years, except explicitly noted otherwise.

3.1 Non-Advanced Scenarios

Even though the main goal of the present work is to compare different multi-recycling scenarios, once-through and mono-recycling scenarios are also investigated to test the functions of SEPAR and provide a basis for comparison for the multi-recycling scenarios. The analysis of mono-recycling scenarios also allows to develop an idea on what the most impactful factors are.

The equilibrium conditions, natural uranium consumption as well as the plutonium and minor actinide production, for a chosen reference case with 10 years cooling times and 2 years fabrication time [41], are reported together for all non-advanced cycles in Tables 3.7, 3.8 and 3.9 respectively. A reference case is chosen because large variations in many of the parameters exist with changing cooling and fabrication times, such that comparison between cycles would be difficult otherwise.

3.1.1 Once-through UOx fleet

For the once-through UOx fleet, a **fuel fabrication capacity of 936 t/y** with a **natural uranium consumption of 8022 t/y** is calculated. The plutonium isotopic composition of the spent fuel can be found in Table 3.1. The uranium isotopic composition was already reported in Table 1.1.

Table 3.1: Plutonium isotopic fractions and content (%) of spent (55 GWd/t) UOx fuel

Pu238	**Pu239**	**Pu240**	**Pu241**	**Pu242**	**Am241**	**Pu+Am241**	**Total**
2.976	48.511	25.529	14.572	7.953	0.458	1.244	

3.1.2 UOx-MOx Mono-Recycling Fleet

A schematic representation of the UOx-MOx cycle option can be found in Fig. 3.1.

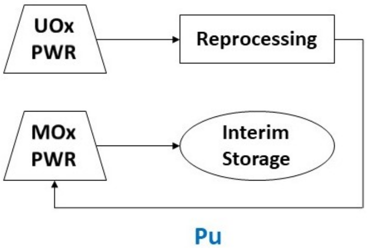
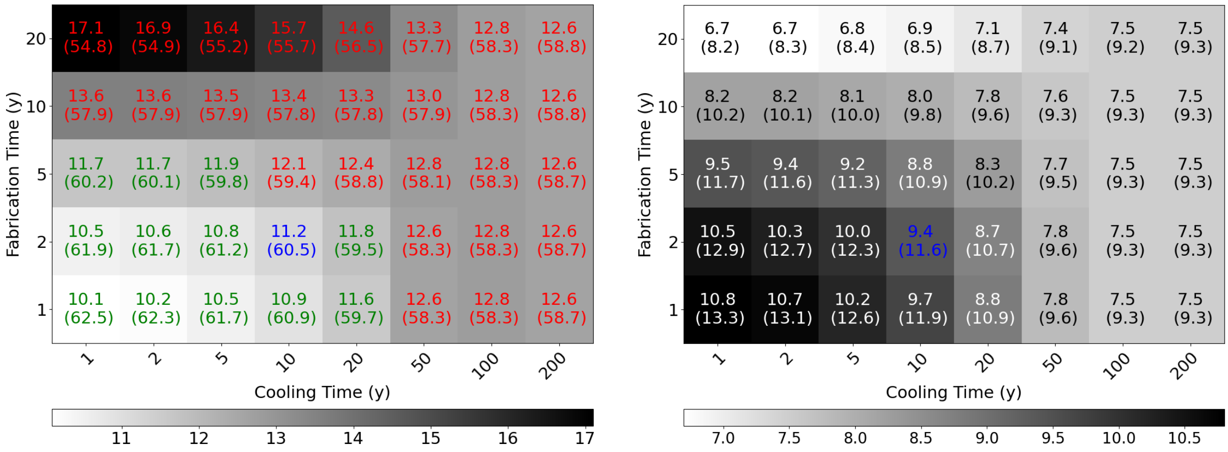


Figure 3.1: Schematic representation of the UOx-MOx cycle

Firstly, the results in Fig. 3.2a show that **the cooling and fabrication times have an important impact on the plutonium grade.** This is due to the decay of ^{241}Pu to ^{241}Am: the depletion of ^{241}Pu and accumulation of ^{241}Am decreases the fissile grade. In general, the shorter the sum of both times is, the better the plutonium quality will be. Nevertheless, since all Am is removed during reprocessing, the fabrication time has the biggest impact on the plutonium grade since after the MOx fuel has been produced, the Am cannot be removed and constitutes a neutron poison in the fuel. In contrast, even if the cooling time is long, and a lot of ^{241}Pu has decayed, a long fabrication time does not decrease the grade dramatically since all Am is removed and there is not enough time for large quantities to build up again.

A decreased grade automatically means that the plutonium content in fresh MOx fuel must become higher to guarantee enough reactivity in the fuel to reach the desired burnup. This effect can also be seen in Fig. 3.2a: with increasing plutonium content, the grade decreases. Since the safety limit for PWRs is at 12% plutonium content (see section 1.2.1), we can conclude that some combinations of cooling/fabrication times are impossible in reality for the chosen (high) burnup. The cases are marked in red in Fig. 3.2a.



(a) Fresh MOx Pu content (%) and grade (%) (in brackets)

(b) MOx batch fraction (%) and reactor numbers (in brackets)

Figure 3.2: Plutonium content (grade) and equilibrium MOx batch fraction (reactor number) for the UOx-MOx cycle. The reactor number assumes 30% MOx assemblies in the core. Red values in (a) indicate that the 12% safety limit is exceeded. Blue values are the values for the reference case.

Currently, the plutonium content in fresh MOx fuel is significantly lower than the minimum 10% calculated here [18]. This is due to the fact that today's MOx fuel is produced from plutonium from historic fuels with much lower burnups than are considered here (up to some 45 GWd/t at maximum but lower on average, compared to 55 GWd/t used here), giving the plutonium a much better grade. This also means that, with historic fuel stocks, other cooling/fabrication time combinations might be possible.

The grade, and thus cooling and fabrication times, have an important effect on the fleet composition (Fig. 3.2b). Fig. 3.2b shows the batch fractions, i.e. the fraction of MOx assemblies in the whole fleet, and the number of reactors with MOx assemblies in parentheses. For the reactor number, a 30% MOx/ 70% UOx ratio is assumed, an approximation since the neutronic calculation was performed on the MOx assembly only, so no UOx/MOx mixing effects in the spectrum are taken into account.

The MOx batch fractions are higher when the plutonium grade is higher, i.e. when cooling and fabrication times are shorter. Firstly, this is due to the fact that higher grade means less plutonium content, so more reactors can be fuelled with the same mass of plutonium, and in addition, less mass of plutonium is available due to decay if the times are long. The ranges for the main fleet parameters, obtained with the used cooling/fabrication times, are reported in Table 3.2.

3.1.3 UOx-ERU Mono-Recycling Fleet

A schematic representation of the UOx-ERU cycle option can be found in Fig. 3.3.

ERU fuel is not as sensitive to changes in cooling/fabrication times as MOx fuel, since, contrary to plutonium, all uranium isotopes are considered stable by SEPAR. The only notable

Table 3.2: Ranges for the main fleet parameters obtained for the UOx-MOx fleet for the different cooling/fabrication times.

Batch fraction (%) (Fuel fabrication (t/y)) UOx	MOx	**MOx Pu content** (%)	**natU consumption** (t/y)
89.2 - 93.3 (835 - 873)	6.7 - 10.8 (62 - 101)	10.1 - 17.1	7155 - 7487

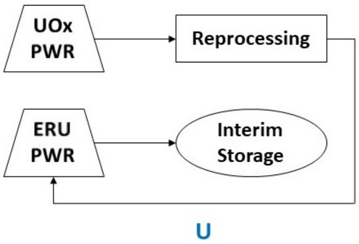


Figure 3.3: Schematic representation of the UOx-ERU cycle

influence of time on the isotopic composition is the slow accumulation of ^{234}U due to the decay of ^{238}Pu with a half-life of 87.7 y. The effect of ^{238}Pu decay is captured by the simulations but remains very small: equilibrium concentrations of ^{234}U in spent and reprocessed uranium before enrichment increase with higher cooling times.

The ranges obtained for the main fleet parameters with the combinations of cooling/fabrication times that were used, are reported in **Table 3.3, which shows that the variation of the required ^{235}U enrichment with times is very small. This enrichment is however significantly higher than the enrichment of fresh UOx fuel (4.58%), which is due to the poisoning effect of other uranium isotopes, most notably ^{236}U**.

Table 3.3: Ranges for the main fleet parameters obtained for the UOx-ERU fleet for the different cooling/fabrication times.

Batch fraction (%) (Fuel fabrication (t/y)) UOx	ERU	**ERU enrichment** (t/y)	**natU consumption** (t/y)
89.9 - 90.0 (841 - 842)	10.0 - 10.1 (94 - 95)	5.23 - 5.29	7212 - 7220

The spent ERU fuel isotopic composition for both uranium and plutonium can be found in Tables 3.4 and 3.5 respectively, and can be compared to the spent UOx fuel composition (Table 1.1). For the uranium composition of the spent ERU fuel, the high fraction of ^{236}U, which is built up upon neutron capture on ^{235}U, is noticeable. The plutonium composition exhibits much higher ^{238}Pu content than spent UOx fuel (see Table 3.1), which is produced upon neutron capture by the large amount of ^{237}Np present in the ERU fuel, that in turn is produced by neutron capture on the significant amounts of ^{236}U. The plutonium grade from spent ERU fuel is only slightly lower than for UOx fuel, but the large quantities of ^{238}Pu are considered problematic (see Section 1.2.1).

Since the isotopic composition and required enrichment do not vary dramatically with the cooling and fabrication times, the batch fractions are also close to constant (Table 3.3). For scenarios with lower cooling times, less ^{234}U is present, thus requiring slightly less enrichment and also allowing a minimally larger ERU batch fraction.

3.1.4 UOx-MOx-ERU Mono-Recycling Fleet

A schematic representation of the UOx-MOx-ERU fleet can be found in Fig. 3.4.

The ranges for the main fleet parameters obtained with the combinations of cooling/fabrication times that were used are reported in Table 3.6, and show that, when both uranium and plutonium

Table 3.4: Uranium Isotopic Composition (%) of reprocessed uranium before and after enrichment for a cooling time of 200 years and for spent ERU fuel (55 GWd/t).

	U234	**U235**	**U236**	**U238**	**U content**
Before Enrichment	0.049	0.808	0.680	98.464	/
Enrichment Factor	1.33	1	0.69	/	/
After Enrichment	0.424	5.287	2.980	91.308	/
Spent ERU	0.08	1.22	3.61	95.09	92.61

Table 3.5: Plutonium isotopic fraction and content (%) of spent (55 GWd/t) ERU fuel

Pu238	**Pu239**	**Pu240**	**Pu241**	**Pu242**	**Am241**	**Pu content**	**Pu grade**
9.49	48.25	22.30	13.44	6.06	0.46	1.38	61.69

are recycled, about 15-20% of UOx reactors can be replaced, leading to a **lower UOx fraction than in an UOx-MOx or UOx-ERU fleet**. Since the origin of the ERU and MOx fuels is the same spent UOx fuel as before, **the isotopic composition and enrichment/plutonium content for ERU and MOx fuel respectively are exactly the same as in the UOx-ERU or UOx-MOx fleets.**

As previously in the UOx-MOx fleet, the MOx batch fraction is higher when both cooling and fabrication times are lower. The sharpest decreases in the MOx fraction are obtained by increasing the fabrication time, an effect that was also observed in the UOx-MOx fleet.

Although the ERU batch fraction is only very slightly time-dependent in the UOx-ERU fleet, this is no longer the case in the UOx-MOx-ERU fleet. The change in the ERU batch fraction is governed by the MOx batch fraction, and is thus more dependent on cooling and fabrication times in the UOx-MOx-ERU fleet. MOx and ERU show a complementary behaviour: if less MOx reactors can be fuelled because of low plutonium grade, more UOx reactors are needed and more uranium is available for ERU reactors, thus raising the ERU fraction.

3.1.5 Comparison of the Mono-Recycling Fleets

The fleet comparison is done here for their ability to save natural uranium and their plutonium and minor actinide production. The comparison is based on the reference case where the UOx assemblies are subjected to a cooling time of 10 years and MOx/ERU fuels are produced and introduced back into the reactor 2 years after the UOx fuel has been reprocessed. This is done to eliminate the effect of extreme outliers with unrealistically long and/or short cooling or fabrication times and provide a realistic estimation.

Equilibrium Conditions

The equilibrium conditions for the mono-recycling cycles are listed in 3.7. The variations of these parameters with respect to cooling and fabrication times are explained above in the respective

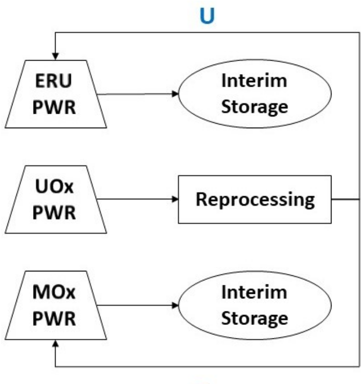


Figure 3.4: Schematic representation of the UOx-MOx-ERU cycle

Table 3.6: Ranges for the main fleet parameters obtained for the UOx-MOx-ERU fleet for the different cooling/fabrication times.

Batch fraction (%) (Fuel fabrication (t/y)) UOx	MOx	ERU	**natU consumption** (t/y)
81.1 - 84.5 (759 - 791)	6.0 - 9.8 (56 - 92)	9.1 - 9.5 (85 - 98)	6503 - 6777

sections. Fleets with MOx reactors have a batch fraction just below 10%, which translates into a **maximum needed MOx fuel fabrication capacity of about 90 t/y**. This amount of fuel fabrication can be served by the current MELOX plant, which is designed to produce up to 150 t/y of MOx fuel [42].

Table 3.7: UOx-MOx fleet equilibrium conditions for the reference case with 10 years cooling and 2 years fabrication time

Fleet	**UOx fraction** (%)	**MOx fraction** (%)	**ERU fraction** (%)	**ERU Enrichment** (%)	**MOx Pu content** (%)	**Pu grade** (%)
UOx-MOx	90.6	9.4	/	/	11.2	60.5
UOx-ERU	89.9	/	10.1	5.23	/	/
UOx-MOx-ERU	82.2	8.6	9.2	5.23	11.2	60.5

Natural Uranium savings

The option that saves the most natural uranium resources is the one where both uranium and plutonium are recycled, with **savings of up to 17.8%** (Table 3.8). Nevertheless, an interesting observation can be made concerning the amount of savings that can be achieved. Once both plutonium and uranium are recycled, ERU and MOx batches are competing to replace a part of the UOx reactors. This results in the fact that **in the UOx-MOx-ERU fleet, the natural uranium savings are smaller than what one could expect by just adding the natural uranium savings from the UOx-MOx fleet and the UOx-ERU fleet**.

Table 3.8: Comparison of natural uranium consumption/savings for the different mono-recycling scenarios for the reference case

	Once-Through	**UOx-MOx**	**UOx-ERU**	**UOx-MOx-ERU**
natU needs (t/y)	8022	7266	7213	6596
Savings (%)	/	9.4	10.1	17.8

Plutonium and Minor Actinide Production

To rank the fleets with regards to plutonium production, we need to specify that, since PWRs are not efficient plutonium burners, and it is thus hard to get rid of this element, **a smaller plutonium production is better for the fleets modelled here.** For scenarios with FRs for example, a large amount of plutonium might be necessary to start the cores of these reactors, in which case more plutonium production is better.

For the minor actinides, a reduction of their production is always a goal, since in no case do they constitute a valuable resource, and must be disposed of (or transmuted). Since minor actinides are the main long term contributors to radiotoxicity and heat production in vitrified waste, their reduction could be an important target to reduce the final storage capacity needs in the future.

The plutonium and minor actinide production for the different fleets can be found in Table 3.9. **While the UOx-ERU fleet does not change the plutonium production by much, the UOx-MOx and UOx-MOx-ERU fleets produce a significant reduction in plutonium production of over a third (compared to the once-through case), with a slight ad-**

vantage for the UOx-MOx fleet. Although the total amounts of plutonium produced in the once-through and UOx-ERU fleet are similar, the plutonium grade is slightly lower in the UOx-ERU case and the ^{238}Pu fraction is significantly higher.

Table 3.9: Plutonium and minor actinide production for the mono-recycling fleets and change with respect to the once-through cycle. Values in bold indicate equilibrium values reached after 1000 years.

Cycle Option	**Pu** (t/y)	**Total MA** (t/y)	**Np** (%)	**Am (241)** (%)	**Cm** (%)
Once-Through	**9.74** - 11.62	1.17 - **2.68**	65.5	23.0 (4.4)	11.5
UOx-MOx	**6.09** - 7.31 (-37.5%)	2.05 - **2.96** (+10.4%)	45.7	36.6 (10.7)	17.8
UOx-ERU	**9.80** - 11.09 (+0.6%)	1.94 - **2.87** (+7.1%)	70.2	19.9 (3.9)	9.9
UOx-MOx-ERU	**6.46** - 7.83 (-33.7%)	2.12 - **3.11** (+16.0%)	52.2	32.3 (9.4)	15.6

For all cases with recycling of fissile material, the MA production is increased overall compared to the once-through case. In the ERU reactor, the Np production is increased compared to all other reactor types, which is driven by neutron capture on the significant amounts of ^{236}U present in the fuel due to re-enrichment.

MOx reactors mainly increase the production of Am and Cm. MOx reactors create ^{243}Am by neutron capture on ^{242}Pu, so its production is favoured in MOx reactors with high plutonium content and plutonium from high burnup fuel. The production of ^{241}Am is due to the decay of ^{241}Pu, therefore, a higher MOx batch fraction greatly increases the fraction of ^{241}Am in the total Am production, as larger quantities of Pu, and therefore ^{241}Pu , are in the reactors at each time.

The total minor actinide production in a fleet with MOx reactors is strongly dependent on the cooling/fabrication times, because those times determine how much ^{241}Pu can decay before being put back into the reactor and therefore also the production of ^{241}Am and by extension ^{237}Np. Recycling plutonium can be seen as a race against time: the faster it is re-introduced in the reactor, the larger part of ^{241}Pu can fission instead of creating ^{241}Am. Fig. 3.5 shows that short times are essential to reduce the MA production, and that if times get very long, mono-recycling fleets can produce in excess of 3.7 t/y at equilibrium.

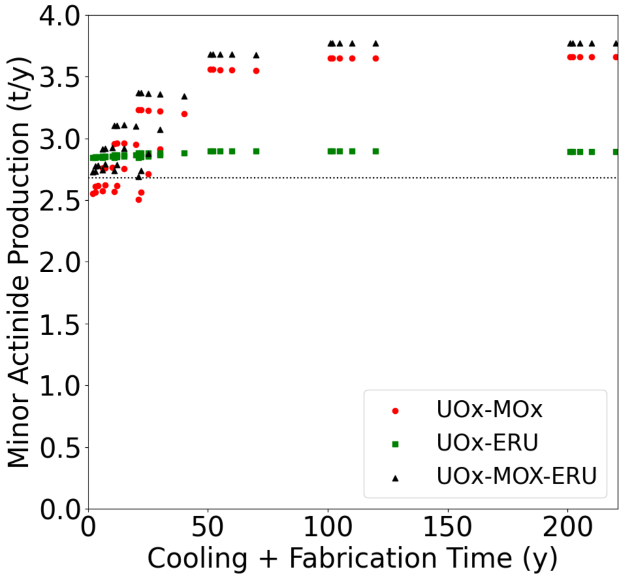


Figure 3.5: Variation of the total minor actinide production (equilibrium value) as a function of cooling + fabrication times for the mono-recycling cycles. The black dotted line indicates the production for the once-through fleet.

If we compare the minor actinide production from a dynamic scenario simulation for a UOx-MOx-

ERU fleet, as performed in [43], we can see that the minor actinide production is underestimated since we find 3.11 t/y as a maximum value compared to 3.3 t/y for the reference. This can be attributed to the fact that historic fuels have lower burnups and, on average, longer cooling times. Even if lower burnups should favour lower minor actinide production, longer cooling times can quickly lead to higher minor actinide production as Fig. 3.5 shows.

The once-through equilibrium values correspond well with results from [43]: for the Pu production, the reference of 9.9 t/y lies well within the determined range of 9.74 - 11.62 t/y. The MA production values are consistent as well: the reference of 2.7 t/y is nearly identical with the range maximum of 2.68 t/y.

3.2 Partial Multi-Recycling Scenarios

This section investigates scenarios where spent fuel, produced from reprocessed materials, is reprocessed again to produce new fuel. A distinction should be made between partial and full multi-recycling fuel cycles: in a full multi-recycling cycle, the total flux of one valuable element (for example plutonium) is recycled, leading to a closed cycle for this material, while in a partial multi-recycling scenario, some of the material accumulates in spent fuel stocks.

Some of the following cycle options are optimised such that they could be achieved with current cycle facilities, whereas other options are only possible once new facilities are built. The limiting factors are always those facilities that are handling plutonium, namely we will use a 50 t/y limit on the reprocessing of MOx SF and the limit on ^{238}Pu at the reprocessing and fuel fabrication steps, as described in Section 1.2.1. To achieve full multi-recycling scenarios, be it in PWRs or SFRs, new fuel cycle facilities, with improved processes that are adapted to the changing fuel composition, need to be designed, built and commissioned in the future.

The simulations are performed for the same cooling/fabrication times as for the mono-recycling fleets (cooling: 1, 2, 5, 10, 20, 50, 100, 200 years; fabrication: 1, 2, 5, 10, 20 years), except if explicitly noted otherwise. As before, equilibrium fleet conditions and natural uranium consumption as well as plutonium and minor actinide production of the reference case (cooling time 10 years, fabrication time 2 years) are summarized for all partial multi-recycling scenarios in Tables 3.17 and 3.18 to allow for a better comparison between cycles.

3.2.1 UOx-MOx cycle with partial recycling of MOx SF

This fleet was modelled under the assumption of using currently existing fuel cycle facilities with a limit on MOx SF reprocessing of 50 t/y. A schematic representation of this cycle option can be found in Fig. 3.6. SEPAR calculations were used to iteratively find the maximum MOx SF reprocessing while keeping the fresh MOx plutonium content below the safety limit of 12%.

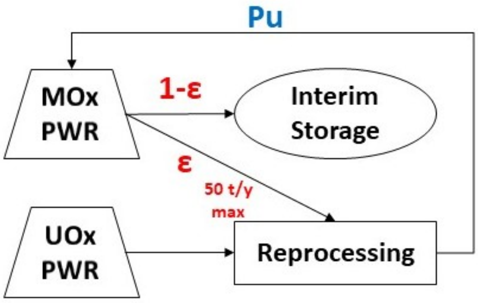


Figure 3.6: Schematic representation of the UOx-MOx cycle with partial recycling of MOx SF. The reprocessed MOx fraction is denoted as ε.

The results show that **for shorter cooling/fabrication times, more spent MOx can be reprocessed, which is due to the higher plutonium grade that can be extracted from spent fuel with shorter times. As for the UOx-MOx fleet, the grade is most sensitive to the fabrication time**, which in this case translates to the fact that the reprocessed MOx fraction decreases the quickest with increased fabrication times. In general, the limit on ε, for the high burnups of 55 GWd/t used here, is almost always the 12% safety limit rather than the 50 t/y reprocessing capacity. Only for two cases with the shortest possible fabrication time (cooling time of 1 or 2 years and fabrication time of 1 year), the limit was found to be the reprocessing capacity. The results in reality would be different since the historic stock of SF in France has significantly lower burnup.

The ranges for the main fleet parameters at equilibrium for the combinations of cooling/fabrication times used here are reported in Table 3.10. Depending on the times, the reprocessed MOx SF fraction varies between 0%, for those cases where the safety limit of 12% was already surpassed without MOx reprocessing, and 41%, in cases where the upper limit of 50 t/y of MOx reprocessing is reached. The results also show that the MOx batch fraction is slightly increased compared to the mono-recycling fleet (Table 3.2) since more plutonium is available in the fleet, and that the natural uranium consumption is slightly reduced compared to the mono-recycling fleet.

Table 3.10: Ranges for the main fleet parameters obtained for the UOx-MOx fleet with partial MOx reprocessing for the different cooling/fabrication times.

Batch fraction (%) (Fuel fabrication (t/y))		ε (%)	**natU consumption** (t/y)
UOx	MOx		
86.9 - 93.3 (813 - 873)	6.7 - 13.1 (62 - 123)	0 - 41	6972 - 7487

3.2.2 UOx-MOx-ERU fleet with partial MOx SF reprocessing

This fuel cycle option is the second option that is assumed to be possible with current cycle facilities, and is therefore of special interest. It is analogous to the previous cycle option that was presented with the only addition being some ERU reactors, however no ERU fuels are reprocessed (see Fig. 3.7).

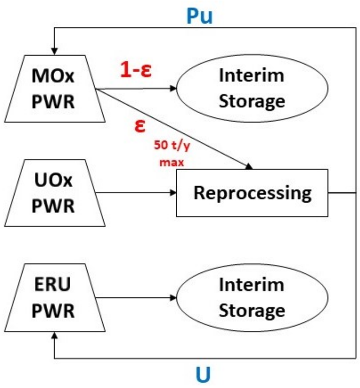


Figure 3.7: Schematic representation of the UOx-MOx-ERU cycle with partial recycling of MOx SF. The reprocessed MOx fraction is denoted as ε.

The ranges for the main fleet parameters at equilibrium can be found in Table 3.11. The batch fractions are similar to the UOx-MOx-ERU mono-recycling fleet (Table 3.6) with higher MOx fraction, at the expense of UOx. The MOx reprocessed fraction is slightly larger than in the UOx-MOx fleet with partial MOx recycling, but the absolute reprocessed fuel mass is about the same (the batch fraction is just smaller), because the Pu grade from ERU SF is similar to that from UOx SF.

Since the ^{238}Pu content could be a crucial limit, it is important to note that results (not shown here) confirm that the equilibrium content of ^{238}Pu is certainly higher than without MOx recycling, but the maximum value, which is reached for very short cooling times, lies below 3.4%. This means that, **the ^{238}Pu content should not pose a problem for the reprocessing process at La Hague and the fabrication process at MELOX, provided that MOx and UOx fuels are reprocessed together.**

Table 3.11: Ranges for the main fleet parameters obtained for the UOx-MOx-ERU fleet with partial MOx reprocessing for the different cooling/fabrication times.

Batch fraction (%) (Fuel fabrication (t/y)) UOx	MOx	ERU	ε (%)	**natU consumption** (t/y)
79.0 - 84.5 (739 - 791)	6.0 - 12.1 (56 - 114)	8.9 - 9.5 (83 - 89)	0 - 44	6337 - 6777

3.2.3 UOx-MOx-ERU fleet with plutonium recycling from ERU SF

In this cycle option, plutonium from both UOx and ERU SF is recovered to produce fresh MOx fuel, but uranium is only reprocessed from spent UOx (see Fig. 3.8). The ranges of the main equilibrium parameters for the cycle option are listed in Table 3.12.

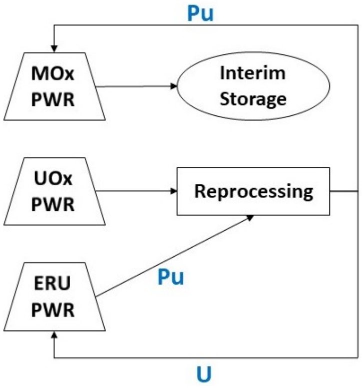


Figure 3.8: Schematic representation of the UOx-MOx-ERU cycle with plutonium recycling from ERU SF. All ERU SF is sent to reprocessing, the "Pu" label on the arrow means that only the plutonium fraction is used for further fuel fabrication.

Table 3.12 shows that compared to the UOx-MOx-ERU case with plutonium recycling from MOx SF (Table 3.11), **this cycle does not allow such high MOx fractions since the plutonium mass recovered from ERU SF is much lower than the one recoverable from MOx SF, but the higher ERU fraction allows to reduce the natural uranium consumption.** The ERU enrichment is equal to the one found in the UOx-MOx-ERU mono-recycling fleet since the material to produce the ERU fuel is exactly the same, the same is true for the MOx Pu content, because the ERU SF plutonium grade is just slightly lower than that of UOx SF.

Table 3.12: Ranges for the main fleet parameters obtained for the UOx-MOx-ERU fleet with plutonium recycling from ERU SF for the different cooling/fabrication times.

Batch fraction (%) (Fuel fabrication (t/y)) UOx	MOx	ERU	**MOx Pu content** (%)	**natU consumption** (t/y)
80.1 - 83.8 (750 - 748)	6.8 - 10.9 (63 - 102)	9.0 - 9.4 (84 - 88)	10.1 - 17.0	6425 - 6724

The equilibrium isotopic composition of the plutonium fluxes in the fleet is interesting to analyze since we know that the ^{238}Pu fraction can be a limiting factor. Table 3.13 shows the plutonium isotopic composition of spent ERU fuel and the equilibrium input isotopic composition in fresh MOx fuel. We can see that the ERU spent fuel has a ^{238}Pu close to 10% which can pose severe problems at the reprocessing step. Even if UOx and ERU fuels were mixed during reprocessing to dilute the ^{238}Pu in solution, the fraction would still be above 3.4%. **This shows that this fuel cycle option is not possible with current facilities, at least not if all ERU SF should be reprocessed.**

Table 3.13: Plutonium equilibrium isotopic composition (%) for spent (55 GWd/t) ERU fuel and fresh MOx fuel for the UOx-MOx-ERU fleet with plutonium recycling from spent ERU fuel. Data for the reference case.

	Pu238	**Pu239**	**Pu240**	**Pu241**	**Pu242**	**Am241**	**Pu+Am241 Total**
Spent ERU	9.49	48.31	22.27	13.44	6.05	0.46	1.38
Fresh MOx	3.59	51.77	26.87	8.63	8.27	0.88	11.19

3.2.4 UOx-MOx-ERU Fleet with Plutonium and Uranium Recycling from ERU SF

For this fleet, plutonium and uranium are both recycled from UOx and ERU SF (see Fig. 3.9), meaning that this cycle option is not possible with current facilities. **This cycle option could only be modelled for cooling times up to 100 years because else, the ^{238}Pu and ^{234}U contents would be out of range of the training data.** The ranges for the main equilibrium parameters for the different fabrication/cooling times are listed in Table 3.14. The ERU batch fraction is quite stable at a high level while longer cooling/fabrication times decrease plutonium grade and with it the MOx batch fraction, which is compensated by higher UOx batch fraction. The high ERU fraction and the MOx fraction similar to the UOx-MOx-ERU fleet with plutonium recycling from ERU SF makes it possible to reduce the natural uranium consumption even more compared to the latter cycle option (Table 3.12).

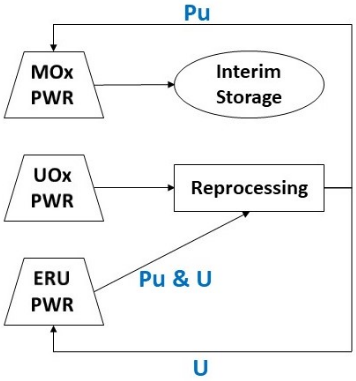


Figure 3.9: Schematic representation of the UOx-MOx-ERU cycle with plutonium and uranium recycling from ERU SF.

Table 3.14: Ranges for the main fleet parameters obtained for the UOx-MOx-ERU fleet with plutonium and uranium recycling from ERU SF for the different cooling/fabrication times.

Batch fraction (%) (Fuel fabrication (t/y))			**MOx Pu content**	**ERU enrichment**	**natU consumption**
UOx	MOx	ERU	(%)	(%)	(t/y)
78.6 - 82.2 (736 - 769)	6.8 - 11.0 (64 - 103)	10.5 - 11.0 (98 - 103)	10.1 - 17.0	5.73 - 5.99	6303 - 6596

3.2.5 UOx-MOx-ERU Fleet with Plutonium Recycling from ERU and MOx SF

In this cycle option, all plutonium is, at least partially, reprocessed from all fuels in the cycle (see Fig. 3.10). The ranges obtained for the main fleet parameters for the cooling/fabrication times used are listed in Table 3.15. Although Pu from ERU SF has a higher grade than the one from MOx, the limited quantities available do not allow to increase the fresh MOx Pu grade significantly, therefore the MOx Pu content is nearly identical with the one from the UOx-MOx-ERU fleet with partial MOx recycling (Section 3.2.1).

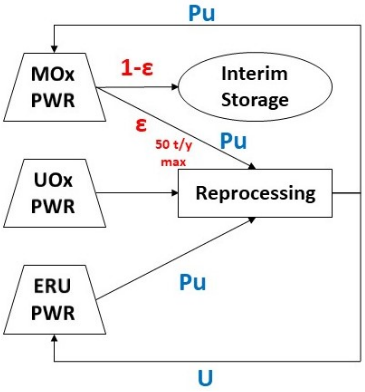


Figure 3.10: Schematic representation of the UOx-MOx-ERU cycle with plutonium recycling from ERU and MOx SF. The reprocessed MOx fraction is denoted as ε.

Table 3.15: Ranges for the main fleet parameters obtained for the UOx-MOx-ERU fleet with plutonium recycling from ERU and MOx SF for the different cooling/fabrication times.

Batch fraction (%) (Fuel fabrication (t/y))			**MOx Pu content** (%)	ε (%)	**ERU enrichment** (%)	**natU consumption** (t/y)
UOx	MOx	ERU				
78.0 - 83.8 (730 - 784)	6.8 - 11.0 (63 - 103)	8.8 - 9.4 (82 - 88)	11.5 - 17.0	0 - 40	5.24 - 5.29	6258 - 6724

3.2.6 UOx-MOx-ERU Fleet with Plutonium and Uranium Recycling from ERU SF and Plutonium Recycling from MOx SF

This fleet recycles all valuable material (at least partially), excluding uranium from MOx, which is depleted and therefore not interesting (see Fig. 3.11). The ranges obtained for the main fleet parameters for the cooling/fabrication times used are listed in Table 3.16. As before, when U and Pu from ERU SF were recycled (section 3.2.4), only cooling times up to 100 years could be modelled. The cycle option is very similar to the same one without uranium recycling from ERU SF but with a higher ERU fraction, that allows lower natU consumption.

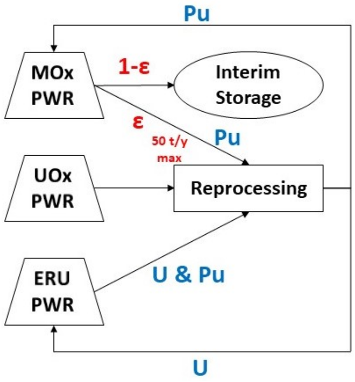


Figure 3.11: Schematic representation of the UOx-MOx-ERU cycle with uranium and plutonium recycling from ERU and plutonium recycling from MOx SF. The reprocessed MOx fraction is denoted as ε.

3.2.7 Comparison of the Partial Multi-Recycling Fleets

Equilibrium Conditions and Natural Uranium Consumption

The first observation that can be made from the results of the partial multi-recycling scenarios is that, compared to the respective mono-recycling fleets, **partial multi-recycling only allows very small shifts to lower UOx batch fractions, which translates to only small additional natural uranium savings (compare Tables 3.7/3.8 and 3.17).**

Table 3.16: Ranges for the main fleet parameters obtained for the UOx-MOx-ERU fleet with plutonium recycling from ERU and MOx SF and uranium recycling from ERU SF for the different cooling/fabrication times.

Batch fraction (%) (Fuel fabrication (t/y))			**MOx Pu content** (%)	ε (%)	**ERU enrichment** (%)	**natU consumption** (t/y)
UOx	MOx	ERU				
76.5 - 82.2 (716 - 769)	6.8 - 13.3 (64 - 125)	10.2 - 11.0 (95 - 103)	11.5 - 16.9	0 - 40	5.73 - 5.99	6138 - 6596

Table 3.17: Partial multi-recycling fleet equilibrium conditions for the reference case with 10 years cooling and 2 year fabrication time. E(B) indicates that element E is recycled from batch B. Note that for Pu(MOx) only a fraction of plutonium is recycled.

Cycle Option	**UOx/MOx/ERU batch fractions** (%)	**ERU Enrichment** (%)	**MOx Pu Content** (%)	**MOx SF Reprocessing Fraction** (%)	**natU Consumption** (t/y)
UOx-MOx, Pu(MOx)	89.8 / 10.2 / N.A.	/	12.0	21.0	7201
UOx-MOx-ERU, Pu(MOx)	81.5 / 9.3 / 9.2	5.24	12.0	21.0	6542
UOx-ERU, U(ERU)	88.2 / N.A. / 11.8	5.76	/	/	7078
UOx-MOx-ERU, Pu(ERU)	81.3 / 9.5 / 9.1	5.24	11.2	/	6525
UOx-MOx-ERU, Pu & U(ERU)	79.8 / 9.6 / 10.6	5.76	11.2	/	6398
UOx-MOx-ERU, Pu(ERU, MOx)	80.6 / 10.4 / 9.0	5.24	12.0	21.0	6466
UOx-MOx-ERU, Pu(ERU, MOx) U(ERU)	79.0 / 10.5 / 10.5	5.76	12.0	22.0	6335

For all cases, two different equilibrium ERU enrichment values are possible: **if uranium is only recycled from UOx, we have 5.24% enrichment, if uranium from ERU is recycled as well, the needed enrichment rises to 5.76%.** The fresh MOx plutonium content changes if Pu from ERU or MOx SF are recycled, in which case **the Pu from MOx tends to increase the content** (up to 12%, since this is the set limit) **and the Pu from ERU tends to decrease the content, but generally the quantities of Pu from ERU are too small compared to Pu from MOx SF to be able to make a real change**. Cases with MOx partial reprocessing but with $\varepsilon = 0$ have MOx Pu contents higher than 12%, meaning that these cases are not feasible in reality.

The MOx batch fraction is highly time-sensitive, an effect that was already observed for the mono-recycling cycles but that is more notable when plutonium is multi-recycled. **Generally, the faster fuels are reprocessed and re-fabricated, the higher the MOx batch fraction and the higher the natU savings, and for this, short fabrication times are most important.**

Plutonium and Minor Actinide Production

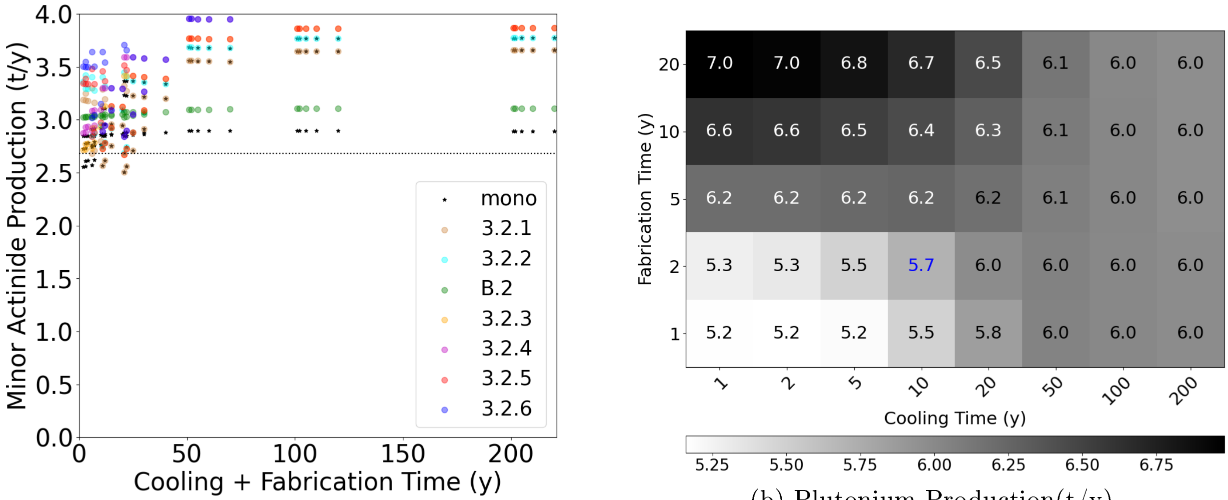
The results from Table 3.18 show that partial multi-recycling scenarios allow for a reduction in the plutonium production and the best cycle options for reduced plutonium production are the ones with partial MOx recycling. Introducing ERU reactors does not penalize plutonium production greatly. **Compared to mono-recycling scenarios, the achieved reduction in plutonium production is limited: for the best case, plutonium production is reduced by merely 6% with respect to the best mono-recycling case**. The best cooling/fabrication time combination to reduce plutonium production is for short times, where short fabrication times are the most important factor (Fig. 3.12b)

Table 3.18: Pu and MA production for the partial multi-recycling fleets. E(B) indicates that element E is recycled from batch B. Note that for Pu(MOx) only a fraction of plutonium is recycled. Values in bold indicate equilibrium values reached after 1000 years.

Cycle Option	**Pu** (t/y)	**Total MA** (t/y)	**Np** (%)	**Am (241)** (%)	**Cm** (%)
UOx-MOx, Pu(MOx)	**5.69** - 6.33	2.65 - **3.19**	43.4	38.4 (11.7)	18.2
UOx-MOx-ERU, Pu(MOx)	**6.09** - 6.85	2.73 - **3.32**	49.9	34.0 (10.3)	16.1
UOx-ERU, U(UOx, ERU)	**9.81** - 10.65	2.53 - **3.05**	74.1	17.3 (3.5)	8.5
UOx-MOx-ERU, Pu(ERU)	**6.11** - 6.87	2.63 - **3.13**	50.7	33.2 (9.8)	16.1
UOx-MOx-ERU, Pu & U(ERU)	**6.12** - 6.92	2.80 - **3.30**	55.3	30.2 (9.0)	14.6
UOx-MOx-ERU, Pu(ERU, MOx)	**5.71** - 6.39	2.80 - **3.36**	48.4	35.7 (10.8)	16.5
UOx-MOx-ERU, Pu(ERU, MOx) U(ERU)	**5.68** - 6.40	2.98 - **3.54**	52.8	32.1 (9.9)	15.1

The MA production for the partial multi-recycling cycles shows an increase over mono-recycling cases, nevertheless, the increase is limited (Fig. 3.12a) since the recycling is limited. **Recycling more reprocessed uranium, from ERU SF in this case, increases the Np production through neutron capture on ^{236}U.** As before, recycling more Pu increases the Am and Cm production. While the fractions of Am and Cm are similar to the mono-recycling cases, the absolute values are higher since the total MA production is higher.

A major reduction in minor actinide production can be achieved for very short cooling/fabrication times, Fig. 3.12a shows the large range of minor actinide production. This phenomenon was already explained for the mono-recycling UOx-MOx fleet (section 3.1.2) and is even more important here since larger quantities of plutonium are involved.



(a) Minor Actinide Production(t/y)

(b) Plutonium Production(t/y)

Figure 3.12: Minor actinide and plutonium production as a function of cooling/fabrication times. Panel (a) shows values for all partial mulit-recycling cases, the legend shows the section where they are presented for simplicity. Panel (b) shows the results from the cycle with plutonium recycling from ERU and MOx as an example, the blue value is for the reference case.

3.3 Full Multi-Recycling Scenarios

In this section, scenarios with full multi-recycling of plutonium and/or uranium in PWRs are modelled. As shown in the partial multi-recycling cases, recycling of all plutonium in the fleet cannot be achieved using standard UOx, MOx and ERU fuels since the plutonium grade would drive the plutonium content in MOx fuel above 12%. To close the plutonium cycle, the scenarios investigated here use the MIX fuel concept that relies on an enriched uranium support for the plutonium based fuel to compensate for the degraded plutonium grade. The MIX reactors are assumed to have the same load factor, thermal and electrical power, core mass, EFPD and refuelling fractions as the reactors used previously (see at the beginning of Chapter 3). The plutonium content in fresh MIX fuel is constant at 8%, which is a value similar to today's MOx fuels [19].

Contrary to plutonium, uranium multi-recycling does not require novel fuel designs, instead normal ERU fuels are used here. All reprocessed uranium is going to ERU fuels, although MIX fuels with re-enriched uranium could also be an option for further studies. For the natural uranium consumption calculation, the enriched uranium support is assumed to come from natural uranium.

As previously, the equilibrium fleet conditions and natural uranium consumption as well as plutonium and minor actinide production for the reference case with 10 years cooling and 2 years fabrication time are summarized for all full multi-recycling scenarios in Tables 3.24 and 3.25.

3.3.1 UOx-MIX fleet

A schematic representation of this cycle can be found in Fig. 3.13. The MIX fresh and spent fuel isotopic composition for plutonium and the required enrichment can be found in Table 3.19. **The composition shows the degradation of the plutonium with extremely high ^{238}Pu, ^{240}Pu and ^{242}Pu contents which make it such that, on top of 8% plutonium content, a uranium enrichment above 4% is needed.**

Table 3.19: Isotopic composition (%) of fresh and spent (55 GWd/t) MIX fuel for a case with 10 years cooling time and 2 years fabrication time

	Pu+Am241	Pu238	Pu239	Pu240	Pu241	Pu242	Am241	Enrichment
Fresh	8.0	3.9	36.7	27.0	8.6	22.8	0.9	4.31
Spent	6.5	4.4	29.2	25.0	14.4	25.7	1.4	2.25

The UOx-MIX fleet behaves very differently as a function of cooling and fabrication times than

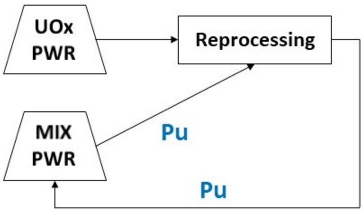
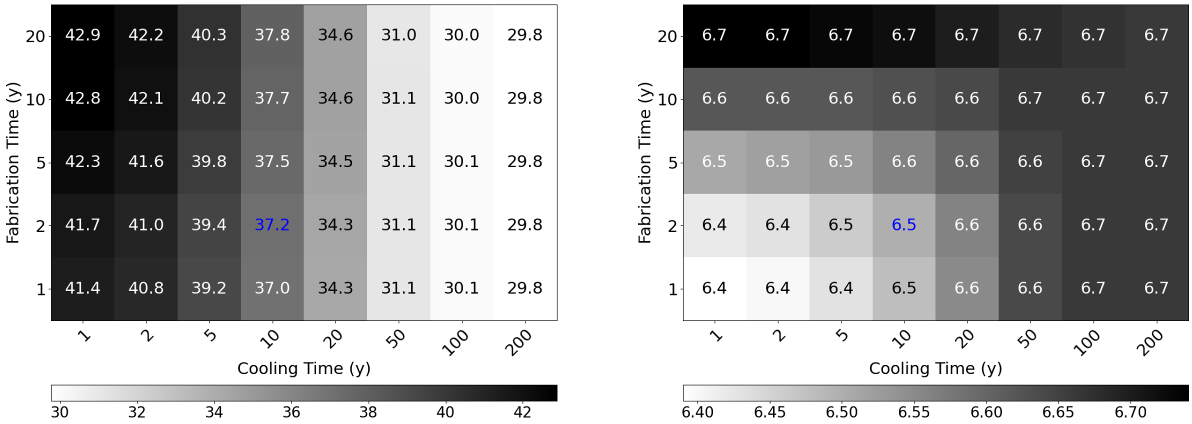


Figure 3.13: Schematic representation of the UOx-MIX cycle. All MIX SF is sent to reprocessing, the "Pu" label on the arrow means that only the plutonium fraction is used for further fuel fabrication.

the UOx-MOx fleet for example. **Since the plutonium content is fixed in MIX fuel and reactivity is adjusted by uranium enrichment, it is the total available mass of plutonium that determines the MIX batch fraction.** To explain the variation of the MIX fraction in the fleet with changing cooling and fabrication times (Fig. 3.14a), one has to understand how the plutonium quantity and grade are affected.

Generally, longer times, but especially longer fabrication times, reduce the plutonium grade, as was shown for the mono-recycling cases. At the same time, longer times increase the amount of plutonium (^{241}Pu and ^{238}Pu) that decays, so **with longer times, the total amount of plutonium is reduced.** Finally, **the grade**, and therefore mainly the fabrication time, **changes the efficiency of plutonium burning through fission, and therefore the residual plutonium content in the fuel. Higher residual plutonium content in the fuel means that more MIX reactors can be fuelled**, regardless of plutonium grade (Fig. 3.14b), contrary to the case of the UOx-MOx fleet.

Combining these factors, we observe that for given cooling time, an increase in fabrication time increases the MIX fraction due to higher residual plutonium content in the MIX fuel. On the other hand, for given fabrication time, an increase in cooling time decreases the MIX fraction due to more plutonium decay.



(a) MIX fraction (%)

(b) Spent MIX Pu content (%)

Figure 3.14: Variation of the MIX batch fraction and spent MIX plutonium content as a function of cooling and fabrication time. Values in blue are for the reference case.

Table 3.20 lists the ranges of the main fleet parameters obtained with the cooling/fabrication times and shows that **the required MIX enrichment is on the order of usual UOx enrichments for a Pu content of 8%.** Since all Pu is recycled in MIX fuels with a constant content, **the MIX fraction is much higher than the MOx fractions for fleets previously described. Because both UOx and MIX fuels require a high enrichment, the natU consumption is on the order of the once-through fleet and can even become higher.**

Table 3.20: Ranges for the main fleet parameters obtained for the UOx-MIX fleet for the different cooling/fabrication times.

Batch fraction (%) (Fuel fabrication (t/y))		**MIX enrichment** (%)	**natU consumption** (t/y)
UOx	MIX		
57.1 - 70.2 (534 - 657)	29.8 - 42.9 (279 - 401)	3.98 - 5.56	7337 - 8454

3.3.2 UOx-MIX-ERU fleet with plutonium recycling from ERU SF

This cycle is the result of adding ERU reactors, fed only by uranium from UOx SF, to the previous fleet (Fig. 3.15). To keep the plutonium cycle closed, plutonium from ERU SF is also used in MIX production. The ranges for the main fleet parameters obtained with the cooling/fabrication times are listed in Table 3.21 and show that this cycle is very similar to the previous one. **The ERU reactors displace UOx reactors but their fraction is small. MIX enrichment is similar to the UOx-MIX fleet** since the small amounts of higher-grade Pu from ERU SF cannot change the overall isotopic composition due to the large flux of Pu from MIX SF. **The ERU fraction is able to bring the natU consumption well below that of the once-through fleet.**

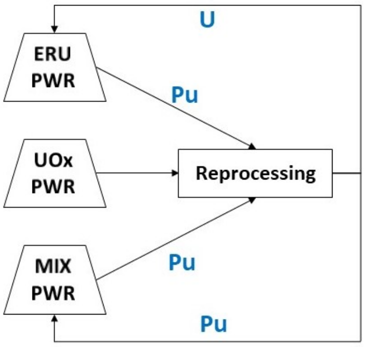


Figure 3.15: Schematic representation of the UOx-MIX-ERU cycle with plutonium recycling from ERU SF. All MIX SF is sent to reprocessing, the "Pu" label on the arrow means that only the plutonium fraction is used for further fuel fabrication.

Table 3.21: Ranges for the main fleet parameters obtained for the UOx-MIX-ERU fleet with plutonium recycling from ERU SF for the different cooling/fabrication times.

Batch fraction (%) (Fuel fabrication (t/y))			**MIX enrichment** (%)	**ERU enrichment** (%)	**natU consumption** (t/y)
UOx	MIX	ERU			
51.2 - 63.2 (479 - 591)	29.8 - 43.1 (279 - 403)	5.7 - 7.0 (54 - 66)	3.97 - 5.54	5.23 - 5.29	6852 - 7980

Except for higher ^{238}Pu content in both fresh and spent MIX fuel, the plutonium recycling from ERU SF does not have a significant effect, as can be seen when the MIX isotopic compositions of the UOx-MIX fleet (Table 3.19) are compared with the UOx-MIX-ERU fleet (Table 3.22).

3.3.3 UOx-MIX-ERU fleet with plutonium and uranium recycling from ERU SF

For this fleet, both plutonium and uranium from spent ERU fuel are recovered and reintroduced into MIX and ERU fuels respectively (see Fig. 3.16). As previously when uranium from ERU was recycled, the cycle could only be modelled for cooling times up to 100 years. **Table 3.23 shows that introducing more reprocessed uranium back into the reactors increases the ERU batch fraction at the expense of the UOx fraction, while the MIX fraction can be increased slightly due to more available plutonium.** The ERU enrichment is increased

Table 3.22: Isotopic composition (%) of fresh and spent MIX fuel for a case with 10 years cooling time and 2 years fabrication time

	Pu+Am241	**Pu238**	**Pu239**	**Pu240**	**Pu241**	**Pu242**	**Am241**	**Enrichment**
Fresh	8.0	4.3	36.8	26.9	8.6	22.5	0.9	4.30
Spent	6.5	4.7	29.3	25.0	14.4	25.3	1.4	2.24

compared to the previous fleet since uranium recycled from ERU SF needs to be over-enriched even more than uranium from UOx SF. The recycling of both uranium and plutonium from ERU also allows for an additional small reduction in natU consumption.

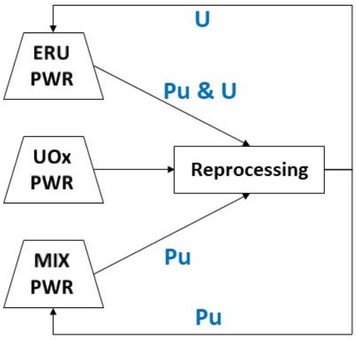


Figure 3.16: Schematic representation of the UOx-MIX-ERU cycle with plutonium and uranium recycling from ERU SF. All MIX SF is sent to reprocessing, the "Pu" label on the arrow means that only the plutonium fraction is used for further fuel fabrication.

Table 3.23: Ranges for the main fleet parameters obtained for the UOx-MIX-ERU fleet with plutonium and uranium recycling from ERU SF for the different cooling/fabrication times.

Batch fraction (%) (Fuel fabrication (t/y))			**MIX enrichment** (%)	**ERU enrichment** (%)	**natU consumption** (t/y)
UOx	MIX	ERU			
50.1 - 61.6 (467- 577)	30.0 - 43.2 (281- 404)	6.7 - 8.4 (62 - 78)	3.96 - 5.52	5.73 - 5.99	6766 - 7893

3.3.4 UOx-MIX-ERU fleet with plutonium and uranium recycling from both ERU and MIX SF

In this cycle option, all uranium and plutonium is recycled from UOx, ERU and MIX fuels (Fig. 3.17). **The high residual enrichment of spent MIX fuel (Table 3.22) makes reprocessing uranium from spent MIX fuel highly interesting.** The goal was to simulate the fleet for the same cooling/fabrication times as previously, but the ^{238}Pu content in ERU SF (exceeding 11%) yields very high ^{234}U contents in fresh ERU (exceeding 0.3%), and both are out of the range in the training data for many combinations of cooling/fabrication times. To investigate the effect of time on the fleet equilibrium conditions, new training data needs therefore to be generated from APOLLO2 simulations with higher ^{238}Pu and ^{234}U input fractions for MIX and ERU fuels respectively.

Nevertheless, for the reference case, all isotopic compositions were in range of the training data, so the results for this case can be trusted.

3.3.5 Comparison of Full Multi-Recycling Scenarios

Equilibrium conditions and Natural Uranium consumption

For the full multi-recycling fleets, it can be noticed that **the fraction of plutonium-based fuels (MIX) is much higher than in the mono-recycling cases (MOx)**. This comes from the fact that the plutonium content in MIX fuels is constant and lower than in the simulated MOx

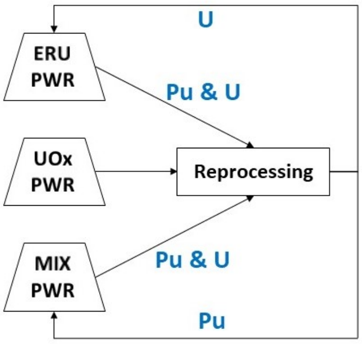


Figure 3.17: Schematic representation of the UOx-MIX-ERU cycle with plutonium and uranium recycling from both ERU and MIX SF.

fuels, and the addition of enriched uranium allows for more MIX reactors in the fleet at a given available Pu mass. In addition, the quantities of plutonium available in the fleet are much higher in full multi-recycling scenarios compared to mono-recycling or partial multi-recycling because all plutonium is recycled.

The high proportion of MIX in the equilibrium fleets entails that large amounts of plutonium based fuels need to be produced. With 37% MIX fraction in the fleet, **346 t/y of MIX fuels need to be produced, which is more than double the amount of plutonium based fuels that can be produced at the MELOX facility. Knowing that plutonium-handling facilities are generally much more expensive than facilities that only handle uranium, this could increase the cycle cost significantly.**

The natU consumption for most full multi-recycling scenarios are only slightly lower to the once-through case because, except for ERU, all fuels in these fleets need considerable amounts of enriched uranium from natU origin. **The only very notable reduction in natural uranium consumption can be achieved if uranium is recycled from MIX fuels since they contain a very high residual enrichment compared to UOx fuels.**

Table 3.24: Full multi-recycling fleet equilibrium conditions for the reference case with 10 years cooling and 2 year fabrication time. E(B) indicates that element E is recycled from batch B. Plutonium from MIX spent fuel is always recycled. Fresh MIX plutonium content is 8% for all cases.

Cycle Option	**UOx/MIX/ERU batch fractions** (%)	**MIX Enrichment** (%)	**ERU Enrichment** (%)	**natU Consumption** (t/y)
UOx-MIX	62.8 / 37.2 / N.A.	4.37	/	7650
UOx-MIX-ERU Pu(ERU)	56.3 / 37.4 / 6.3	4.35	5.24	7127
UOx-MIX-ERU, Pu & U(ERU)	55.2 / 37.5 / 7.4	4.32	5.76	7030
UOx-MIX-ERU, Pu & U(ERU) U(MIX)	40.5 / 37.9 / 21.6	4.25	5.61	5834

Plutonium and Minor Actinide Production

Even if all plutonium from spent fuel is recycled into fresh fuel, **there is still overall production of plutonium in the fleet. This is not a computational error but this plutonium comes from the decay of ^{244}Cm, ^{243}Cm and ^{242}Cm** that decay into ^{240}Pu, ^{239}Pu and ^{238}Pu with half-lives of 18, 30 and 0.4 years respectively. This means that the **net production of plutonium happens exclusively in the vitrified waste packages** and is one order of magnitude smaller than the plutonium production in mono-recycling scenarios.

The minor actinide production of the full multi-recycling fleets is characterized by **lower Np**

Table 3.25: Plutonium and minor actinide production for the full multi-recycling fleets. E(B) indicates that element E is recycled from batch B. Plutonium from MIX is always recycled. Values in bold indicate equilibrium values reached after 1000 years.

Cycle Option	**Pu** (t/y)	**Total MA** (t/y)	**Np** (%)	**Am (241)** (%)	**Cm** (%)
UOx-MIX	0.09 - **0.50**	**3.88** - 4.28	22.7	50.2 (10.9)	27.1
UOx-MIX-ERU Pu(ERU)	0.08 - **0.50**	**3.99** - 4.38	25.3	48.5 (10.7)	26.2
UOx-MIX-ERU, Pu & U(ERU)	0.08 - **0.50**	**4.10** - 4.49	27.9	46.8 (10.4)	25.3
UOx-MIX-ERU, Pu & U(ERU) U(MIX)	0.08 - **0.51**	**4.54** - 4.79	34.2	42.7 (9.7)	23.1

fraction in the total production compared to mono-recycling or partial multi-recycling fleets. This is because full plutonium multi-recycling increases the Am and Cm production dramatically. Even if U from ERU of MIX SF is recycled, the Np fraction is increased but remains well below the values for mono- or partial multi-recycling cycles.

Although the Cm fraction is increased artificially since only the in-reactor production is considered, it can be a limiting factor for the vitrified waste packages due to its low solubility and stability in the glass matrix [14].

The production of MA is greatly influenced by cooling/fabrication times, an effect that was already observed in mono- and partial multi-recycling cycles, but shows even stronger with full multi-recycling. The large variations with the times can be seen in Fig. 3.18 and the magnitude of the effects compared to previously discussed fleets. **For realistic cooling/fabrication times, multi-recycling scenarios produce significantly more minor actinides than mono-recycling scenarios.** For the UOx-MIX fleet, results from the literature [19], obtained with a dynamic simulations, find the same equilibrium batch fractions and MA production values between 4.2 and 4.5 t/y. The simulation however takes into account historic spent fuel and variable cooling times, with a minimum cooling time of 5 years and fabrication time of 2 years, so that a direct comparison is not possible, but the determined ranges are consistent.

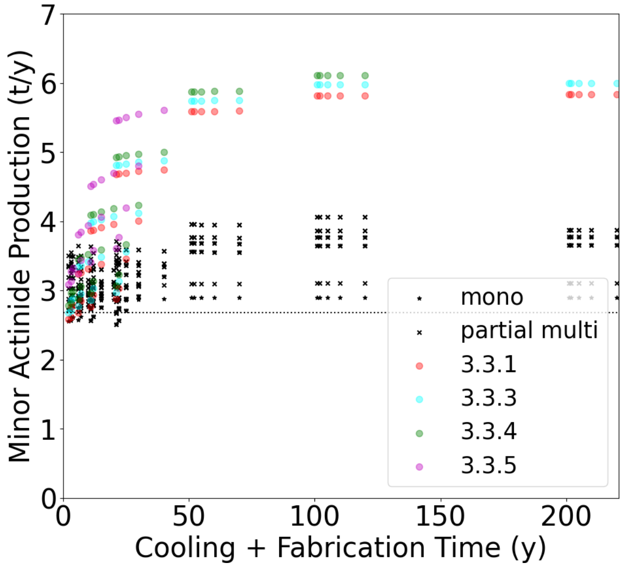


Figure 3.18: Variation of MA production with fabrication + cooling time for full multi-recycling cycles. The black dotted line indicates the once-through production.

Chapter 4: Conclusion

First, the SEPAR code has allowed to perform a systematic study of the effect of cooling and fabrication times on mono-, partial multi-, and full multi-recycling nuclear fuel cycle options in PWRs. For non-advanced mono-recycling cases, the SEPAR results have been found to be in line with results from dynamic calculations, for advanced scenarios, the code still needs to be validated.

A method to calculate the Pu and MA production has been devised, which has shown that it is impossible to attribute an exact value for the Pu and MA production to a given fuel cycle option, but rather a range of values, which are dependent on time, should be given.

All cases with recycling of fissile material reduce the natural uranium consumption, nevertheless, these savings always come at the expense of increased minor actinide production. In the end, the decision for a particular fleet type is a trade-off between natural resource savings and waste production, since both cannot be optimized simultaneously.

The use of fuels with high plutonium contents (MOx or MIX) increase the production of Am and Cm in the reactor through neutron capture on the main plutonium isotopes (and decay of ^{241}Pu for ^{241}Am). At the same times, fuels based on reprocessed uranium increase the Np production through neutron capture on higher ^{236}U fraction in the uranium.

Partial multi-recycling scenarios are very close to mono-recycling scenarios in all respects: the fleet compositions are not drastically different, which means that neither natural uranium consumption nor plutonium or minor actinide production change heavily. The real advantage of partial multi-recycling scenarios is mainly to reduce the rate at which the spent fuel inventory increases. Reprocessing of spent MOx fuel at La Hague could also be used to gain knowledge about the performance of current processes with these fuels. The facility could even be used to conduct full-scale testing of processes and concepts that might be used in a future new reprocessing facility. Limited reprocessing of MOx SF could give invaluable information for the research and development efforts going on for new reprocessing facilities and could complement laboratory-scale testing. Scenarios which require new fuel cycle facilities are probably not useful for partial multi-recycling because the high investment does not warrant the limited advantages, but instead, full multi-recycling cycle could directly be implemented with new facilities.

Full multi-recycling cycles are a necessity if the plutonium cycle should be closed. This entails that a new generation of reprocessing and plutonium fuel fabrication plants, with larger capacities, need to be designed, constructed and commissioned. This is a capital-heavy undertaking and, on top of that, requires a lot of R&D spending to develop. Since fast reactor projects are postponed to (at least) the second half of the century and current fuel cycle facilities are ageing, it seems that, if France wants to continue using nuclear power generation with reprocessing, there is no way around a new generation of fuel cycle facilities.

The simulation of full multi-recycling cycles underlines what was already found in mono-recycling and partial multi-recycling cases, although to a lesser extent: if the plutonium cycle should be closed while minimizing the minor actinide production, short cooling and fabrication times are of the essence. This puts a stress on the requirements of transport, reprocessing and fuel fabrication systems, and a deeper analysis must be performed to allow for an optimized techno-economic operating mode of these facilities.

How short cooling and fabrication times can be is limited by the restrictions on the reprocessing and fuel fabrication facilities as well as on transport of spent fuel. Due to heat loads and radiological protection issues, these steps need a certain cooling time before the fuel can be handled, especially in the case of MOx fuel. Nevertheless, the results show, that minimizing the cooling and fabrication times could have a large beneficial effect on minor actinide and plutonium production, if technologically feasible and economically sound solutions can be found.

At the same time as cooling and fabrication times, the achieved burnup for the different fuels is also of great importance: using the plutonium from lower-burnup UOx fuel can reduce the MIX fraction in the fleet while keeping the plutonium cycle closed, which could be interesting since less plutonium-based fuel means lower cycle costs.

The simulations also show that ERU reactors are an important component of advanced cycles if MIX fuels are involved because of the high residual enrichment in the MIX fuels. The recovery of this uranium is key to reducing the natural uranium consumption while closing the plutonium cycle.

Equilibrium scenarios have shown to be a useful tool to investigate advanced fuel cycle options. They allow to make projections about the R&D challenges that will arise if these advanced cycles are chosen. Most notably, the prediction of the evolution of the isotopic composition of the material will allow to set requirements for future transport, reprocessing and fuel fabrication facilities.

In future work, as a first step, post-processing methods, developed in python by the author, need to be included directly into the SEPAR code to further the code's capacities. In further steps, the simulation results for the advanced scenarios should be validated using a validated dynamic code. More sensitivity studies are also needed around the scenarios modelled here, especially in terms of fuel burnup, to estimate the effect of a historic stock of SF. From there, more complicated cycle options, including fast reactors, other advanced reactors or transmutation systems can be modelled with SEPAR. For a more detailed view of isotope production in the fleets, the SEPAR code can also be developed to include more isotopes such as other minor actinides or fission products.

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Appendix A: Including Decay Chains Into the SEPAR Master Equation

A.1 Introduction

Until now, the SEPAR code is able to handle the decay and accumulation of species with one parent isotope. The main importance for this is the accurate modelling of the decay of ^{241}Pu to ^{241}Am which is invaluable to get accurate predictions. Nevertheless, SEPAR currently neglects decay over decay chains that have more than one generation in between. The 'config.xml' file currently contains two such decay chains that have two generations of decay: $^{241}Pu \xrightarrow{14y} {}^{241}Am \xrightarrow{432.2y} {}^{237}Np$ and $^{242}Cm \xrightarrow{14y} {}^{238}Pu \xrightarrow{88y} {}^{234}U$.

The solution for the evolution of members of a decay chain of arbitrary length has been worked out by Bateman and can be included to model the decay chains accurately in SEPAR. Here, a convenient form of the solution for each isotope in the decay chain is presented. No proof is given but is available in [32].

A.1.1 Problem Statement

Since the decay in SEPAR is modelled for a system outside the reactor, the Bateman equations only contain natural decay terms (no source due to fission or capture is present). In this case, the equations take the form of a Genealogically Ordered Exit-Only Decay (GOED) Chain. This means that for each isotope in the chain, the sink for this isotope is its natural decay and the only source is the natural decay of a precursor. In addition, the chain is ordered, meaning that a parent isotope decays to a daughter nucleus, but the daughter nucleus cannot decay to the parent isotope through some chain. The formalism takes into account branching ratios. Mathematically, the decay chain of length m is governed by the following differential equations:

$$\frac{dN_1(t)}{dt} = -\lambda_1 N_1 \tag{A.1}$$

$$\frac{dN_i(t)}{dt} = -\lambda_i N_i + \sum_{j=1}^{i-1} \rho_{j,i}\lambda_j N_j \quad i \in \{2, ..., m\} \tag{A.2}$$

Here, $\rho_{j,i}$ is the branching fraction for the decay of nucleus j to i, the λ are the decay constants of the respective nuclei and the N denote the quantity of the respective nuclei present at time t. The nucleus with index 1 has no precursor and is called the ancestor. In the case of SEPAR, N is generally a mass. Notice here that the sum in Eq. A.2 only goes to $i-1$ since we assumed that only nuclei higher up in the chain can decay to nucleus j and a nucleus cannot decay into itself, so $\rho_{j,i} = 0$ for $j \geq i$.

A.1.2 General Solutions to the Bateman Equations

It has been shown that the solution to this problem can be conveniently written as a linear combination of decaying exponentials:

$$N_i(t) = \sum_{j=1}^{i} C_{i,j} e^{-\lambda_j t} \quad for\, i \in \{1, ..., m\} \tag{A.3}$$

The coefficients $C_{i,j}$ are defined recursively:

$$C_{i,j} = \frac{1}{\lambda_i - \lambda_j} \sum_{k=j}^{i-1} \rho_{k,i} \lambda_k C_{k,j} \quad for\, i \in \{2, ..., m\}\, and\, j \neq i \tag{A.4}$$

$$C_{i,i} = N_i^0 - \sum_{k=1}^{i-1} C_{i,k} \quad for\, i \in \{2, ..., m\} \tag{A.5}$$

$$C_{1,1} = N_1^0 \tag{A.6}$$

$$C_{i,j} = 0 \quad if\, j \geq i \tag{A.7}$$

Here, N_i^0 denotes the quantity of nucleus i at time $t = 0$, i.e. the initial conditions. The matrix C containing the coefficients $C_{i,j}$ can easily be computed in a recursive algorithm and the evolution of each nucleus in the decay chain can thus easily be computed.

A.2 Implementation in SEPAR

To allow SEPAR to deal with longer decay chains, the master equation needs to be changed. The current form is equivalent to what would be obtained with the described method if each isotope did not have a "grand-parent" isotope. To adjust for longer decay chains, the expression for $N(t; \overrightarrow{N^0})$ can simply be used. Here, the dependence on the initial quantities of the isotopes involved in the decay chain, $\overrightarrow{N^0}$, is explicitly noted since the master equation needs to take into account the decay during the transfer from the reactor to the reprocessing facilities and from the reprocessing facility back to the reactor, and the initial quantities for both steps are obviously changed by decay and removal due to reprocessing. Taking all this into account, SEPAR can be easily adjusted to incorporate decay chains of arbitrary length.

Appendix B: Additional Cycle Option

The results for this case are moved to the appendix because of a lack of space due to the page limitation. This specific case was chosen since it is rather an academic exercise than an interesting case for implementation in reality.

B.1 UOx-ERU fleet with uranium recycling from ERU SF

This fuel cycle option is included for completeness, however, its real world usefulness is arguable at best. If we assume that reprocessed uranium from spent ERU is reused, it would only make sense to reuse the plutonium from ERU SF as well since it would be readily available as a product from the PUREX process. A schematic representation of this cycle option can be found in Fig. B.1. With the criteria set above, this cycle option is not feasible with current facilities.

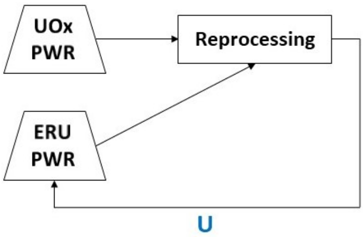

Figure B.1: Schematic representation of the UOx-ERU cycle with uranium recycling from ERU SF.

The ranges for the main fleet parameters at equilibrium can be found in Table B.1. **Due to higher ^{234}U and ^{236}U contents in the reprocessed uranium, the needed enrichment is higher than in the mono-recycling case**. The effect of long cooling times, during which ^{238}Pu can decay to ^{234}U, is more notable here than in the mono-recycling case (section 3.1.3). The ERU batch fraction can be increased from the mono-recycling case and the equilibrium conditions for the fleet in the reference case can be found in Table 3.17.

Table B.1: Ranges for the main fleet parameters obtained for the UOx-ERU fleet with uranium recycling from ERU SF for the different cooling/fabrication times.

ERU batch fraction (%) (ERU fuel fabrication (t/y))	**ERU enrichment** (%)	**natU consumption** (t/y)
11.7 - 12.0 (110 - 112)	5.73 - 6.04	7062 - 7080

Appendix C: Sensitivity Studies

This appendix explores a number of sensitivity studies on the UOx burnup that have been performed around one chosen cycle option from each of the following groups: mono-recycling, partial multi-recycling and full multi-recycling scenarios. For mono-recycling the UOx-MOx-ERU fleet was chosen, for partial multi-recycling, the UOx-MOx-ERU fleet with partial recycling of Pu from MOx SF was chosen, and for full multi-recycling, the UOx-MIX-ERU fleet with plutonium and uranium recycling from MIX and ERU was chosen. In addition to this, a sensitivity study was performed on the factor of plutonium losses at the reprocessing step for the UOx-MIX-ERU fleet with plutonium and uranium recycling from MIX and ERU.

C.1 Sensitivity on the UOx Burnup

For this sensitivity study, the UOx burnup was varied between 50, 55 and 60 GWd/t, while other batches retained their burnup of 55 GWd/t as before. Historically, the burnup of fuel assemblies has steadily increased over the years from 33 GWd/t to 52 GWd/t [44], as fuel manufacturing and modelling has advanced and allowed to safely operate the fuel assemblies to higher burnup. The interest of higher burnup fuels is clearly on the generating side, since higher burnup fuels allow longer refuelling cycles, which translates into fewer outages and more revenue. A burnup of 50 GWd/t can be seen as a representative value of what has been reached in France today, while the a burnup of 60 GWd/t is similar to what EDF expects to be able to achieve with the EPR [44].

Since the general evolution with cooling/fabrication times for a given fleet is the same with a burnup of 50, 55 or 60 GWd/t, the comparison is done for the reference case of 10 years cooling and 2 years fabrication time.

C.1.1 UOx-MOx-ERU mono-recycling fleet

Table C.1 shows that increasing the UOx burnup decreases the MOx and ERU batch fractions. For MOx this is linked to the degraded Pu grade with increasing burnup, whereas for ERU, this is linked to the degraded isotopic composition of U, with more ^{236}U present in the UOx spent fuel. The lower Pu grade and degraded U composition make higher Pu content in MOx, and higher enrichment for ERU, necessary. Since the MOx and ERU batch fractions are decreased, the UOx batch fraction increases, and the natU consumption also increases along with it.

Table C.1: Fleet equilibrium parameters and natU consumption for the UOx-MOx-ERU fleet with different UOx burnups.

UOx burnup	**Batch fraction** (%)			**MOx Pu content** (%)		**Pu grade**	**ERU enrichment**	**natU consumption**
(GWd/t)	UOx	MOx	ERU	Fresh	Spent	(%)	(%)	(t/y)
50	81.1	9.1	9.8	10.9	8.2	61.3	5.19	6520
55	82.2	8.6	9.2	11.2	8.5	60.5	5.24	6596
60	83.2	8.1	8.8	11.5	8.7	59.8	5.28	6686

The MA production (Table C.2) decreases with increased burnup since the fuel types with the highest MA production (MOx and ERU) see their fractions decreased. Although the overall MA production is decreased with higher burnup, the Cm fraction in that production is increased since the Pu in MOx has a lower grade, which also increases the ^{243}Am fraction in production, which is shown by the decreasing ^{241}Am fraction. As before, the overall Am production decreases with decreasing MOx fraction, i.e. with increasing burnup.

The most important factor on the Pu production is the Pu content in MOx and the MOx batch fraction since the MOx fuels contain by far the largest part of Pu (much larger than the Pu accumulated in ERU SF). Although the Pu contents in fresh and spent MOx increase with increasing burnup, the fraction of Pu that is burnt (or transmuted), i.e. the difference between the fresh and spent content, is about the same in all cases since the MOx burnup is fixed. The Pu production is therefore reduced with reduced MOx fraction, i.e. with increased UOx burnup.

Table C.2: Pu and MA production for the UOx-MOx-ERU fleet with varying UOx burnups.

UOx burnup (GWd/t)	**Pu** (t/y)	**Total MA** (t/y)	**Np** (%)	**Am (241)** (%)	**Cm** (%)
50	6.69 - 8.12	2.10 - 3.15	52.0	32.6 (9.6)	15.4
55	6.44 - 7.83	2.12 - 3.11	52.2	32.2 (9.4)	15.6
60	6.23 - 7.60	2.13 - 3.07	52.4	31.8 (9.1)	15.8

C.1.2 UOx-MOx-ERU fleet with partial MOx SF reprocessing

Compared to the previous fleet, the UOx-MOx-ERU fleet with partial reprocessing of MOx SF shows an increased MOx fraction C.3 because more Pu is available to fuel the MOx reactors. With increasing burnup, the absolute mass, and the fraction of the yearly flux, of MOx fuel that can be reprocessed decreases. This is because the reprocessed fraction is limited by the degradation of Pu quality in UOx SF that accompanies the increased burnup. The Pu grade in fresh MOx fuel reaches a value of about 58.7 % for all of the cases, which is the grade required to reach 55 GWd/t at a Pu content of 12% in fresh MOx. As before, increasing the UOx burnup increases the UOx fraction and therefore the natU consumption.

Table C.3: Fleet equilibrium conditions for the UOx-MOx-ERU fleet with partial MOx SF reprocessing for different UOx burnups.

UOx burnup	**Batch fraction** (%)			**MOx SF reprocessing**	**Pu grade**	**ERU enrichment**	**natU consumption**
(GWd/t)	UOx	MOx	ERU	(t/y)	(%)	(%)	(t/y)
50	80.2	10.2	9.7	26 (27%)	58.7	5.19	6446
55	81.5	9.3	9.2	18 (21%)	58.6	5.24	6542
60	82.8	8.5	8.7	11 (14%)	58.6	5.28	6653

For the same reasons as in the previous fleet, the Pu and MA productions are reduced when the UOx burnup is increased C.4. This time, however, since the Pu grade and contents are the same for all cases, the variations in the Cm and Am fractions are not very large, and only governed by the change in batch fractions, and not Pu content or grade.

Table C.4: Pu and MA production for the UOx-MOx-ERU fleet with partial MOx SF reprocessing for different UOx burnups.

UOx burnup (GWd/t)	**Pu** (t/y)	**Total MA** (t/y)	**Np** (%)	**Am (241)** (%)	**Cm** (%)
50	6.16 - 6.94	2.81 - 3.45	48.8	35.1 (10.9)	16.1
55	6.07 - 6.85	2.73 - 3.32	49.9	34.0 (10.3)	16.1
60	6.00 - 6.79	2.66 - 3.19	51.0	32.9 (9.7)	16.0

C.1.3 UOx-MIX-ERU fleet with U and Pu reprocessing from MIX and ERU SF

For the full multi-recycling case, the first thing to notice is the decrease of the Pu grade with increasing UOx burnup (Table C.5). As before, since the Pu and U isotopic compositions are degraded with increased UOx burnup, the enrichments in fresh MIX and ERU fuels need to be increased with increased UOx burnup. The lower Pu grade also decreases the MIX fraction with increased burnup. Large quantities of U are recycled, yielding a high ERU batch fraction, but also increasing the ERU enrichment significantly over the mono- or partial multi-recycling cases. The natU consumption increases along with increased burnup due to higher UOx fraction and generally higher enrichment in UOx, MIX and ERU fuels.

Table C.5: Equilibrium conditions for the UOx-MIX-ERU fleet with U and Pu reprocessing from MIX and ERU SF for different UOx burnups.

UOx burnup (GWd/t)	**Batch fraction** (%) UOx	MIX	ERU	**MIX enrichment** (%)	**Pu grade** (%)	**ERU enrichment** (%)	**natU consumption** (t/y)
50	39.4	38.6	22.0	4.22	45.8	5.58	5781
55	40.5	37.9	21.6	4.25	45.5	5.61	5834
60	41.4	37.3	21.3	4.28	45.3	5.64	5893

The MA production of the fleet is reduced when UOx burnup is increased because the batch fraction of MIX and ERU reactors, which produce more MA than UOx reactors, is decreased. Since the Pu prodcution for this fleet with a closed Pu cycle is only due to Cm in the waste, and since the quantities of Cm do not vary dramatically, we can say that the Pu production is not really affected by the UOx burnup (Table C.6). The Am fraction decreases slightly with decreasing MIX fraction, which was already observed when MOx reactors were involved.

Table C.6: Pu and MA production for the UOx-MIX-ERU fleet with U and Pu reprocessing from MIX and ERU SF for different UOx burnups.

UOx burnup (GWd/t)	**Pu** (t/y)	**Total MA** (t/y)	**Np** (%)	**Am (241)** (%)	**Cm** (%)
50	0.08 - 0.51	4.57 - 4.82	33.8	43.0 (9.9)	23.2
55	0.08 - 0.51	4.54 - 4.79	34.2	42.7 (9.7)	23.1
60	0.08 - 0.51	4.50 - 4.75	34.6	42.4 (9.6)	23.0

C.2 Sensitivity on Pu losses at the reprocessing step for the UOx-MIX-ERU fleet with U and Pu reprocessing from MIX and ERU SF

The sensitivity on Pu losses at the reprocessing step is particularly interesting for multi-recycling fleets because if the losses of Pu were too large, the Pu cycle would not really be closed since significant quantities of Pu could end up in the waste. The loss of plutonium would reduce the fraction of reactors fuelled with plutonium-based fuels and could therefore have an important effect on the equilibrium conditions. The PUREX process exhibits high recovery and purification yields (99.9% and 10^6 respectively [45]), such that only very small quantities of Pu and U are lost to the waste stream and the obtained Pu and U streams are very pure.

For the next generation of extraction processes, one promising type of extraction molecule are monoamides or diamides, which can achieve improved performances over the PUREX process, most notably to the resistance of the process to radiolysis. In addition, the processes developed based on these molecules are able to separate the U and Pu streams without the need for a reducing agent, like used in the PUREX process, which would reduce the liquid waste significantly [46]. With these characteristics, advanced extraction processes will be suited to reprocess fuels with high Pu content and higher self-irradiation, which is necessary for full multi-recycling cycles. It is therefore important to know if the reprocessing losses have an important impact on the cycle, and what limits need to be imposed on future processes.

The UOx-MIX-ERU fleet with U and Pu reprocessing from MIX and ERU SF was modelled for plutonium losses at reprocessing of 0% and 0.1%, as well as for extreme values of 1.0% and 5.0%. It was assumed that there are no losses of U and all reactors were modelled for burnups of 55 GWd/t.

Table C.7 shows that the MIX fraction decreases with increased Pu losses since the fraction is determined by the mass of Pu available in the fleet. Losing Pu during reprocessing increases the Pu grade available to feed MIX production because the fraction of Pu from UOx or ERU SF, with higher grade, becomes higher. This also reduced the needed enrichment in the MIX fuel. To the contrary, the ERU enrichment is very slightly increased if the Pu losses are higher because the fraction of U from MIX, with higher residual enrichment than U from UOx, is decreased. All in all, the natU consumption is increased slightly with increasing Pu losses due to higher UOx batch fractions, and this increased need for enriched uranium cannot be compensated by the lower MIX enrichment.

Table C.7: Equilibrium conditions for the UOx-MIX-ERU fleet with U and Pu reprocessing from MIX and ERU SF for different losses of Pu during reprocessing.

Pu losses (%)	**Batch fraction** (%) UOx	MIX	ERU	**MIX enrichment** (%)	**Pu grade** (%)	**ERU enrichment** (%)	**natU consumption** (t/y)
0	40.5	37.9	21.6	4.35	45.8	5.61	5834
0.1	40.6	37.8	21.5	4.24	45.6	5.61	5836
1.0	42.1	36.8	21.1	4.19	45.9	5.64	5852
5.0	47.7	32.9	19.4	3.99	47.3	5.62	5923

The MA production (Table C.8) is decreased with more Pu losses because the fraction of UOx, with lower MA production, is increased in this case. The Pu production is now the sum of the Pu lost during reprocessing and the production due to the decay of Cm, which is more or less constant, meaning that the overall Pu production increases with the Pu losses.

Table C.8: Pu and MA production for the UOx-MIX-ERU fleet with U and Pu reprocessing from MIX and ERU SF for different losses of Pu during reprocessing.

Pu losses (%)	**Pu** (t/y)	**Total MA** (t/y)	**Np** (%)	**Am (241)** (%)	**Cm** (%)
0	0.08 - 0.51	4.54 - 4.79	34.2	42.7 (9.7)	23.1
0.1	0.11 - 0.53	4.53 - 4.78	43.3	42.7 (9.7)	23.1
1.0	0.35 - 0.74	4.48 - 4.69	34.8	42.4 (9.7)	22.9
5.0	1.35 - 1.57	4.27 - 4.37	36.9	41.0 (9.8)	22.0