FACULTY OF TECHNOLOGY

OVERVIEW OF RECYCLED NUCLEAR FUEL FOR ENERGY PRODUCTION

Sanni Kalaoja

ENVIRONMENTAL ENGINEERING
Bachelor's thesis
March 2024

ABSTRACT

Overview of recycled nuclear fuel for energy production

Sanni Kalaoja

University of Oulu, Programme of Environmental Engineering

Bachelor's thesis, pp. 32

Supervisor at the university: Antonio Caló

The purpose of this thesis is to constitute an understanding of the development, the current state and the future of spent nuclear fuel recycling. This thesis explores the possibility of closing the nuclear fuel cycle by recycling uranium and plutonium in fast breeder reactors. The development of already existing technologies for uranium and plutonium recycling are presented along with the environmental and economic impacts of spent nuclear fuel recycling. The standard open fuel cycle without any degree of fuel recycling is compared to a partially closed fuel cycle and the prospective fully closed fuel cycle. This thesis is conducted as a literature review and serves as an overview of the wider topic of nuclear fuel recycling.

Nuclear energy is relatively environmentally friendly as nuclear power plants do not produce greenhouse gases during operation. The environmentally challenging parts of nuclear energy production lie significantly in uranium mining and fuel production as well as in nuclear waste management and storage. This thesis examines how nuclear fuel recycling affects those aspects and what kind of advantages and disadvantages it brings. Technical challenges of recycling are examined, for example, the behavior of fuel made of recycled materials in reactors is compared to typical uranium fuel.

This thesis found that there are well established technologies for spent nuclear fuel recycling currently in use. Advantages of spent nuclear fuel recycling include the reduced need for uranium mining and reduction of high-level waste volumes and radiotoxicity of the waste. Disadvantages of recycling are largely economic as the production of uranium fuel is cheaper than recycling. Closing the nuclear fuel cycle is possible with fast reactors but it is not yet deployable commercially. Fast reactors are under development, but they

are not expected to become commercially available to produce power until the latter half of this century.

Keywords: nuclear reprocessing, spent nuclear fuel, mixed oxide fuel (MOX), plutonium uranium reduction extraction (PUREX), fast reactors

TIIVISTELMÄ

Yleiskatsaus käytetyn ydinpolttoaineen kierrätyksestä energian tuotantoa varten

Sanni Kalaoja

Oulun yliopisto, Ympäristötekniikan tutkinto-ohjelma

Kandidaatintyö 2024, 32 s.

Työn ohjaaja yliopistolla: Antonio Caló

Tämän kandidaatintyön tavoitteena on muodostaa kattava käsitys ydinpolttoaineen kierrätyksen kehityksestä, nykytilasta sekä tulevaisuudesta. Työn tarkoituksena on selvittää mahdollisuutta sulkea ydinpolttoainekierto kierrättämällä uraania ja plutoniumia nopeissa hyötöreaktoreissa. Työssä esitellään olemassa olevia käytetyn ydinpolttoaineen kierrätysmenetelmiä ja kierrätyksen ympäristöllisiä sekä taloudellisia vaikutuksia. Työssä verrataan tavanomaista avointa uraani-ydinpolttoainekiertoa, osittain suljettuun polttoainekiertoon sekä tulevaisuudessa mahdolliseen täysin suljettuun ydinpolttoainekiertoon. Työ toteutetaan kirjallisuuskatsauksena ja se toimii yleiskuvauksena ydinpolttoaineen kierrätyksestä.

Ydinenergia on verrattain ympäristöystävällistä, sillä ydinvoimalat eivät tuota toiminnassaan kasvihuonekaasupäästöjä. Ympäristönäkökulmasta ydinenergian tuotannossa haastavimpia vaiheita ovat erityisesti kaivostoiminta uraanin louhinnassa, polttoaineen tuotanto sekä ydinjätteen käsittely ja varastointi. Tässä työssä tutkitaan, miten käytetyn ydinpolttoaineen kierrätys vaikuttaa edellä mainittuihin polttoainekierron vaiheisiin sekä muita kierrätyksen etuja ja haittoja. Työssä kerrotaan kierrätyksen teknisistä haasteista, kuten kierrätetyistä materiaaleista tehdyn polttoaineen eriävästä toiminnasta reaktoreissa.

Työssä todetaan, että käytetyn ydinpolttoaineen kierrätykseen on käytössä hyvin vakiintuneita teknologioita. Kierrätyksen etuihin lukeutuu uraanin louhinnan tarpeen väheneminen sekä korkea-aktiivisen jätteen radiotoksisuuden ja tilavuuden pieneneminen. Kierrätyksen haittapuoliin kuuluu sen kalleus. Kierrätys on laajasti taloudellisesti kannattamatonta, sillä ydinpolttoaineen tuottaminen louhitusta uraanista on halpaa. Ydinpolttoainekierron sulkeminen nopeiden reaktoreiden avulla on mahdollista, mutta sitä ei olla vielä tehty. Nopeat reaktorit ovat vasta kehityksen alla ja

niiden odotetaan yleistyvän kaupallisessa voimantuotannossa vasta tämän vuosisadan puolivälin jälkeen.

Asiasanat: ydinjätteen uudelleenkäsittely, käytetty ydinpolttoaine, sekaoksidipolttoaine (MOX), PUREX-prosessi, nopeat hyötöreaktorit

TABLE OF CONTENTS

SYMBOLS AND ABBREVIATIONS

BWR	boiling-water reactor
DGR	deep geological repository
FBR	fast breeder reactor
FR	fast reactor
GIF	Generation IV International Forum
HLW	high level waste
HNO_3	nitric acid
LWR	light water reactor
MOX	mixed oxide
Pu	plutonium
PuO_2	plutonium dioxide
PUREX	plutonium uranium reduction extraction
PWR	pressurized-water reactor
REDOX	reduction oxidation
RepU	reprocessed uranium
SNF	spent nuclear fuel
TBP	tributyl phosphate
U	uranium
U-235	uranium 235
U-238	uranium 238
UO_2	uranium dioxide
wt%	weight percent

1 INTRODUCTION

This thesis explores nuclear fuel recycling and the prospect of closing the uranium based nuclear fuel cycle by reusing spent nuclear fuel (SNF) from light water reactors (LWRs) in fast breeder reactors (FBRs). The current state of commercial SNF recycling is examined in the thesis along with the prospective FBRs to fully close the nuclear fuel cycle. This thesis' purpose is to obtain a basic understanding of the technical challenges and advantages of SNF recycling. The environmental and economic impacts of recycling SNF are compared to the once-through nuclear fuel cycle. The thesis is conducted as a literature review.

The thesis deals mostly with the back end of the nuclear fuel cycles. The traditional once-through fuel cycle is discussed briefly along with the currently deployed partially closed fuel cycle and the prospective fully closed fuel cycle. The advantages of recycling SNF include the decreased demand for mining uranium and minimization of waste. It does not eliminate the need for final disposal sites, but it affects the amount and the nature of the waste requiring final disposal. Challenges discussed in this thesis include the economics of the recycling.

FBRs are discussed in this thesis as they are thought to be the next major step in making the nuclear fuel cycle more circular. FBRs have the potential to create more plutonium than they consume. FBRs can use fuel made with already existing technology utilizing the stocks of plutonium created in LWRs and stockpiles of depleted uranium created in the uranium fuel fabrication process.

As the world is switching to greener economies and technologies, nuclear energy plays an important part in it. Resources should be used more mindfully and thoroughly to produce as little waste and pollution as possible. Nuclear power is a relatively environmentally friendly energy source, especially compared to fossil fuels as nuclear power plants do not produce greenhouse gas emissions. Nuclear power is reliable and safe thanks to strict regulations. (World Nuclear Association 2023)

Uranium is a common element in the Earth's crust and seawater. Technologies to utilize all of its potential as a fuel are under development. Most of the pollution caused by nuclear energy production comes from the mining and enrichment process. Enriching uranium produces massive amounts of depleted uranium that could still be processed into more nuclear fuel. (World Nuclear Association 2021a)

2 NUCLEAR FUEL CYCLE AND SPENT NUCLEAR FUEL

Currently, there are around 410 nuclear power reactors in operation worldwide. (IAEA 2024a) Nuclear energy became commercially available around the 1960s. Ever since then and until 2016 nuclear power reactors have generated about 390 000 tonnes of spent uranium fuel. Only one third of the SNF has been reprocessed and the rest remains in storage. (IAEA 2022) p. 11 The most widely used nuclear reactors are light water reactors (LWRs). The two common types of LWRs are the pressurized-water reactor (PWR) and the boiling-water reactor (BWR). LWRs use normal water as coolant and neutron moderator. Because of the neutron moderator, neutrons slow down and work at thermal energies. LWRs are thermal reactors because of this. LWRs typically use fuel assemblies made of uranium oxide with zircalloy cladding. The typical PWRs and BWRs require slightly different uranium fuels of composition. (Spinrad and Marcum 2024) A typical nuclear fuel composition can be seen in Figure 1.

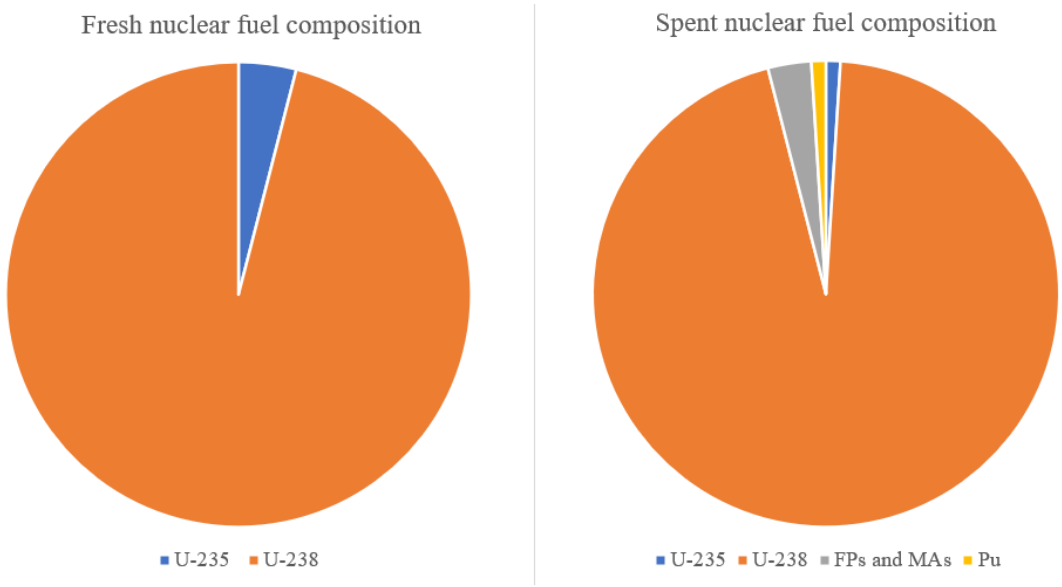

Figure 1: Nuclear fuel and spent nuclear fuel compositions (World Nuclear Association 2021a)

A nuclear reactor typically requires fuel that contains between 3,5% and 5% of U-235, which is a fissile isotope in natural uranium. The rest of the uranium in the fuel is U-238, as is depicted in Figure 1. U-238 is a fertile isotope of uranium. Depending on the reactor type and operation style, nuclear fuel spends 18-36 months in the reactor, until using it is no longer viable due to buildup of fission products. Used fuel contains about 95% U-238,

3% fission products and minor actinides, 1% U-235 and 1% Pu (of which about 60% is fissile Pu). (World Nuclear Association 2021a) The fissile isotope of plutonium, Pu-239, is created in nuclear reactors through neutron capture of the fertile U-238. Pu-239 is the most useful of the plutonium isotopes, because it contributes to the energy output of the reactor by fission. In a typical reactor more Pu-239 is created than destroyed which results in its presence in SNF. (Bodansky 2004) p. 208

When the SNF is taken out of the reactor, it is highly radioactive mostly due to the fission products. The SNF still produces heat and needs to be cooled down and shielded from emitting radiation to the environment. SNF assemblies are typically placed in pools of water at the reactor site to manage the decay heat and reduce the amount of radiation to safer levels. This can take months to years after which there are two options. The SNF can be treated as a waste and put into a final disposal, or it can be reprocessed. The goal of reprocessing is to separate the remaining fissile and fertile materials, mainly uranium and plutonium, from the fission products. (World Nuclear Association 2021a) The standard steps of the nuclear fuel cycle are depicted in Figure 2.

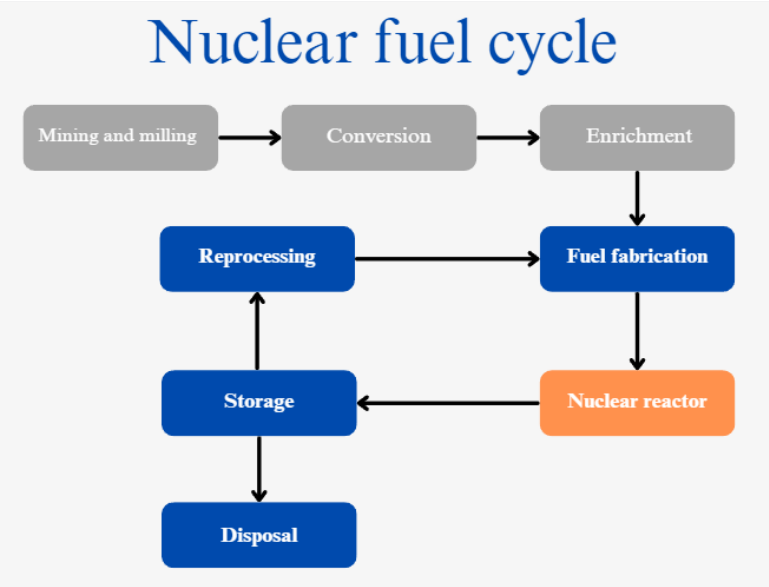

Figure 2: Nuclear fuel cycle (World Nuclear Association 2021a)

The front end of the nuclear fuel cycle is not within the scope of the thesis and is marked gray in Figure 2. This thesis focuses on the back end of the fuel cycle, which is marked blue. Fuel fabrication is generally deemed to be part of the front end of the cycle, but since reprocessed materials have to be fabricated into fuel, it is partially within the scope

of this thesis, and thus also marked blue. Reprocessed fuel behavior in reactors, marked orange, is also examined in this thesis.

If SNF is not reprocessed, it is considered to be high level waste (HLW). Only less than 1% of all radioactive waste is HLW but it contains about 95% of the total radioactivity in all radioactive waste. After the initial cool down period, the SNF is moved to an interim storage where it will further cool down for several decades. Then the SNF will be sealed in corrosion and leach resistant containers and finally be disposed of in deep geological repositories (DGRs). (IAEA 2022) p. 27

In the future, fast neutron reactors, or fast reactors (FRs), are expected to become the industry standard. They have the potential to use uranium resources far more efficiently and be a crucial part of the closed nuclear fuel cycle. Most FRs built so far have been fast breeder reactors (FBRs). (World Nuclear Association 2021b) Fast reactors do not use a neutron moderator in order to produce fission at higher energies, and fast neutrons have higher kinetic energy than thermal neutrons used in LWRs. Fast reactors can be breeders, converters or burners depending on the fertile to fissile nuclei conversion ratio and fissile nuclei burning ratio. Fast reactors utilize U-238 as fertile fuel to produce and burn Pu-239. Fast breeders produce more Pu-239 than they consume, converters roughly produce as much Pu-239 as they consume, and burners consume or burn more Pu-239 than they produce. FBRs can use Pu-239 as fissile fuel more easily along with other not so readily burnable isotopes in LWRs. Some of the Generation IV International Forum's (GIF's) reactors under research are FRs and could be operated as FBRs. (Bodansky 2004) p.186

3 CURRENT STATE OF RECYCLING SPENT NUCLEAR FUEL

Interest in recycling SNF arises from the need to safely manage the produced plutonium as well as the aspects of waste minimization and reduction of the radiotoxicity of the waste. Recycling can enhance the environmental friendliness and sustainability of nuclear power. (IAEA 2003)

As of 2022, only three countries, France, Russia and India, operate large scale reprocessing facilities for SNF. Most countries are planning to place their SNF as waste in DGRs, including Finland. Some countries, such as Bulgaria and the Netherlands, have their SNF reprocessed in another country, such as France, that operate the reprocessing facilities. Management of SNF as well as transporting and reprocessing it are under strict regulations. (IAEA 2022) p. 27 It is to be noted that regulations and licenses are crucial to safely manage all processes associated with radioactive materials, but they will not be discussed deeply in this thesis.

Reprocessing SNF works in the same way as processing metal minerals. There are three broad types of metallurgical processes that include pyrometallurgy, electrometallurgy and hydrometallurgy. They can be applied to reprocessing SNF, but the most common way is a hydrometallurgical process called the PUREX process. The PUREX acronym comes from plutonium uranium reduction extraction. After reprocessing SNF with the PUREX process, the extracted plutonium can be made into mixed oxide (MOX) fuel. (World Nuclear Association 2020) Of the processes developed for recycling SNF this thesis will focus on the PUREX separation process and the production of MOX fuel. Their placement on the nuclear fuel cycle can be seen in Figure 3.

The PUREX process is the most widely used separation process for SNF reprocessing. All of the countries with used nuclear fuel reprocessing plants use the PUREX process with some variety in the details. In 2005 there has not been another way to reprocess SNF, at least on an industrial scale. (IAEA 2005) p.10

Fission products, along with minor actinides that are separated in the PUREX process, are treated as HLW. Reprocessing SNF also produces intermediate and lower-level

wastes. The separated uranium from the PUREX process, also referred to as RepU, can go back to the fuel cycle to go through conversion and enrichment again. (IAEA 2022) RepU is not very attractive in the economic sense of recycling because it contains some impurities and unwanted isotopes of uranium. They make RepU more difficult to handle and RepU requires more complicated processes so treat it compared to natural uranium. (Nawada et al. 2007) p. 5

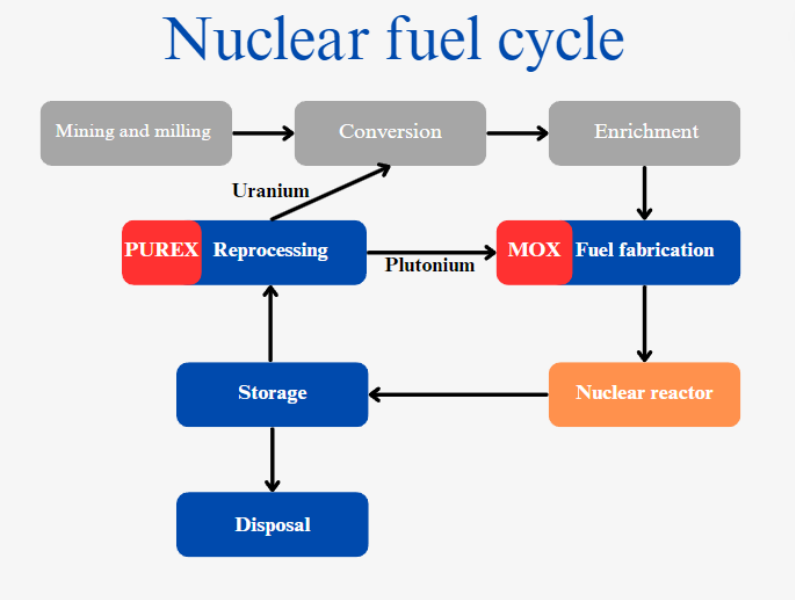

Figure 3: Recycling processes on the nuclear fuel cycle (World Nuclear Association 2021a)

Separated plutonium can be processed into mixed oxide (MOX) fuel that can be used in some nuclear reactors, mainly LWRs. MOX fuel consists of depleted uranium and plutonium oxides. MOX fuel has been used for decades in LWRs around the world. MOX fuel is currently produced in France, and it has been produced in Belgium and the UK. (IAEA 2022) Japan has a MOX fuel production plant planned to start operation in 2024 (International Panel on Fissile Materials 2023). Producing MOX fuel is a well-established way to recycle nuclear fuel. Additionally, there are innovative developments to recycle plutonium and uranium in MOX fuel that are not yet commercially available. (World Nuclear Association 2017)

3.1 The standard PUREX process

The separation of plutonium first became of interest to build atomic weapons in the Cold war. There were many types of batch process experiments based on centrifugation, precipitation and repeated dissolution to separate plutonium, but a continuous process was needed. A continuous reduction oxidation (REDOX) process was created for the purpose, but it proved to be not the ideal solution. The salting agent, aluminum nitrate ($Al(NO_3)_3$), used in the REDOX process, could not be recycled and the used solvent hexone ($C_6H_{12}O$) has a low flashpoint, which makes it susceptible to exploding. (Gerber 1993) Apart from the salting agent and solvent, the REDOX and PUREX processes are quite similar. (Irish and Reas 1957) The first PUREX plant was built in 1955 in the USA, and it began processing radioactive materials in 1956. (Gerber 1993)

The PUREX process is an aqueous separation process to extract plutonium and uranium from irradiated nuclear fuel. PUREX can be used for oxide and metallic fuels. The process uses nitric acid (HNO_3) as a salting agent and tributyl phosphate (TBP) as the organic solvent. PUREX forms two product streams, one includes plutonium and the other uranium. The waste of the PUREX process includes minor actinides (neptunium, americium and curium) and fission products. The waste streams are treated as HLW. (Dunn Lee et al. 2023) p. 101

PUREX process can consist of six major steps that include the extraction of plutonium (Pu) and uranium (U) from aqueous solution into the TBP, partitioning of U and Pu, decontamination and recovery of U and Pu, recovery of the solvent as well as the recovery of the salting agent. A process diagram of the PUREX process is presented in Figure 4. In the PUREX process the solvent is safer as it has a higher flashpoint, making it less likely to explode compared to the solvent used in the REDOX process. Additionally, the salting agent is recyclable, which makes PUREX better than the earlier developed REDOX process. (Irish and Reas 1957)

In the PUREX process, 99.9% of the fission products are left in the aqueous phase in the first cycle. Multiple cycles further purify the products. Most of the impurities and nitric acid get separated together and this aqueous waste stream is sent to acid recovery. The salting agent HNO_3 is recovered to save waste storage space. The waste stream of the acid

recovery process is a high activity acid concentrate which contains the fission products. The waste is neutralized by adding sodium hydroxide. (Irish and Reas 1957)

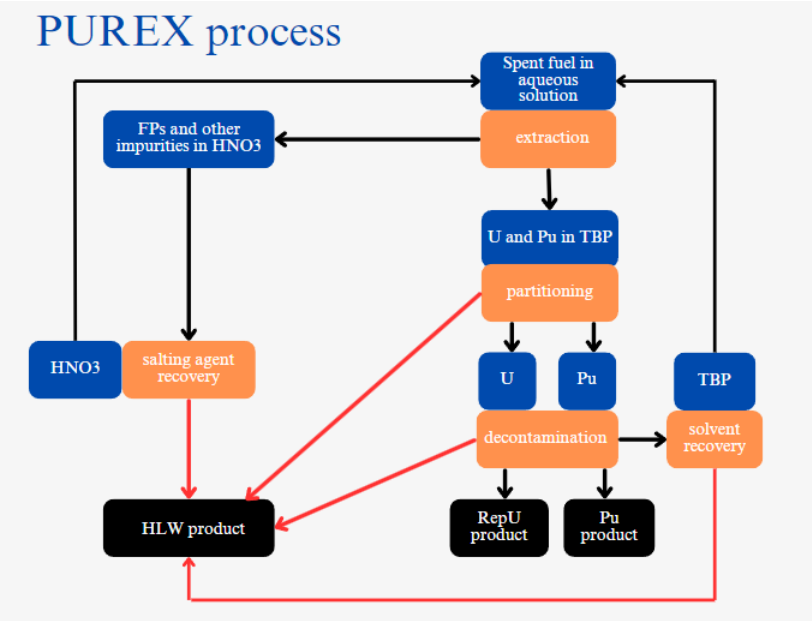

Figure 4: Standard steps of the PUREX process (Irish and Reas 1957)

The other major waste streams in the PUREX process come from the organic solvent recovery process. The solvent which includes uranium and plutonium is decontaminated further to purify the final U and Pu products of remaining fission products. The U and Pu products are purified and concentrated, and the remaining solvent is sent to solvent recovery. The solvent is washed with a sodium carbonate solution and centrifugated to remove solids and remaining aqueous phase. The solids form a waste slurry. The carbonate wash removes the residual radioactive elements in the solvent. They will go to waste storage along with depleted sodium carbonate. (Irish and Reas 1957)

As can be seen in Figure 4, every step in the PUREX process produces waste, marked with red arrows. The wastes from the PUREX process are treated as HWL. They are typically vitrified first in glass or ceramic matrices. The materials have to be durable chemically and be able to withstand radiation and heat. Borosilicate glass for example is deemed suitable for vitrification. After vitrification the waste is packed and eventually disposed of in a DGR. (IAEA 2022)

One prospect of separation processes in SNF recycling is the separation of minor actinides along with uranium and plutonium. Minor actinides include neptunium, americium and

curium. Returning them back to a nuclear reactor would greatly reduce the radioactivity of the remaining waste further. In a reactor, the minor actinides would be converted to other elements through neutron reactions. (Bodansky 2004) p. 214

3.2 MOX fuel fabrication

Producing MOX fuel is the next step in the commercially deployed recycling cycle of spent nuclear fuel. MOX fuel is a mixture of uranium and plutonium oxides, mainly uranium dioxide and plutonium dioxide (UO_2 and PuO_2 respectively). MOX fuel is mainly used in thermal reactors. The purpose of MOX fuel is to use plutonium that forms in a nuclear reactor and reduce the already existing stocks of weapons grade plutonium to reduce the risk of proliferation. Nuclear proliferation in short means that materials suitable for making nuclear weapons end up in the wrong hands. (Bodansky 2004) p. 205 Proliferation is a critical aspect that needs to be addressed when managing fissile materials, as they are managed in SNF recycling, but it is outside the scope of this thesis.

In the PUREX process, plutonium IV, plutonium with the oxidation state +4, forms a stable complex with the nitrate ions from nitric acid and the TBP. (Irish and Reas 1957) Plutonium nitrate can be converted into PuO_2 which is necessary in the production of MOX fuel. There are many different ways to convert plutonium nitrate to PuO_2. They include precipitation, thermal de-nitration, co-precipitation and gel precipitation methods. In commercial scale, precipitation of plutonium oxalate is the most widely used method. The final plutonium product is purer especially compared to de-nitration and handling of the product is easier. (IAEA 2003) p. 13

The precipitation of plutonium oxalate process uses hydrogen peroxide to condition the plutonium nitrate to Pu (IV) before precipitation. Then, an excess of oxalic acid is added to the product to reduce its solubility. Plutonium oxalate decomposes to PuO_2, which can be made into ceramic fuel. The PuO_2 product is filtered, washed, dried and calcined before storage and MOX fuel fabrication. (IAEA 2003)

The conventional MOX fuel fabrication process consists mainly of mixing PuO_2 with UO_2 and possibly scraps generated in the process, blending, granulation, mixing, pelletizing and sintering before pellets are stacked into fuel rods. The fuel rods are welded shut and assembled into fuel assemblies. In the fabrication process, important

characteristics of the ready fuel include an even distribution of plutonium and uniformity of the isotopic composition of plutonium. The isotopic composition depends on what kind of reactor the SNF is from. Typically, recyclable fuel comes from PWRs and the plutonium in it contains 60-70% fissile isotopes. (IAEA 2000) p. 49

The UO_2 in MOX fuel is a carrier material for plutonium. It is possible to use natural, tails or recycled uranium, RepU. Most economic option is to use tails uranium, which contains 0.4-0.7 wt% U-235. Tails uranium comes from the uranium fuel fabrication process, specifically from the enrichment process. Using RepU as the carrier material has proved to be more difficult due to varying isotopic composition of the recycled uranium. (IAEA 2003) p. 51

After MOX fuel has been used in a reactor, it can be reprocessed to recycle the remaining plutonium. In 2012 the prevailing reprocessing technologies practically allowed only two to three plutonium recycles. A lot of research and development is underway to increase the number. The number of recycles is limited due to the build-up of even plutonium isotopes that are non-fissionable. Also, other heavy element isotopes are created, that make the spent MOX fuel more difficult to reprocess. (NEA and OECD 2013) p. 30 Most importantly higher contents of Pu-238, Pu-241, americium and curium make spent MOX fuel more difficult to manage and reprocess compared to used uranium fuels. The difficulties are due to the elements increasing the radiotoxicity of spent MOX fuel. (IAEA 2003) p. 93

Additionally, weapons grade plutonium can be fabricated into MOX fuel. Research and development are being conducted, mainly in Russia and the US, to efficiently reduce the weapons grade plutonium stocks by burning it as MOX fuel in LWRs and FRs. There are some differences and difficulties with making weapons grade plutonium into MOX fuel compared to normal reactor grade plutonium. (IAEA 2003) p. 10 Using weapons grade plutonium to make MOX fuel is one way to reduce the plutonium stocks by recycling. This touches on this thesis's topic, which makes it worth mentioning but not in depth.

4 RECYCLED FUEL IN REACTORS

4.1 MOX fuel in light water reactors

As of 2001, MOX fuel has been used in over 30 thermal reactors around the world. MOX is used mainly in LWRs and both PWRs and BWRs can use it. Countries with experience in using MOX include Belgium, France, Germany, Japan and Switzerland. (IAEA 2003) p. 8 MOX was used in Europe in several reactors but only in France and the Netherlands in 2022. Using MOX fuel affects the reactor's safety requirements and a license to use MOX fuel is needed. MOX fuel also affects the performance of the reactor, and it has to be adapted to use the fuel. (Euratom Supply Agency 2022) p. 8

Most reactors licensed to use MOX are allowed to use only up to one third of the fuel assemblies made of MOX. This is due to the different properties of MOX fuel and what has been found safe in testing. Full reactor cores of MOX fuel are being investigated. MOX fuel behaves differently in the reactor core compared to uranium fuel because different plutonium isotopes have different neutronic properties. MOX made of weapons grade plutonium behaves more similarly to uranium fuel than MOX made of reactor grade plutonium. Due to plutonium's different behavior in a reactor, a concentration of 7.5 wt% Pu is calculated to be equivalent to between 4.0-4.3 wt% U-235. (IAEA 2003) p. 46 A comparison of the equivalent concentrations of uranium and plutonium can be seen in Figure 5.

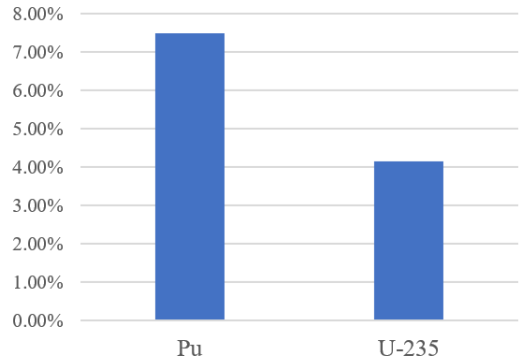

Figure 5: Concentration of plutonium equivalent to U-235 in MOX fuel (IAEA 2003)

Plutonium concentration requirements depend on the quality of the plutonium. The quality is determined by the amount of fissile plutonium isotopes, Pu-239 and Pu-241. Figure 5 shows the concentration of total plutonium content. In reactor grade plutonium, there is usually between 60-70% fissile plutonium. The different neutronic properties of plutonium affect the reactivity of the fuel. (IAEA 2003)

Reactivity of MOX fuel decreases more slowly when the burnup, representative of the time a fuel element spends in the reactor, increases compared to uranium fuel. This means MOX fuel releases more fission gas. While the concentration of plutonium in MOX fuel for LWRs is low, the fuel rods behave quite similarly. At higher burnups the fuel rods gather more pressure due to the higher reactivity and buildup of fission gases. MOX fuel reactivity has to be adjusted along with dissipated power and accumulated burnup to increase compatibility with uranium fuel to avoid harm to reactor operation and safety. Plutonium also affects the thermal conductivity of the fuel. (IAEA 2003)

These variables cannot be made identical in MOX and uranium fuel, but reasonable equivalency can be reached with adequate fuel rod and assembly design. A wide range of tests around the world with different conditions and variables in the reactor core and fuel have been conducted to find that MOX fuel in LWRs is at least as safe and reliable as uranium fuel. (IAEA 2003)

MOX fuel utilization in LWRs is a mature technology, but MOX can be used in other types of reactors too. Another thermal reactor which could use MOX fuel is a pressurized heavy water reactor (PHWR). Plutonium recycling as MOX and other advanced fuels could be utilized in PHWRs with high burnup. (Gupta et al. 2008) In the year 2022 nearly 95% of all operational nuclear power reactors were either LWRs or PHWRs. (IAEA 2023) p. 9 This means that a vast majority of the current nuclear power reactor fleet could adopt the practice of MOX fuel utilization.

4.2 Spent nuclear fuel recycling in fast reactors

MOX fuel can also be used in fast reactors. Of the six reactors in the GIF program the supercritical water-cooled reactor, sodium-cooled fast reactor, lead-cooled fast reactor and gas-cooled fast reactor could use MOX fuel. (Chauvin 2021)

As of 2021, there have been about 20 operational FBRs worldwide with over 400 reactor-years of combined operation. According to World Nuclear Association, Russia, India, China and Japan had operational experimental or demonstrational fast reactors that used MOX fuel in 2021. (World Nuclear Association 2021b) At the time of early development, FBRs were supposed to use plutonium derived from uranium as fuel. Using U-238 as fertile fuel would increase the available energy by 50-fold, according to IAEA. MOX fuel use in thermal reactors was thought of as a secondary alternative to the plutonium use in FBRs. (IAEA 2000) p.6

In the last century, FBRs were expected to become far more common. By the year 2000, there were expected to be over 500 FBRs in operation. That is why large-scale civilian plutonium separation processes were developed and allowed to continue after the need for weapons grade plutonium separation stopped. In reality, there were no plutonium utilizing FBRs of commercial scale in operation anywhere in the year 2000. (Schneider and Marignac 2008) p. 5 Another point justifying MOX fuel production to continue is that the facilities are not yet at the end of their service life. Decommissioning nuclear facilities is expensive so it is economically wiser to run the facilities as long as they last before eventual decommissioning.

The implementation of planned FRs was postponed in many countries because in the 1980s it was noticed that the demand for nuclear energy was far smaller than expected. (NEA and OECD 2013) p. 41 In Europe, France has been the only country to operate a commercial-scale FBR, the Superphénix. Other countries, such as Italy, Germany and the UK were onboard in the program. The reactor shut down in 1996 and the FBR program in France and practically rest of Europe was suspended. (Schneider and Marignac 2008)

Interest in developing FRs and FBRs has risen again in recent years. As of 2021, multiple countries, including the USA, Belgium, Romania and South Korea to name a few, had FR designs under active development. (World Nuclear Association 2021b) The development of FRs is essential to sustain and widen SNF recycling as its recycling currently is not economically really viable. This point is discussed further in the next chapter. Despite wide interest in FRs, they are estimated to become more common after 2050s. Technologies for FRs systems will not be available soon enough for them to replace reactors coming to their end of service life in the next two decades. The following large-scale replacement need of aging nuclear reactors will be around 2080-90s, which is

when FRs could become the industrial standard more realistically. (NEA and OECD 2013) p. 123 The age distribution of the current nuclear reactor fleet can be seen in Figure 6.

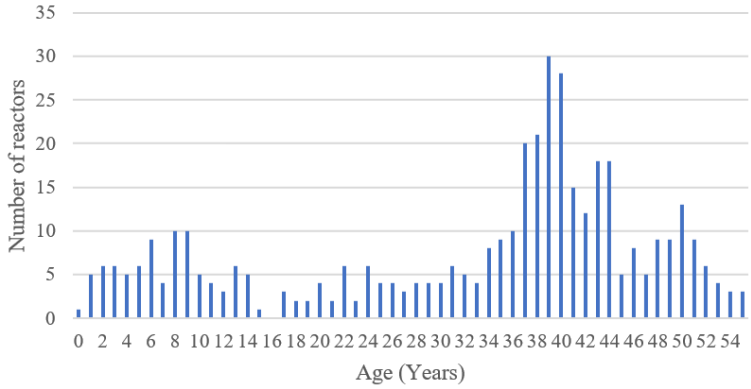

Figure 6: Current nuclear power fleet age distribution (IAEA 2024b)

An advantage of FRs considering the recycling of SNF is their ability to utilize many types of fuels and components unusable to thermal reactors. Fast reactors can burn MOX and RepU reprocessed from LWR fuel and thus reduce the stocks of plutonium and otherwise difficultly usable RepU. Additionally, fast reactors are able to burn other actinides that are fissionable to fast neutrons. Separation and incineration of the actinides from nuclear waste would reduce the radiotoxicity of the waste greatly. (Guidez and Prêle 2017) p. 301

MOX fuel behaves similarly in FRs compared to uranium fuel, as have been found in experimental and prototype FRs around the world, mainly in France and Russia. To make FRs competitive, MOX fuel should reach high burnups. Burnups of MOX fuel as high as 21% were analyzed in a report made in 2000. The report concluded that burnups of 20% should be safe for a commercial-scale reactor. (IAEA 2000) p. 49 Spent MOX fuel has not been widely recycled because the build-up of non-fissile isotopes of plutonium makes the fuel less efficient in LWRs. Also, the creation of other transuranic elements makes the spent fuel harder to recycle. Spent MOX fuel could be used in fast reactors instead, which are more suitable for recycling of fissile and fertile materials several times. More of the unwanted isotopes in LWR fuel are fissile in the fast neutron spectrum. However, the neutron reactions in FRs do not work entirely ideally and there are always some residual actinides and fission products that need to be disposed of as HLW. (NEA and OECD 2013) p. 30

5 ADVANTAGES AND DISADVANTAGES OF SPENT NUCLEAR FUEL RECYCLING

This chapter makes comparisons between the open nuclear fuel cycle, the partially closed nuclear fuel cycle and the fully closed fuel cycle in the environmental and economic aspects. The social and political aspects concerning SNF recycling are outside the scope of this thesis.

Recycling fissile and fertile materials from SNF increases the energy obtained from uranium. The World Nuclear Association estimates that recycling plutonium in MOX fuel gives 12% more energy from uranium compared to the open cycle. If the uranium in SNF is also recycled, 22% more energy can be obtained. (World Nuclear Association 2017) Estimates vary between sources, but Nuclear Energy Agency claims that between 30 to 60 times more energy could be extracted from uranium with a closed fuel cycle with advanced or fast reactors compared to an open fuel cycle. This is due to FRs´ ability utilize U-238 as fertile fuel. (Nuclear Energy Agency 2015) p. 13 The amounts of energy obtained from uranium compared between the fuel cycles can be observed in Figure 7.

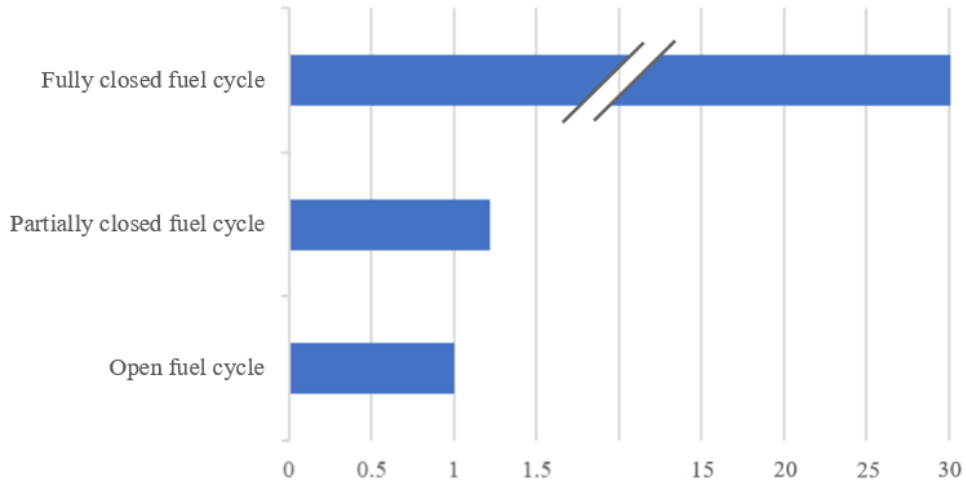

Figure 7. Energy obtained from uranium in relation to the open fuel cycle (World Nuclear Association 2017), (Nuclear Energy Agency 2015)

Recycling fissile and fertile materials in SNF reduces the need for mining uranium. In 2022 ESA estimated that using MOX fuel in reactors saved 3% of all-natural uranium fuel in the EU. (Euratom Supply Agency 2022) p. 25 An article summarizing a life cycle

analysis of the French nuclear fuel cycle finds that an open nuclear fuel cycle compared to the partially closed fuel cycle the French use would use approximately 16% more natural uranium to produce the same amount of energy. (Poinssot et al. 2014) A comparison of natural uranium consumption of the fuel cycles can be observed in Figure 8. Additionally, a closed fuel cycle reduces the need for importing uranium to countries that do not have exploitable uranium reserves of their own, contributing to energy security. FBRs that could use U-238 as fertile fuel, would lessen the need for enrichment facilities too. (Dunn Lee et al. 2023)

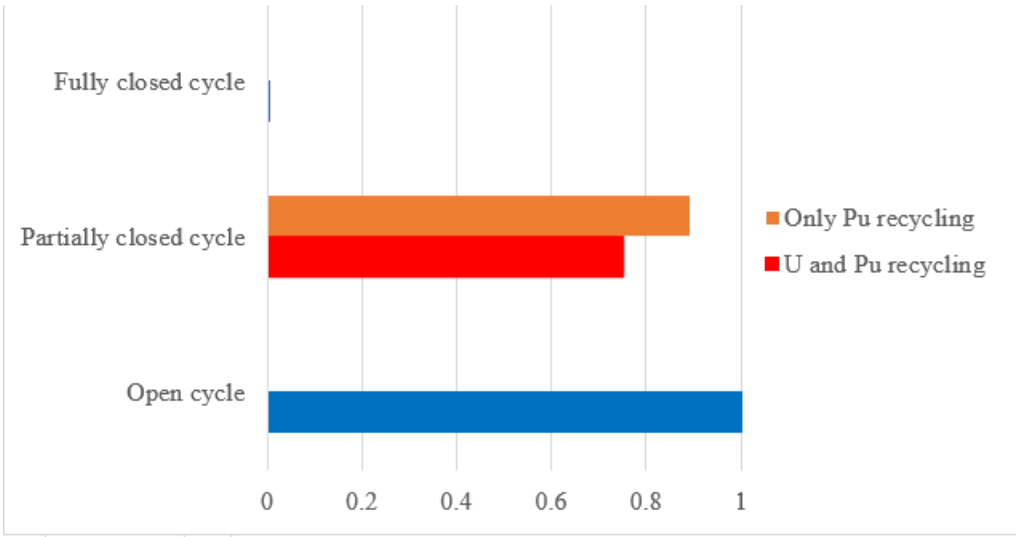

Figure 8: Natural uranium consumption in relation to open fuel cycle (OECD Nuclear Energy Agency 2006)

In the French nuclear fuel cycle assessment, the open fuel cycle is estimated to produce more HLW by volume and have an overall bigger environmental footprint. The vast majority of the selected indicators are in favor of the partially closed fuel cycle. Some gaseous radionuclide emissions are heightened with the partially closed fuel cycle, because the fission gases normally only get released in reprocessing. However, the heightened emissions represent only 1% of radioactivity compared to natural radioactivity annually. Characteristics concerning this thesis can be observed in Figure 9. Other characteristics in the assessment include greenhouse gas emissions, atmospheric and water pollution and ecotoxicity, to name a few, of which all are in favor of the partially closed fuel cycle. (Poinssot et al. 2014)

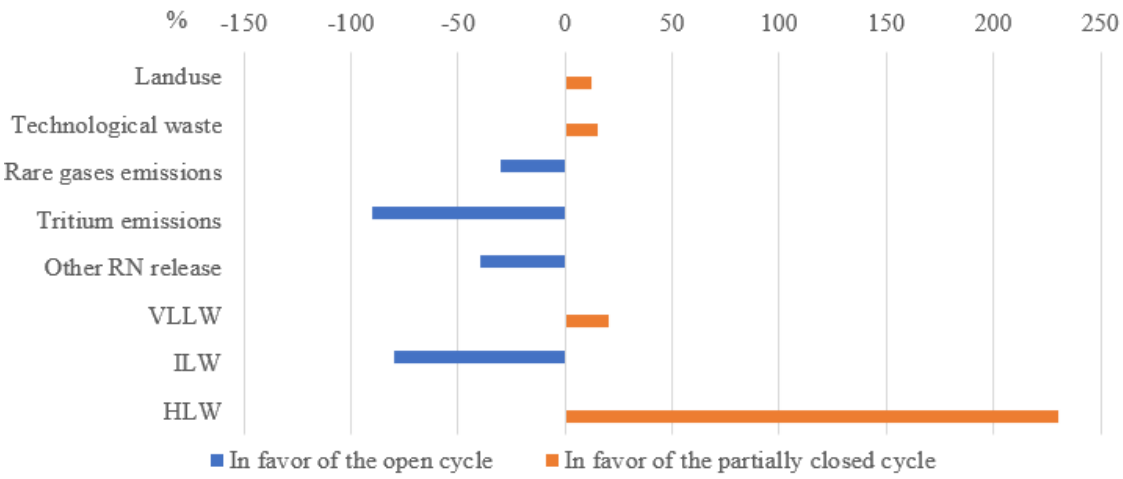

Figure 9: A comparison of open fuel cycle and partially closed fuel cycle characteristics (Poinssot et al. 2014)

Recycling changes the nature of the SNF waste, depending on what is recycled and how. The mass of the HLW material can be reduced by a lot if the uranium is recycled by returning it to a reactor or separated to depleted uranium. Separation of plutonium and minor actinides decreases the decay heat production and radiotoxicity. (Bodansky 2004) p. 193 If uranium, plutonium and minor actinides are separated from the HLW, the radiotoxicity of the waste will lower to the levels of mined uranium after 1000 years. For open cycle HLW that will take 'several 100 000 years'. Thermal load of the HLW would reduce to the same levels in 10 000 years in the open fuel cycle and in 300 years with uranium, plutonium and minor actinide recycling. Radiotoxicity and thermal load of the HLW affect the design requirements for DGRs. HLW emitting less radiation and heat needs smaller repositories. (González-Romero 2011)

Amounts of HLW in different fuel cycles in relation to the open fuel cycle can be seen in Figure 10. Additionally, a comparison of decay heat reduction of different fuel cycles between 50 and 200 years in storage can be seen in Figure 11.

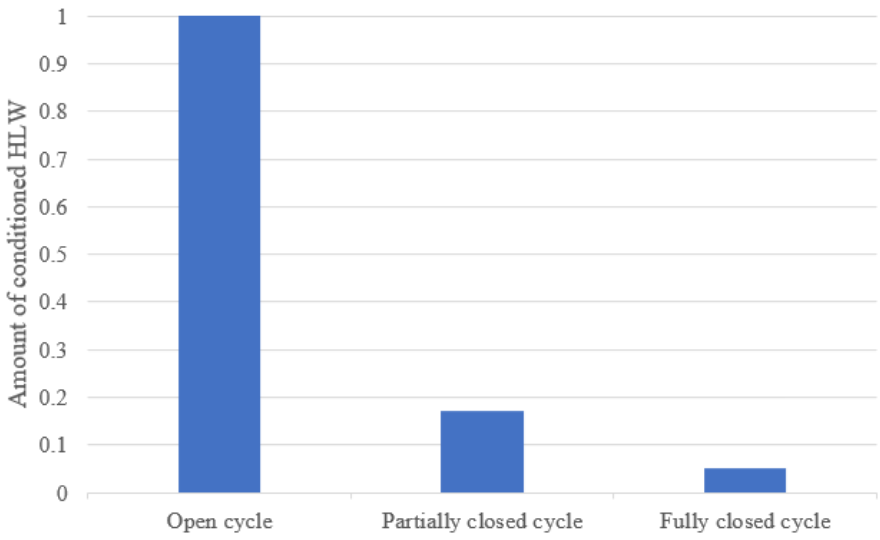

Figure 10: Volumes of HLW in fuel cycles in relation to the open fuel cycle (NEA and OECD 2021)

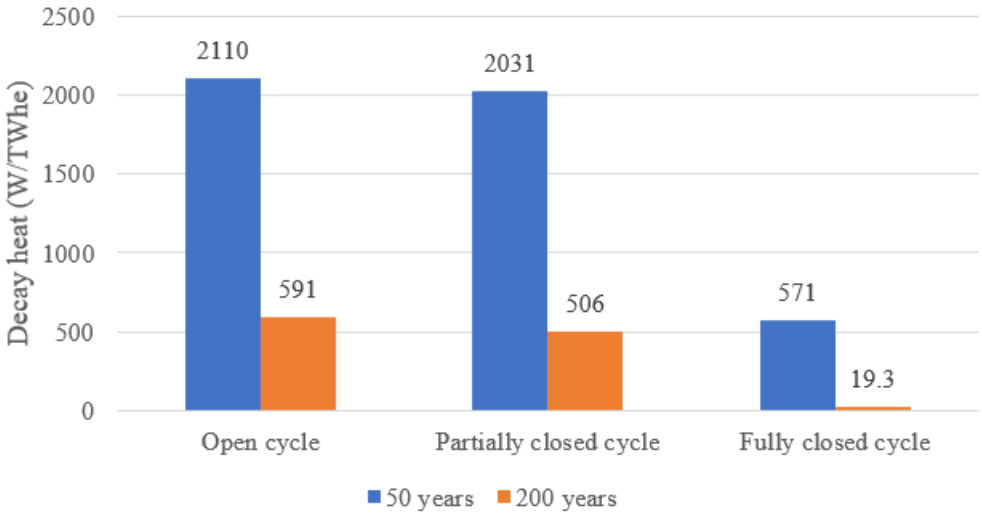

Figure 11: Decay heat at 50 years and 200 years for fuel cycle options (OECD Nuclear Energy Agency 2006)

A research report on SNF reprocessing in France finds shortcomings in the claim that reprocessing dramatically reduces the volume of radioactive waste and its effect on DGRs. They include not taking into account the wastes from reprocessing facilities decommissioning, lower-level waste generation of the reprocessing and the larger required final disposal sites for MOX fuel due to its higher heat generation compared to the used uranium fuel. (Schneider and Marignac 2008) p. 3

The cost of recycling SNF is a big disadvantage. Recycling costs have been mostly higher than the cost of making uranium fuel from newly mined ore. The advantages of recycling might become more relevant if the use of nuclear energy expands a lot and the reactor fleet that can use recycled nuclear fuels becomes vaster. (Bodansky 2004) A large share of the costs for recycling arise from investments in new facilities and equipment. Establishing PUREX and MOX fabrication plants is costly as well as modifying already existing enrichment plants for managing RepU. (Dunn Lee et al. 2023) p. 128

In 2009 the Electric Power Research Institute made a report comparing costs between a once-through and plutonium single-recycling fuel cycles. Results in the report presented that there would not be economic incentive for spent nuclear fuel recycling in the US under present unit costs of uranium and the costs for PUREX processing and MOX manufacturing. (Supko et al. 2009) A summary of fuel cycle strategies by country shows that only countries with a large nuclear fleet have adopted a partially closed fuel cycle. Recycling SNF can be viable for a country with a small nuclear fleet if the spent fuel is transported to another country to be reprocessed. (IAEA 2022)

Fast reactor fuel cycle costs are hard to estimate. Fast reactor fuel cycle requires reprocessing and fuel fabrication plants, which increase the cost of energy produced with fast reactors. The diminished amount and radiotoxicity of HLW lessens the need for space in DGRs which decreases the cost of energy produced. (NEA and OECD 2013) p.123 According to J. Guidez and G. Prêle FRs enable electricity production for millennia. There is so much depleted uranium in stocks that there would be no need for uranium mines or enrichment facilities. (Guidez and Prêle 2017) This would further decrease the costs of a FR fuel cycle compared to the once-through and partially closed cycles. Additionally, the environmental impact of no more uranium mining would be substantial.

6 CONCLUSIONS

Spent nuclear fuel recycling has reached maturity in technologies available. PUREX process to extract plutonium and uranium has proved to be an efficient part in the SNF recycling process line. MOX fuel fabrication and use in LWRs has become standard practice in France where SNF recycling is commercially deployed of European countries. There is room to develop these processes to match future needs in fuel design, for example, extracting minor actinides from the waste of PUREX process. It will become more topical when FRs systems become available.

Closing the nuclear fuel cycle by deploying fast reactors has significant environmental benefits. MOX fuel use lessens the demand for uranium mining. Fast reactors could utilize uranium and plutonium in a way that uranium mining is minimized greatly. Fast reactors could extract 30 times and up the amount of energy from uranium fuel as LWRs do. Many environmental impacts of nuclear power are lowered with SNF recycling. However, recycling increases the release of radionuclides that only get released in the reprocessing process. Designing recycling facilities to safely manage these emissions is crucial but it adds up to the costs of recycling.

Aspects speaking against closing the fuel cycle are largely economic. The production of MOX fuel is more expensive than making uranium fuel from mined ore. Building and operating the recycling facilities add to the cost of nuclear energy. While uranium is plentiful and easily available currently, the additional costs from recycling make it widely non-viable economically. Countries with large nuclear fleets are the exception, where other interests, such as the need to reduce plutonium stocks, override the disadvantage of the costs. Countries with smaller fleets might find it of interest to have their spent fuel reprocessed in a country with the facilities.

Research and development of advanced reactors and fuel cycles have proved that closing the nuclear fuel cycle is possible but is yet to be deployed. FRs technologies will not come in time for the replacement of the widely ageing current nuclear fleet. However, the fleet after could widely consist of fast reactors.

This thesis studied reprocessing technologies for spent nuclear fuel and systems already utilizing fuel made from recycled materials. This thesis also found that closing the nuclear

fuel cycle fully is a possibility. Next topics that could be studied are the ones intentionally left out of this thesis, although they are important aspects of recycling nuclear fuel. They include topics more in the legislative, political and social side of nuclear energy production. For example, security of recycling and the regulations and licenses in order to prevent proliferation were a common point for and against SNF recycling.

REFERENCES

Bodansky, D., 2004. *NUCLEAR ENERGY* [online]. Available from: http://www.gammaexplorer.com/wp-content/uploads/2014/03/Nuclear-Energy-Principles-Practices-and-Prospects-Bodansky-Springer-2004.pdf [Accessed 25 Jan 2024].

Chauvin, N., 2021. *MOX FUEL FOR ADVANCED REACTORS* [online]. Available from: https://gif.jaea.go.jp/webinar/Series49/gen_iv_webinar_series_49_dr._nathalie-chauvin_28_jan_2021_final.pdf [Accessed 12 Feb 2024].

Dunn Lee, J., Baisden, P. A., Ewing, R. C., Chu, M. S. Y., Dickman, P. T., Hansen, C. S., Lee, J. C., Lyman, E. S., Macfarlane, A. M., Machiels, A. J., Poinssot, C., Semancik, J. D., Sorenson, K. B., Vujic, J. L., Wall, N. A. and Wood, H. G., 2023. *Merits and Viability of Different Nuclear Fuel Cycles and Technology Options and the Waste Aspects of Advanced Nuclear Reactors* [online]. Merits and Viability of Different Nuclear Fuel Cycles and Technology Options and the Waste Aspects of Advanced Nuclear Reactors. National Academies Press. Available from: http://nap.nationalacademies.org/26500 [Accessed 15 Jan 2024].

Euratom Supply Agency, 2022. *ANNUAL REPORT 2022* [online]. Available from: https://euratom-supply.ec.europa.eu/system/files/2023-10/ESA%20Annual%20Report%202022%20-%20Final%20%28website%29_2.pdf [Accessed 24 Jan 2024].

Gerber, M. S., 1993. Brief history of PUREX. [online]. Available from: https://www.osti.gov/servlets/purl/10115226-JdCrRH/native/ [Accessed 26 Jan 2024].

González-Romero, E. M., 2011. Impact of partitioning and transmutation on the high level waste management. *Nuclear Engineering and Design* [online], 241 (9), 3436–3444. Available from: https://www.sciencedirect.com/science/article/pii/S0029549311002366 [Accessed 5 Mar 2024].

Guidez, J. and Prêle, G., 2017. *Superphenix Technical and Scientific Achievements* [online]. Available from: https://link.springer.com/book/10.2991/978-94-6239-246-5 [Accessed 12 Mar 2024].

Gupta, H. P., Menon, S. V. G. and Banerjee, S., 2008. Advanced fuel cycles for use in PHWRs. *Journal of Nuclear Materials* [online], 383 (1–2), 54–62. Available from: https://www.sciencedirect.com/science/article/abs/pii/S0022311508004777 [Accessed 12 Feb 2024].

IAEA, 2000. *PROCEEDINGS MOX Fuel Cycle Technologies for Medium and Long Term Deployment INTERNATIONAL ATOMIC ENERGY AGENCY* [online]. Available from: https://www-pub.iaea.org/MTCD/Publications/PDF/CSP3P.pdf [Accessed 12 Feb 2024].

IAEA, 2003. *Status and Advances in MOX Fuel Technology INTERNATIONAL ATOMIC ENERGY AGENCY,VIENNA, 2003* [online]. Available from: https://www-pub.iaea.org/MTCD/Publications/PDF/TRS415_web.pdf [Accessed 12 Mar 2024].

IAEA, 2005. *FISSILE MATERIAL MANAGEMENT STRATEGIES FOR SUSTAINABLE NUCLEAR ENERGY* [online]. Available from: https://www-pub.iaea.org/MTCD/Publications/PDF/P1288_web.pdf [Accessed 12 Dec 2023].

IAEA, 2022. *IAEA Nuclear Energy Series @ Status and Trends in Spent Fuel and Radioactive Waste Management Status and Trends in Spent Fuel and Radioactive Waste Management* [online]. Vienna. Available from: https://www-pub.iaea.org/MTCD/Publications/PDF/PUB1963_web.pdf [Accessed 12 Dec 2023].

IAEA, 2023. Nuclear Power Reactors in the World. [online]. Available from: https://www-pub.iaea.org/MTCD/Publications/PDF/RDS-2-43_web.pdf [Accessed 20 Feb 2024].

IAEA, 2024a. Power Reactor Information System. [online]. Available from: https://pris.iaea.org/PRIS/home.aspx [Accessed 19 Feb 2024].

IAEA, 2024b. Age Distribution. [online]. Available from: https://pris.iaea.org/PRIS/WorldStatistics/OperationalByAge.aspx [Accessed 8 Mar 2024].

Irish, E. R. and Reas, W. H., 1957. *THE PUREX PROCESS-A SOLVENT EXTRACTION REPROCESSING METHOD FOR IRRADIATED URANIUM GENERAL^ELECTRIC UNCLASSIFIED* [online]. Available from: https://www.osti.gov/servlets/purl/4341712 [Accessed 8 Jan 2024].

Nawada, H. P., Ganguly, C. and International Atomic Energy Agency., 2007. *Management of reprocessed uranium : current status and future prospects.* [online]. International Atomic Energy Agency. Available from: https://www-pub.iaea.org/MTCD/Publications/PDF/te_1529_web.pdf [Accessed 5 Mar 2024].

NEA and OECD, 2013. *The Economics of the Back End of the Nuclear Fuel Cycle* [online]. Available from: https://www.oecd-nea.org/jcms/pl_14764/the-economics-of-the-back-end-of-the-nuclear-fuel-cycle?details=true [Accessed 1 Feb 2024].

NEA and OECD, 2021. *Strategies and Considerations for the Back End of the Fuel Cycle* [online]. Available from: https://www.oecd-nea.org/jcms/pl_55928/strategies-and-considerations-for-the-back-end-of-the-fuel-cycle?details=true [Accessed 5 Mar 2024].

Nuclear Energy Agency, 2015. *Nuclear Energy: Combating Climate Change* [online]. Available from: https://www.oecd-nea.org/jcms/pl_14914/nuclear-energy-combating-climate-change?details=true [Accessed 12 Mar 2024].

OECD Nuclear Energy Agency, 2006. *Advanced nuclear fuel cycles and radioactive waste management.* [online]. Nuclear Energy Agency, Organisation for Economic Co-operation and Development. Available from: https://www.oecd-nea.org/upload/docs/application/pdf/2019-12/5990-advanced-nfc-rwm.pdf [Accessed 5 Mar 2024].

Poinssot, Ch., Bourg, S., Ouvrier N., Combernoux N., Rostaing C., Vargas-Gonzalez M. and Bruno J., 2014. Assessment of the environmental footprint of nuclear energy systems. [online], 69, 199–211. Available from:

https://www.sciencedirect.com/science/article/pii/S0360544214002035?via%3Dihub [Accessed 5 Mar 2024].

Schneider, M. and Marignac, Y., 2008. *International Panel on Fissile Materials Spent Nuclear Fuel Reprocessing in France* [online]. Available from: https://fissilematerials.org/library/rr04.pdf [Accessed 12 Mar 2024].

Spinrad, B. I. and Marcum, W., 2024. Nuclear Reactor. *Encyclopedia Britannica* [online]. Available from: https://www.britannica.com/technology/nuclear-reactor [Accessed 7 Feb 2024].

Supko, E., Meade, T. and Schwartz, M., 2009. *Nuclear Fuel Cycle Cost Comparison Between Once-Through and Plutonium Single-Recycling in Pressurized Water Reactors*[online]. Available from: https://www.epri.com/research/products/1018575 [Accessed 12 Mar 2024].

World Nuclear Association, 2017. Mixed oxide (MOX) fuel. [online]. Available from: https://world-nuclear.org/information-library/nuclear-fuel-cycle/fuel-recycling/mixed-oxide-fuel-mox.aspx [Accessed 12 Dec 2023].

World Nuclear Association, 2020. Processing of Used Nuclear Fuel. [online]. Available from: https://world-nuclear.org/information-library/nuclear-fuel-cycle/fuel-recycling/processing-of-used-nuclear-fuel.aspx [Accessed 5 Mar 2024].

World Nuclear Association, 2021a. *Nuclear Fuel Cycle Overview* [online]. World Nuclear Association. Available from: https://world-nuclear.org/information-library/nuclear-fuel-cycle/introduction/nuclear-fuel-cycle-overview.aspx [Accessed 12 Dec 2023].

World Nuclear Association, 2021b. Fast Neutron Reactors. [online]. Available from: https://world-nuclear.org/information-library/current-and-future-generation/fast-neutron-reactors.aspx [Accessed 12 Feb 2024].

World Nuclear Association, 2023. How Can Nuclear Combat Climate Change. [online]. Available from: https://world-nuclear.org/nuclear-essentials/how-can-nuclear-combat-climate-change.aspx [Accessed 12 Dec 2023].