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Physical Analysis and Prospects of Accelerator-Driven Systems for Transmutation of Spent Nuclear Fuel

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Abstract

La gestione dei rifiuti radioattivi, in particolare la minimizzazione della radiotossicità a lungo termine dovuta ad Attinidi Minori (MA) e Prodotti di Fissione a Lunga Vita (LLFP), rappresenta una sfida cruciale per l'energia nucleare. Questa tesi analizza i Sistemi Accelerator - Driven (ADSs) come potenziali soluzioni. Un ADS integra un acceleratore di particelle, che genera neutroni tramite spallazione su un target pesante, con un reattore nucleare operante in regime sottocritico ($k_{\text{eff}} < 1$). I neutroni esterni pilotano il reattore, sostenendo la fissione e la trasmutazione dei radionuclidi problematici.

Vengono discussi i fondamenti fisici e neutronici degli ADS, includendo il guadagno energetico, la cinetica in presenza di sorgente esterna e il bilancio neutronico essenziale per la trasmutazione. Si evidenziano i vantaggi intrinseci di sicurezza della sottocriticità e la flessibilità nella composizione del combustibile, favorendo l'incenerimento degli MA. La trattazione include i componenti chiave (acceleratore, target, nocciolo sottocritico) e le relative sfide tecnologiche. Infine, si offre una panoramica su proposte storiche e progetti di ricerca internazionali, come MYRRHA, volti a dimostrare la fattibilità di tali sistemi per un ciclo del combustibile nucleare più sostenibile.

Abstract

The management of nuclear waste, particularly the minimization of long-term radiotoxicity from Minor Actinides (MA) and Long-Lived Fission Products (LLFPs), represents a crucial challenge for nuclear energy. This thesis analyzes Accelerator-Driven Systems (ADSs) as potential solutions. An ADS integrates a particle accelerator, which generates neutrons via spallation on a heavy target, with a nuclear reactor operating in a subcritical state ($k_{\text{eff}} < 1$). The external neutrons drive the reactor, sustaining fission and the transmutation of problematic radionuclides.

The physical and neutronic fundamentals of ADS are discussed, including energy gain, kinetics in the presence of an external source, and the neutron balance essential for transmutation. The inherent safety advantages of subcriticality and the flexibility in fuel composition, which favors the incineration of MAs, are highlighted. The discussion covers the key components (accelerator, target, subcritical core) and their related technological challenges. Finally, an overview of historical proposals and international research projects, such as MYRRHA, is provided. These projects aim to demonstrate the feasibility of such systems for a more sustainable nuclear fuel cycle.

Contents

Introduction

The question of how energy can be produced in the most economically, politically and environmentally viable way has been shaping government policy all around the world for the last decades. Climate change poses a great challenge to humanity, so finding and using the cleanest energy source possible is the key to solving the problem. In 2023, nuclear power accounted for about 10% of electricity production in the world and is considered to be one of the safest and to have the lowest CO_2-equivalent emissions along with wind power of all energy sources.

One of the challenges of Nuclear Power is the management of nuclear waste. Depending on their classification regarding the hazard they pose to human health, nuclear wastes are either stored in temporary/permanent surface storage, reprocessed using chemical processes or stored in geological repositories. Different nations choose their preferred method. For example, Finland built the Onkalo spent nuclear fuel repository, which is the first deep geological repository for final and permanent disposal of spent nuclear fuel, the most hazardous nuclear waste produced by Nuclear reactors. France, the country with the highest share of nuclear power production in the world, reprocesses almost 100% of all Spent Nuclear Fuel (SNF) produced, which gets reused as MOX (Mixed Oxide fuel). However, only 96% of SNF can be reprocessed, while the other 4%, composed mostly of Minor Actinides (MAs) and Fission Products (FPs), is the most problematic part as it is difficult to manage.

Solutions for managing MAs and FPs include fourth-generation reactors, such as Fast Neutron Reactors, and the main topic of this thesis: Accelerator-Driven Systems (ADSs).

Chapter 1

Nuclear reactors and the nuclear fuel cycle

1.1 Principles of nuclear fission

Nuclear reactors are based on nuclear fission, which releases a lot of energy with each process. In the 1930s it was found that some nuclei, such as ^{235}U, would break apart if bombarded with neutrons, resulting in reaction products with a lower atomic number Z than the initial atom. The basic reaction at the base of nuclear fission involving ^{235}U is

$$\mathrm{n} + {}^{235}\mathrm{U} \rightarrow {}^{236}\mathrm{U}^{*} \rightarrow \mathrm{FPs} + \mathrm{a\ few\ n} + \mathrm{energy} \tag{1.1.1}$$

where FPs are Fission Products. FPs, as mentioned in the introduction, are of particular interest as some of them cannot be reprocessed and have long half-lives. For each fission of ^{235}U, an average of 2.44 neutrons is produced along with ~ 200 MeV, distributed as described in Table 1.1. After ^{235}U captures a neutron, it briefly becomes ^{236}U*, where * indicates an excited state of U which quickly decays.

The neutrons produced during the process make it possible to sustain a self-sustaining nuclear chain reaction, where one atom of Uranium is hit with one neutron, producing multiple neutrons, which then hit a different atom that undergoes fission, etc. This is essentially how reactors are able to produce a large amount of energy and how nuclear power is exploited by the military.

Energy Component	**Average Energy (MeV)**
Instantaneously Released Energy	
Kinetic energy of fission fragments	168
Kinetic energy of prompt neutrons	5
Energy carried by prompt γ-rays	7
Energy from Decaying Fission Products	
Energy of β^- particles	8
Energy of delayed γ-rays	7
Total Recoverable Energy	**195**
Energy of neutrinos (lost)	12
Total Energy per Fission	**207**

Table 1.1: *A breakdown of the average energy released (in MeV) per fission event of a ^{235}U nucleus. The table distinguishes between instantaneously released energy and the delayed energy from decaying fission products.* [1]

1.1.1 Fission Products

As previously mentioned, besides energy and prompt neutrons, fission products are produced as well in a fission process. Moreover, the fission fragments are not uniquely defined and there is a specific distribution of masses for the two fission nuclei. The distribution of the yields is shown in Figure 1.1. As can be seen from the graph, the likelihood of having two FPs with the same mass is negligible, while the most probable outcome is two FPs with $A \sim 95$ and $A \sim 135$, therefore a wide range of nuclides gets produced around these numbers. While most of them have short to medium half-lives, some of them have half-lives in the order of millions of years, making them particularly difficult to manage in the long term.

Even though they make up only about 5% of the mass of spent fuel, FPs are the dominant source of radioactivity in spent fuel. For example, ^{131}I and ^{133}Xe have half - lives of 8.04 and 5.25 days respectively, making them harmful in the short term. After several years of cooling, most radioactivity is from the fission products ^{137}Cs and ^{90}Sr, which are each produced in about 6% of fissions, and have half-lives of about 30 years. Other fission products with similar half-lives have much lower fission product yields, lower decay energy, and several are also quickly destroyed by neutron capture while still in the reactor, so they are not responsible for more than a tiny fraction of the radiation production at any time. Long-Lived Fission Products (LLFPs), such as ^{129}I and especially ^{135}Cs and ^{99}Tc

which have a yield of ∼6%, represent a significant proportion of the remaining radioactivity, along with longer-lived actinides like ^{237}Np and ^{242}Pu.

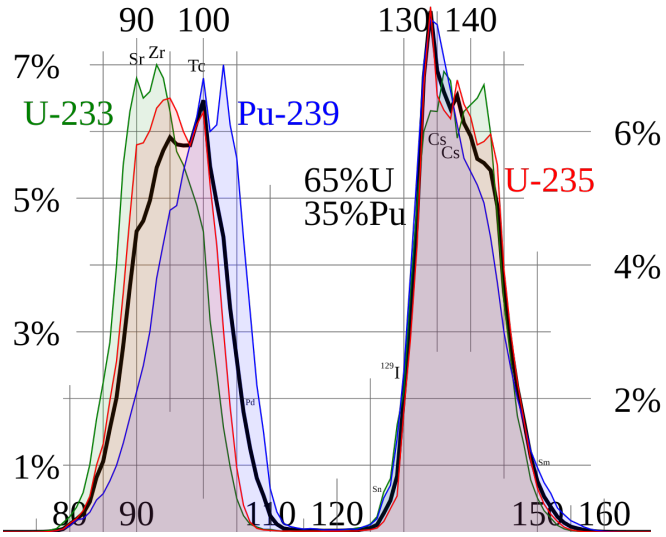

Figure 1.1: *Mass distribution of fission products for various fuels under thermal neutron fission. The characteristic double-humped curve shows the high probability of asymmetric fission, with peaks around mass numbers A∼95 and A∼135. The fission yields for* ^{235}*U,* ^{239}*Pu,* ^{233}*U, and a typical LWR fuel mix are compared.* [2]

Other fission products, such as ^{135}Xe and ^{149}Sm, have high neutron absorption cross sections. Since a nuclear reactor must balance neutron production and absorption rates, fission products that absorb neutrons tend to poison or shut the reactor down; this is controlled with burnable poisons and control rods. Build-up of ^{135}Xe during shutdown or low-power operation may poison the reactor enough to impede restart or interfere with normal control of the reaction during restart or restoration of full power.

1.1.2 Nuclear cross section

One of the most important concepts in nuclear reactor physics is the fission cross section σ_f, measured in barns (b, 1 $b = 10^{-24}\,\mathrm{cm}^2$). It quantifies the probability that a neutron incident on a specific fissile or fissionable nucleus will induce a fission reaction.

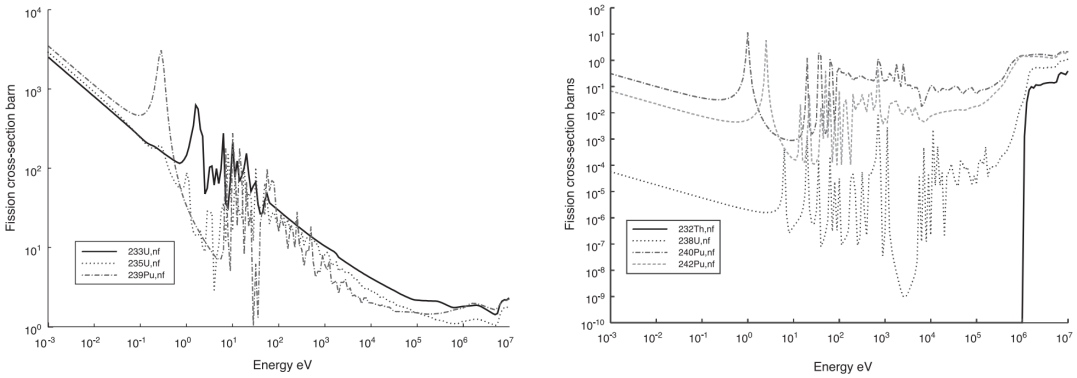

Figure 1.2: *Fission cross-section (in barns) as a function of incident neutron energy (in eV). The left panel displays the high thermal cross-sections characteristic of fissile nuclei (^{233}U, ^{235}U, ^{239}Pu). The right panel shows the behavior of various fissionable isotopes, highlighting that some (like ^{238}U) require high-energy (fast) neutrons to undergo fission effectively.* [3]

As shown in Figure 1.2, σ_f varies significantly depending on the energy of the incident neutron. ^{235}U, like other fissile nuclides, has a greater σ_f at lower energy, in particular at ~ 0.025 eV, which is the energy a thermal neutron has. This is one of the main reasons most nuclear reactors operate with thermal neutrons and not fast neutrons. Fast reactors, which use fast neutrons, can take advantage of the possibility of having a higher neutron flux, despite the lower probability of undergoing fission for some isotopes. Another advantage of fast neutrons is that the fission cross-section σ_f of many fissionable but not fissile nuclides (such as ^{238}U) becomes significant.

Other types of nuclear cross sections exist, such as the capture cross section σ_c. It quantifies the probability that a neutron incident on a specific nucleus will be captured. As said before, there are certain nuclides which have a large σ_c for thermal neutrons; therefore, they act as poison for the reactors because they stop the fission chain. Neutron capture by fertile nuclei eventually leads to the production of a fissile species, usually following beta decay:

$$^{232}\text{Th} + \text{n} \rightarrow ^{233}\text{Th} \xrightarrow{\beta^-,22.3\text{ min}} ^{233}\text{Pa} \xrightarrow{\beta^-,26.97\text{ days}} ^{233}\text{U} \tag{1.1.2}$$

$$^{238}\text{U} + \text{n} \rightarrow ^{239}\text{U} \xrightarrow{\beta^-,23.45\text{ min}} ^{239}\text{Np} \xrightarrow{\beta^-,2.356\text{ days}} ^{239}\text{Pu}. \tag{1.1.3}$$

The ability of ^{238}U to capture neutrons and subsequent decay into ^{239}Pu, a fissile material used as nuclear fuel, significantly extends the usable fuel resources. This process is at the core of Breeder Reactors and of spent fuel reprocessing in order to produce MOX (Mixed Oxide) fuel and will be later explored.

An important quantity, called η, is the average number of fission neutrons produced per neutron absorbed by the fissile material. This quantity is crucial to the possibility of establishing a chain reaction. It can be split into two factors: the probability that an absorption leads to fission, $\frac{\sigma_f}{\sigma_f+\sigma_c} = \frac{1}{1+\alpha}$ where $\alpha = \frac{\sigma_c}{\sigma_f}$, and the mean number of neutrons ν emitted per fission. Then,

$$\eta = \frac{\nu}{1+\alpha} \tag{1.1.4}$$

and the absorption cross section is $\sigma_a = \sigma_c + \sigma_f$. Essentially, η quantifies the neutron yield efficiency of the fissile material itself, accounting for the neutrons lost through non-fission absorption processes.
We can now distinguish between fissile, fissionable and fertile nuclides: fissionable refers to any material capable of undergoing nuclear fission when struck by a neutron, either at low or high energy. Fissile is a subset of fissionable materials, specifically those that readily undergo fission when struck by slow (thermal) neutrons. Fertile materials, on the other hand, are not inherently fissile but can be converted into fissile material through neutron capture and subsequent nuclear decay. In Table 1.2 data is shown for absorption, capture and fission cross section of different nuclides.

Nucleus	σ_f (b)	σ_c (b)	σ_a (b)
^{232}Th	$< 10^{-6}$	7.34	7.34
^{233}U	531	45.3	576
^{235}U	585	98.7	684
^{238}U	1.68×10^{-5}	2.68	2.68
^{239}Pu	748	271	1019
^{135}Xe	≈ 0	2.65×10^6	2.65×10^6
^{149}Sm	0.0	42 000	42 000

Table 1.2: *Fission (σ_f), capture (σ_c), and absorption (σ_a) cross-sections (in barns) for key fertile and fissile isotopes when interacting with thermal neutrons (0.025 eV). The absorption cross-section of reactor poisons like ^{135}Xe is also shown for comparison.* [4]

1.2 Conventional nuclear reactors

1.2.1 Core concepts

We have seen that the average number ν of prompt neutrons emitted in a fission reaction is ~ 2.4 and that these neutrons may induce further fission reactions in other nuclei in a chain reaction. This suggests that the number of neutrons could

increase exponentially, potentially leading to an uncontrolled rise in reaction rate, thus resulting in an explosion of energy. Indeed, this is what happens in an atomic bomb. On the other hand, it is possible to control the number of neutrons so that their number remains constant from one generation of fission reactions to the next. This produces a steady output of energy and this is the situation in the core of a nuclear reactor. Whichever happens, atomic bomb or energy source, depends on how many of the two or three emitted neutrons go on to produce fission in another nucleus.

This discussion focuses on the fission of ^{235}U by thermal neutrons, as it is the process that is used in most commercial reactors, such as the PWR (Pressured Water Reactor) and the BWR (Boiled Water Reactor). We might expect that each thermal neutron will, on average, lead to the production of ν thermal neutrons to induce the following generation of fission reactions in the chain. However, this is not the case. Previously, the number η has been already introduced. A neutron created in a medium (which we first consider infinite) containing fissile nuclei will give birth to k_∞ second generation neutrons. For limited medium one has to replace k_∞ by an effective value of k_{eff} which is less than k_∞ due to neutrons escaping from the system. One should also consider local values, k_s, dependent on the specific location of the apparition of the initial neutron. If k_{eff} is larger than unity the reaction diverges, which means from one initial neutron the final number of neutron goes to infinity, but a controlled divergence allows one to start a reactor. When k_{eff} is kept equal to 1 we obtain a critical reactor. The possibility to keep precisely the condition $k_{\text{eff}} = 1$ is due to the presence of a small fraction of delayed neutrons which allow time to compensate for deviation of the criticality coefficient k_{eff} from unity. If k_{eff} is less than unity an incident neutron gives birth to a finite number of secondary neutrons. The medium is said to be multiplying with a factor of $\frac{1}{1-k_{\text{eff}}}$.

To derive an expression for k_∞ we assume that the only possible reactions are scattering, capture and fission. Since the number k_∞ is the number of secondary neutrons produced, on the average, following absorption of the primary neutron one can write

$$\begin{aligned} k_\infty &= \langle\nu\rangle \frac{\text{probability for fission after absorption}}{\text{probability of absorption}} \\ &= \langle\nu\rangle \frac{\text{number of fissions after absorption}}{\text{number of absorptions}} \end{aligned} \tag{1.2.1}$$

where $\langle\nu\rangle$ is the average number of neutrons emitted per fission.
It is often useful and quite common to write k_∞ as a product of four factors

$$k_\infty = \varepsilon p \eta f \tag{1.2.2}$$

where:

- $\varepsilon \approx 1.02$ is the enhancement factor due to fertile nuclei fissions occurring by fast neutrons;
- $f \approx 0.71$ is the probability that the neutron absorption occurs in the fuel;
- $p \approx 0.87$ is the probability for a neutron absorbed in the fuel to be specifically absorbed by a fissile nucleus;
- $\eta \approx 1.65$ is the mean number of neutrons emitted following an absorption in a fissile nucleus (encountered before).

Clearly for a chain reaction to occur, $k_{\text{eff}} \geq 1$. If a nuclear reactor has $k_{\text{eff}} > 1$ it is said to be *supercritical*. If $k_{\text{eff}} = 1$, it is said to be *critical*, which is the condition for the steady production of energy. For $k_{\text{eff}} < 1$, it is said to be *subcritical*. (For a more complete description of k_∞, see: [5])
We then denote the reactivity as

$$\rho = \frac{k_{\text{eff}} - 1}{k_{\text{eff}}} \tag{1.2.3}$$

1.2.2 Commercial reactors

The neutrons generated from fission have energies ranging from a few hundred keV to about 10 MeV and the average energy of fission neutrons is 1.98 MeV. Probability of interaction is strongly affected by energy of the neutrons, as seen in Figure 1.2. If the energy is 10 keV or upward, the neutron is referred to as *fast*. Neutrons with energies below 10 keV but above 0.5 eV are called *epithermal* neutrons. All neutrons with energies below 0.5 eV are usually called *thermal* neutrons. Thermal neutrons have lost most of its initial energy and have come to thermal equilibrium with their surroundings. In the case of thermal equilibrium at 20 °C, the corresponding neutron energy is about 0.025 eV.
Most of the commercially operating nuclear reactors in the world are thermal reactors. This is because the cross section for nuclear fission reaction is much larger for thermal neutrons than for the fast or epithermal neutrons. The process of neutron slowing down to increase the probability of fission is called moderation. The material used for moderation is called a moderator. In most reactors, the material used for moderation is also used as coolant to extract thermal energy. Depending upon the coolant material used, nuclear reactor can also be classified. If light water is used as coolant, the reactor is called light water reactor (LWR). LWR includes PWR and BWR. Among 450 commercially operating nuclear reactors in the world (as of 2016), 291 are PWRs, 78 are BWRs. A schematic of a typical PWR power plant is shown in Figure 1.3.

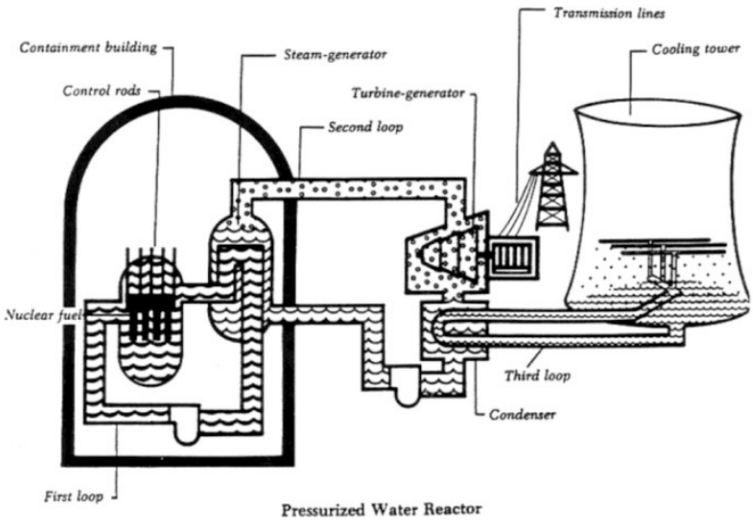

Figure 1.3: *Schematic of a Pressurized Water Reactor (PWR) power plant. It illustrates the three main cooling loops: the primary loop, the secondary loop and the tertiary loop.* [6]

Moderation of fast neutrons is achieved by passing the neutrons through a material of low atomic mass. The neutrons make numerous elastic collisions with the nuclei in the material and steadily lose their energy until they become in thermal equilibrium with the material.
Natural uranium contains 0.72% of ^{235}U and 99.3% of ^{238}U; consequently, light water reactors need to use enriched uranium fuel. Typically, the fraction of the isotope ^{235}U is increased to $\sim 2-5\%$. The ability to control the neutron reproduction factor k_{eff} is essential for the safe operation of a nuclear reactor. This is achieved by inserting control rods into the reactor core. These rods are made from a material such as cadmium that has a very large cross-section for the absorption of thermal neutrons and can therefore remove neutrons from the chain reaction very effectively; furthermore, the rods can be used to stop the chain reaction if necessary. Over time there will inevitably be variations in k_{eff} and it is necessary to manipulate the control rods to take account of these variations.
A schematic diagram of a nuclear reactor based on thermal fission of uranium is presented in Figure 1.4, and shows its main components. The core of the reactor contains the nuclear fuel, the moderator and the control rods. It is surrounded by a neutron reflector that reflects escaping neutrons back into the core. The whole assembly is housed in a steel pressure vessel. The kinetic energy of the fission fragments is absorbed in the reactor core and transformed into thermal energy. The thermal energy is extracted by a coolant, which circulates through the core. The thermal energy is used to produce steam that drives a turbine that, in turn,

drives an electrical generator. A reactor is a powerful source of nuclear radiation and so it is contained within a biological shield, which is typically a thick layer of concrete but also contains heavy metals such as lead that absorb the γ-rays.

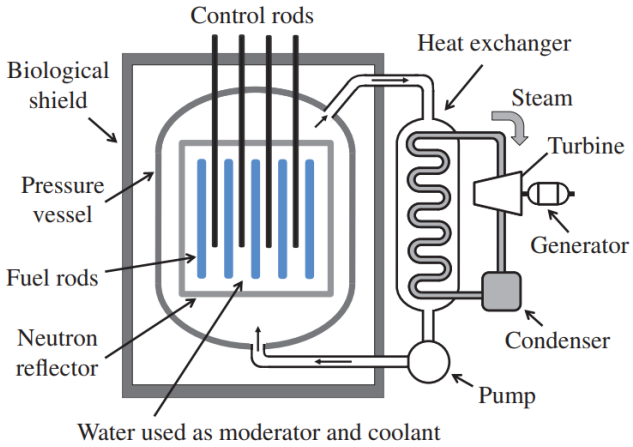

Figure 1.4: *Detailed schematic of a Pressurized Water Reactor (PWR) core and primary circuit. Key components such as fuel rods, control rods, moderator/coolant, and the surrounding pressure vessel and biological shield are highlighted.* [7]

In the operation of a reactor, certain fission fragments are produced that have a large cross-section for the absorption of thermal neutrons. A particular example is ^{135}Xe, which has an absorption cross-section of 2.7×10^6 b. These reactor poisons absorb neutrons, causing the neutron reproduction factor, k_{eff}, to decrease. This loss of neutrons is compensated by withdrawal of the control rods to restore equilibrium at $k_{\text{eff}} = 1$.

1.2.3 Reactor control: delayed neutrons and Temperature dependence of the reactivity

Delayed neutrons are the key of reactivity control. To understand the reason, analyzing how the reactor power change in time is useful:

$$W(t) = W(0)e^{\frac{\rho t}{(1-\rho)\tau_{nf}}} \tag{1.2.4}$$

The time constant associated with the motion of control rods is, typically, measured in seconds. The time delay τ_{nf} between two neutron generations is much shorter, typically 10^{-7} s for fast reactors and 10^{-4} s for thermal reactors. Such numbers would imply a very fast evolution of the reactor, even for very small

positive reactivities. In fact, for a change of reactivity of $\Delta\rho = 0.01$ the power is multiplied by 2 after 70 neutron generations, which means less than 10 μs for fast reactors and less than 10 ms for thermal reactors. With such a fast rise in the reactor power, one might think that reactor control by control rods would be hopeless. In fact, the presence of a small fraction of delayed neutrons makes things tractable.
Delayed neutrons are associated with the beta decay of fission fragments. Indeed, after prompt fission neutron emission the residual fragments are still neutron rich. They undergo a beta decay chain. The eventually emitted neutron is said to be delayed. Delays vary between fractions of a second and several tens of seconds.
Probabilities for delayed neutron emission are of the order of or less than 1% per fission and is called β. We call T_d the mean decay time for delayed neutrons. Then, in first approximation, the time which determines the time constant of the reactor τ:

$$\tau \approx \tau_{nf}(1-\beta) + T_d\beta \tag{1.2.5}$$

which is significantly higher than τ_{nf} for standard reactors. For example, for ^{235}U, $T_d = 8.82$ s and $\beta = 0.00640$. Thus, $T_d\beta$ is $\sim 0.01 - 0.1$ s while τ_{nf} is a lot smaller, making the time constant of the reactor much larger and reactor control with rods possible.

The dependence of reactivity ρ or the multiplication factor k_{eff} on temperature is a critical parameter for the stability and inherent safety of a nuclear reactor. Several effects determine the influence of temperature on the reactivity. The most important is the variation of the number of captures in the region of the resonances.

1. **Doppler Effect:** Fertile nuclei (such as ^{238}U) are characterized by large capture resonances. The apparent width of the resonances is increased by the thermal motion of the nuclei. This enhances the parasitic capture of neutrons within these resonances, removing them from the fission process and thus providing a generally **negative** contribution to the reactivity coefficient ($\frac{d\rho}{dT} < 0$).

2. **Effect of the temperature on the neutron energy spectrum**: An increase in moderator temperature increases the average neutron energy. Since the fission cross-sections of fissile nuclei (like ^{235}U) are typically lower at higher thermal neutron energies, this effect usually contributes negatively to reactivity in standard LWRs.

3. **Density Effects (Dilatation/Void):** Temperature increases cause materials, especially liquid coolants, to expand and decrease in density. In the extreme case, boiling can occur (void effect). The impact on reactivity varies significantly with reactor type:

- In LWRs, lower water density reduces moderation effectiveness and increases neutron leakage, resulting in negative feedback;
- In RBMK reactors (graphite-moderated, water-cooled), reduced water density primarily decreases parasitic neutron absorption in the water without significantly affecting moderation (done by graphite), leading to positive feedback, which was one of the main reason of Chernobyl Incident.

The overall temperature coefficient of reactivity is the sum of these contributions and is a fundamental parameter for ensuring the safe and stable operation of a reactor.

1.3 Nuclear fuel cycle

The life cycle of nuclear fuel, called the nuclear fuel cycle, is the most important source of nuclear waste. Nuclear fuel cycle refers to all of the activities related to preparation, use, and disposal of nuclear fuel. These activities include mining, concentration and purification of uranium, uranium enrichment, fuel fabrication for nuclear reactor operation, and management of used nuclear fuel (and other nuclear wastes and byproducts). Almost all nuclear reactors use fissile isotope of uranium ^{235}U as fuel. Except CANDU and MAGNOX reactors, all other types of nuclear reactors require fuel with ^{235}U at higher concentration than the natural level. Therefore, enrichment of uranium, i.e., increasing the ^{235}U isotope content, is a necessary step as part of nuclear fuel development. Nuclear fuel cycle begins with extracting uranium from uranium mine and ends with disposition of the residual uranium and other byproduct materials. These activities are divided into three stages, which are the *front-end* and the *back-end* of the fuel cycle and reactor operations in between. The front-end process includes all acitivities needed to manufacture the nuclear fuel, which are: 1. Mining and miling to produce U_3O_8, also known as Yellocake; 2. Purification of U_3O_8 and conversion to UF_6 as a preparatory step for enrichment; 3. Enrichment of ^{235}U using the chemical form, UF_6, to the required ^{235}U concentration level; 4. Converting UF_6 to UO_2 and fabrication of nuclear fuel. Operation of nuclear reactor is to use nuclear fuel for energy generation. A key measure of this energy generation and fuel utilization is its burnup, which quantifies the amount of energy extracted per unit mass of initial fuel (typically expressed in gigawatt-days per metric ton, GWd/tHM) or as the percentage of initial heavy metal atoms that have undergone fission. The energy from nuclear fission is converted to electric energy in the plant system. Activities such as fresh fuel loading, reload core fuel management, and plant maintenance are performed as part of reactor operation. This stage eventually produces used

nuclear fuels along with large volume of various other nuclear wastes. The used nuclear fuels are typically called Spent Nuclear Fuel (SNF). These spent nuclear fuels still contain fissile materials (such as ^{235}U, ^{239}Pu) and could be recycled for further energy generation. If the remaining fissile materials in the spent fuel were to be utilized, activities necessary for recycling of spent fuel become part of the back-end fuel cycle.

The back-end process of the fuel cycle is to manage what comes out of a nuclear reactor, i.e. mostly SNF. All of the follow-on activities until these wastes are permanently disposed of belong to the back-end of the fuel cycle. These activities include on-site spent-fuel storage, spent-fuel transport, away-from-reactor storage, and final disposition. When recycling of fissile material back to reactor operation is employed, the back-end of the fuel cycle includes reprocessing of SNF, which is the main solution at the moment to manage and retrieve energy from SNF. Reprocessing includes steps of breaking spent fuel, dissolving fuel materials, separating materials to be reused, and handling of waste.

The nuclear fuel cycle can be classified as once-through/open, modified open or closed, depending on whether nuclear wastes get recycled or not. In the once-through/open cycles, spent fuel is directly disposed as a waste. The modified open cycle employs at least one re-burn of fissile materials, mostly plutonium, through reprocessing of spent fuel. Differently from the closed cycle, spent fuels are disposed as nuclear waste. In fact, closed cycle involves no spent fuel disposal as all useful fissile materials in the SNF gets reused and are repeatedly recycled for full resource utilization. Of course, a closed cylce is preferred, since it uses all, or most, of the useful energy stored in the nuclear fuel.

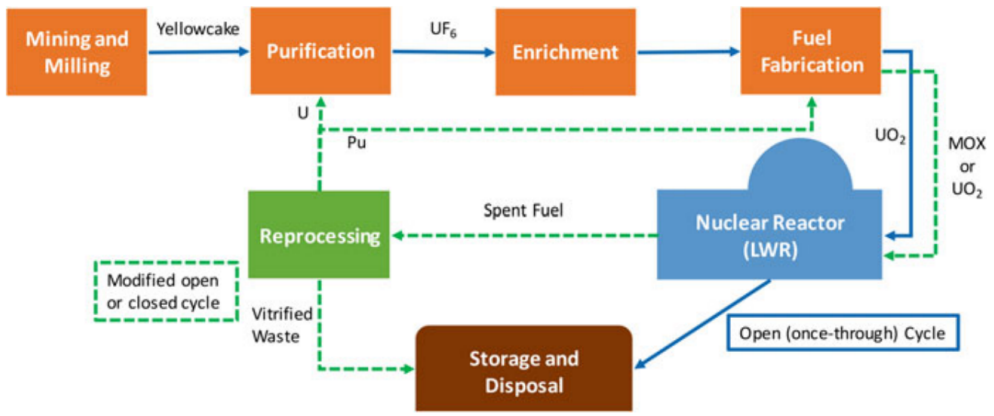

Figure 1.5: *Overview of the nuclear fuel cycle, illustrating the front-end (from mining to fuel fabrication) and back-end processes.* [8]

1.4 Composition and challenges of SNF

Due to the presence of very high levels of radiation field and heat produced by these byproducts, SNF presents extreme levels of hazard, demanding utmost care in handling.
When spent fuel comes out of a nuclear reactor, it is in the form of a fuel assembly. For example, a single PWR spent fuel assembly weighs ~ 0.59 ton. The total radioactivity contained in a single freshly discharged PWR spent fuel assembly is about 3.7×10^3 PBq. To give a perspective, note that these values exceed the total radioactivity released from the 2011 Fukushima accidents ($\sim$ 1000 PBq) in Japan but is slightly lower than the total radioactivity released from the 1986 Chernobyl accident ($\sim$ 5200 PBq). As a result of neutron irradiation, a host of irradiation byproducts are present in used fuel. These byproducts include FPs as well as activation products, which are radioactive isotopes formed when non-radioactive materials capture neutrons. The activity of SNF depends greatly on time. For example, at 1 year after discharge, the level of activity is reduced to about 1% of the freshly discharged spent fuel. The activity is further reduced to $\sim$0.2% at 10 years of cooling post discharge.

Between the fresh fuel and spent fuel, the content of ^{235}U is reduced from about 3–4% to less than 1% remaining in spent fuel. The content of ^{238}U is also reduced from about 96–97% to about 93–94% due to neutron capture. FPs are produced in replacement of the mass of the ^{235}U and ^{238}U, in the amount of about 3-5%, while plutonium and other minor actinides are also produced from the activation of uranium. The plutonium takes up about 1% of mass in spent fuel. In Figure 1.6 an example composition of SNF is shown.

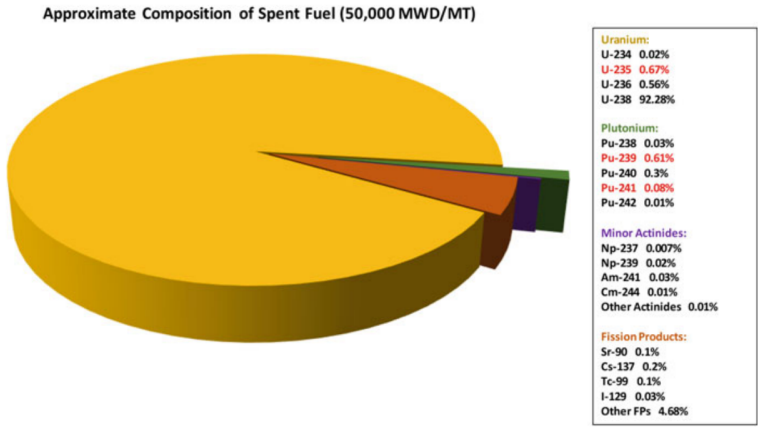

Figure 1.6: *Approximate composition of spent nuclear fuel from a PWR after achieving a burnup of 50,000 MWd/MT. While the vast majority remains uranium, the key components driving long-term radiotoxicity—plutonium, minor actinides, and fission products—constitute about 5% of the total mass.* [9]

Even though they only make up about 5% of the SNF assembly, FPs are the dominant source of radiation. As mentioned, MAs are also produced. Production of various isotopes of uranium, plutonium, and minor actinides in spent fuel comes from various nuclear reactions in the fuel. For example, when ^{235}U absorbs one neutron without being fissioned, it becomes ^{236}U. If ^{236}U subsequently absorbs another neutron, it produces ^{237}U ($t_{1/2} = 6.75$ days). ^{237}U then decays to ^{237}Np through a beta minus decay ($t_{1/2} = 2.14 \times 10^6$ years). Production of ^{237}U and ^{237}Np dominates the activity buildup of actinides, immediately after discharge of spent fuel from the reactor. The same kind of process happen for ^{238}U and ^{239}Pu, during which other minor actinides are created. It can be seen that total activity in spent fuel is dominated by fission products up to several hundred years after reactor discharge. For example, at the time of discharge, the activity is largely contributed by short-lived fission products such as ^{95}Nb, ^{95}Zr, ^{103}Rh, and ^{103}Ru. After 10 years, the important activity contributors are ^{137}Cs, ^{137}Ba, ^{90}Sr, and ^{90}Y. These same radionuclides remain important as key activity contributors after 100 years. After about 1000 years, the activity becomes dominated by the α-emitting actinides such as ^{241}Am, ^{239}Pu and ^{240}Pu. At 10,000 years, ^{239}Pu remains the important activity contributor followed by ^{240}Pu. Even at 100,000 years, ^{239}Pu remains the top activity contributor followed by ^{99}Tc. In Figure 1.7 it is shown how radioactivity of SNF changes in time, relating by their composition.

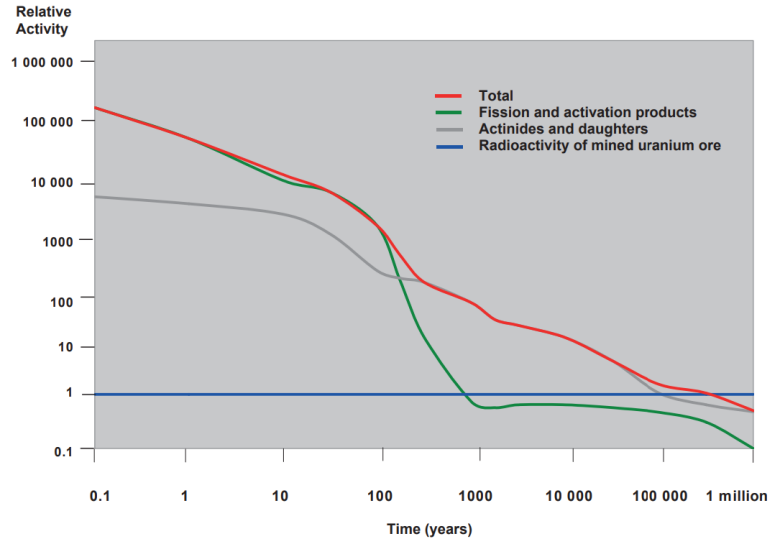

Figure 1.7: *Decay of activity of spent fuel relative to the activity of the uranium ore from which it was mined.* [10]

While the detailed composition of spent fuel varies depending on how long the fuel is irradiated in the reactor or how long it has been cooled since discharged from the reactor, the presence of plutonium in spent fuel is an important feature drawing special attention. Plutonium can be used for nuclear fuel as well as a material for nuclear explosives. One PWR spent fuel assembly contains about 4.2 kg of plutonium, which is about half the amount needed to make one nuclear explosive device. Theoretically speaking, one nuclear weapon can be made with the plutonium available in two of the PWR spent fuel assemblies. Therefore, due to this concern, diversion or theft of spent fuel should be carefully guarded to prevent the use for non-civilian purposes.
Other materials used in nuclear fuel to build fuel rods and assemblies also become a source of radioactivity through neutron activation. These materials include iron, zirconium, and other normally inert materials, including trace impurities such as nitrogen and oxygen.

Chapter 2

Management of nuclear waste

In order to address the question of management of nuclear waste, we introduce its standard classification. This classification allows the necessary handling of the wastes to be commensurate to the hazard and also helps to identify suitable disposal options. In terms of the criteria for classification, the level of potential hazard to humans is considered such that the effort made to handle each class of waste matches the level of effort needed to cope with the potential hazard. Potential hazards of nuclear waste material depend on the amount and type of radionuclides contained, the activity and half-life of the radionuclides contained, the environmental behavior of the contained radionuclides, and the chemical/physical forms of the radionuclides.

The International Atomic Energy Agency (IAEA) provides a comprehensive framework of waste classification, of which the main categories will be outlined next: [11]

- **Low Level Waste** (LLW): This waste contains long lived radionuclides at relatively low level concentration but may include higher levels of short-lived radionuclides. Typically a radionuclide with a half-life of less than about 30 years is considered short lived. It is necessary to isolate and contain this waste from the accessible environment in a robust engineered disposal system (also near surface facilities) for up to a few hundred years;
- **Intermediate Level Waste** (ILW): This type of waste contains higher concentration of long lived radionuclides compared to LLW. The waste may also contain alpha emitting long lived radionuclides. Therefore, compared to LLW, a greater degree of containment and isolation from the accessible environment is needed for the disposal of the waste. Instead of near surface facilities, a disposal system at deeper depth in the order of tens or hundreds of meters is suggested for the waste;

- **High Level Waste** (HLW): It contains large concentrations of both short and long lived radionuclides. The waste also contains significant quantities of decay heat over the periods of several centuries, so a deep geological disposal system with a greater degree of containment and isolation from the accessible environment is used for HLW disposal. The disposal system is usually several hundred meters below the surface using stable geological formations and engineered barriers. Consideration of heat dissipation through the application of temperature limits in the system is an important part of the facility design.

2.1 Strategies for the Management of SNF

The management of SNF stands as a pivotal long-term consideration within the nuclear fuel cycle, as introduced in Chapter 1. Once discharged from a reactor, SNF presents significant challenges due to its intense radioactivity, residual decay heat, and the presence of long-lived radionuclides, particularly minor actinides and certain fission products, whose hazardous nature persists over geological timescales. Consequently, robust and secure strategies are essential to ensure the safe disposition of this material. Globally, approaches to SNF management diverge, primarily following two main pathways, often preceded or accompanied by a period of interim storage.

2.1.1 Extended Interim Storage

The open fuel cycle treats SNF as a final waste product destined for direct disposal after a suitable period of cooling and interim storage. The storage of spent fuel is supposed to be at a water pool inside the fuel building next to the nuclear reactor building only for a limited period of time. These spent fuels are stored on site until they are shipped away from the reactor for storage in another facility, reprocessing, or permanent disposal. However, as of 2025, only one DGR (Deep Geological Repository) is active (Onkalo DGR, Finland), thus the spent fuel is kept at the reactor sites until a centralized interim storage facility or the final disposal facility becomes available. Therefore the originally planned short term storage of spent fuel at a reactor site turned into an extended storage facility.
This usually involves storage in water-filled pools (*wet storage*) and it referred to as a *wait and see* approach. It has its advantages, such as providing a short-medium term solution and the possibility of reusing fissile material, but also disadvantages, such as an increase in the inventory of actinides, maintaining a major hazard on the surface at the storage facility and it doesn't provide a final solution for SNF. In addition to that, it entails ongoing costs for maintenance, security, and institutional

control, along with the responsibility of knowledge transfer to future generations. In Figure 2.1 it is shown where the interim storage is located in relation to the reactor.

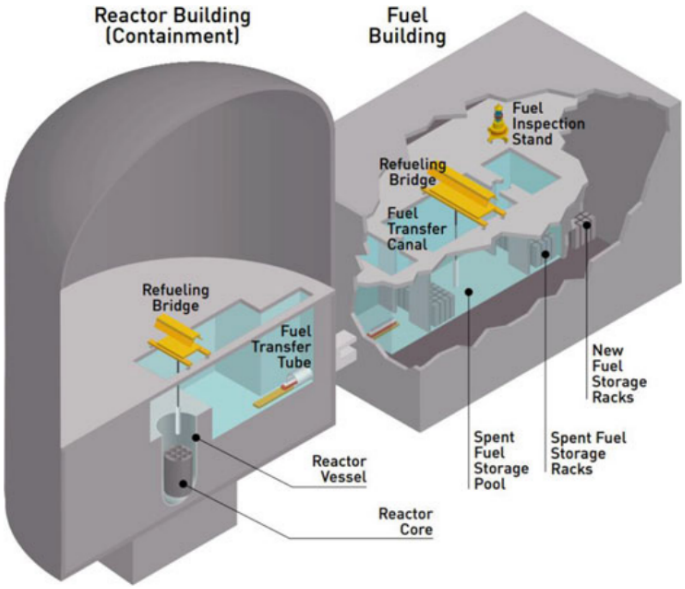

Figure 2.1: *Layout of the fuel handling and storage area in a typical PWR. The diagram shows the path of spent fuel assemblies from the reactor core within the containment building to the spent fuel pool located in the adjacent fuel building for initial cooling and interim storage.* [12]

Following the initial cooling period in spent fuel pools, typically lasting at least one to five years, *dry storage* systems provide an almost-final solution for longer-term interim storage of SNF. In dry storage, the spent fuel assemblies are removed from the pools, dried, and sealed in heavy, shielded casks or modules. These containers are typically filled with an inert gas, such as helium or nitrogen, to prevent fuel degradation and to facilitate heat transfer. Cooling is achieved through natural air convection over the exterior surfaces of the casks or modules, making these systems passively safe as they do not rely on active cooling systems or power.

Dry storage systems are designed for extended operational lifetimes, potentially for periods of 100 years or longer, although they are typically licensed for shorter initial periods (e.g., 20 to 40 years) with provisions for license renewal. This technology is widely used to manage growing inventories of SNF, to free up space in spent fuel pools, and to provide a safe and secure means of storage while awaiting final solutions, such as reprocessing or deep geological disposal.

2.1.2 Reprocessing and recycling

The strategy of closing the nuclear fuel cycle, as said previously, fundamentally relies on the chemical reprocessing of SNF to recover fissile and fertile materials, like uranium and plutonium, for subsequent recycling. The principal objectives of reprocessing are to enhance uranium resource utilisation by recycling these materials and to reduce the volume and long-term radiotoxicity of HLW requiring geological disposal.

The most established and widely deployed aqueous reprocessing technology is the PUREX (Plutonium Uranium Reduction EXtraction) process. In the PUREX process, SNF assemblies are first mechanically sheared and the fuel material is dissolved, typically in nitric acid. Subsequently, through a series of solvent extraction steps, uranium and plutonium are selectively extracted and then separated from each other and from the bulk of the FPs and MAs. These FPs and MAs, which constitute the highly radioactive liquid waste stream, are then directed towards immobilisation, predominantly through vitrification, to form a stable waste form suitable for deep geological disposal.

The recovered plutonium, typically as PuO_2, is the key product. It can be mixed with uranium oxide to fabricate MOX fuel. MOX fuel can be utilized in existing LWRs or more efficiently in fast reactors, which can achieve multiple plutonium recycling and potentially act as breeders or burners of plutonium depending on their core design. Recovered uranium can also be recycled or by conversion for use in other fuel types.

While PUREX technology is mature for U and Pu recovery, it was not originally designed for the efficient separation of MAs. These MAs significantly contribute to the long-term radiotoxicity and heat load of the HLW. Therefore, advanced reprocessing techniques are being developed. These include advanced aqueous processes and pyroprocessing. Pyroprocessing, which involves high-temperature electrochemical separations in molten salt media, is considered particularly promising for treating high burnup fuels and metallic fuels, such as those envisaged for certain types of fast reactors and ADSs.

Despite the benefits of resource recovery and waste management, reprocessing involves complex chemical plants, generates secondary waste streams (LLW and ILW), and necessitates stringent safeguards and security measures due to the handling of separated plutonium, which carries proliferation risks. The economic viability and overall environmental benefit of reprocessing also remain subjects of ongoing assessment and depend heavily on national policies, the price of natural uranium, and the specific technological pathways adopted for recycling and waste management.

2.1.3 Deep Geological Repository (DGR)

Deep Geological Repository is the internationally accepted reference strategy for the long-term management of HLW, which also includes vitrified waste from SNF reprocessing. Its fundamental goal is to isolate these highly radioactive, long-lived materials from the biosphere for periods extending up to a million years, until their radiotoxicity diminishes to levels comparable to natural uranium deposits.

The safety of a DGR relies on a passive multi-barrier system, combining engineered structures with the natural geological environment to ensure containment and isolation of radionuclides. The engineered barriers begin with the waste form itself— typically the robust UO_2 ceramic matrix of SNF or the glass for vitrified HLW. This waste form is encapsulated within highly corrosion-resistant canisters, designed for millennia of integrity. These canisters are then surrounded by a buffer material, such as compacted bentonite clay, which restricts groundwater movement, sorbs radionuclides, and provides mechanical protection and thermal dissipation.

The ultimate barrier is the natural geological formation, situated at depths typically ranging from 300 to 1000 meters. Host rocks are selected based on criteria such as geological stability, low groundwater permeability, and favourable geochemical conditions that retard radionuclide migration. For example, the Onkalo facility in Finland is based of granite.

While facilities like the Onkalo repository in Finland are coming into operation, the deployment of DGRs globally faces considerable technical, societal, and financial challenges. These persistent challenges associated with the long-term burden of HLW provide a strong drive for research into other strategies, such as Transmutation, which we explore next.

2.2 Transmutation

Transmutation is another way of HLW treatment. It is a way of rendering HLW less hazardous through conversion of the nuclide by nuclear reaction. The reaction is through bombardment of subatomic particles such as protons from a source on the target nuclide, in a process called spallation. These sources deliver neutrons which induce transmutation reactions in the energy range from thermal up to about 20 MeV. It should be noted that high-energy spallation neutrons do not significantly contribute to the transmutation reactions, but can influence the activation of components. For example, ^{129}I disintegrates into stable ^{129}Xe with a half-life of 1.57×10^7 years. If ^{129}I could absorb one neutron, it becomes ^{130}I which has a half-life of only 12.36 hours and disintegrates to a stable ^{130}Xe. Similarly,

^{99}Tc could absorb a neutron and become ^{100}Tc, which undergoes radioactive decay into a stable ruthenium within minutes. The process can be considered a way of providing aids to natural decay by effectively shortening half-lives.

Therefore, the candidate nuclides for transmutation include the actinides and LLFPs. For example, ^{99}Tc and ^{129}I are very long-lived with high environmental mobility requiring special disposal strategies for long-term isolation from the biosphere. A prerequisite step for transmutation of nuclear waste is partitioning of target nuclides: the nuclide species under consideration must first be partitioned from the waste and then recovered in a form suitable for neutron bombardment.

2.2.1 Actinide transmutation

For the transmutation of actinides the key reaction is nuclear fission which transforms long-lived and highly radiotoxic actinides into mostly short-lived, less toxic fission products. Other reactions such as capture and (n,2n): ${}^{A}_{Z}X + n \rightarrow {}^{A+1}_{Z} X^{*} \rightarrow {}^{A-1}_{Z} X + 2n$ reactions just transmute actinides into other actinides without a significant effect on long-term radiotoxicity. However, these reactions are useful as they transform fertile actinides with a low fission cross section into fissile actinides with a high fission cross section. The release of additional fission energy is a small extra gain from the transmutation of minor actinides.

The transmutation is completed when the transformation chain (shown in Figure 2.2, for ^{237}Np), which involves generations of neutron reactions and radioactive decays, ends with a fission. The first transformation step uses 0.51 neutrons on average because it is dominated by capture reactions, the second step is a simple decay while the third step produced on average 0.90 neutrons because it is dominated by fissile reactions. Until the second generation, the neutron balance in excess is +0.39 and 66% of the initial atoms are transmuted. An important parameter is the overall neutron balance, which is the neutron balance over all generations until original atoms are fissioned. It depends on the type of transmutation system. Some observations regarding this parameter:

1. Fast system (Fast Reactors or ADS) are characterized by a better overall neutron balance for the transmutation of actinides than thermal systems and provide many excess neutrons which could be utilized for transmutation of FPs;

2. The overall neutron balance of thermal system does not constrain the transmutation of typical LWR discharged transuranic mixtures;

3. A deficit in the overall neutron balance of thermal systems does not allow a complete transmutation of minor actinides.

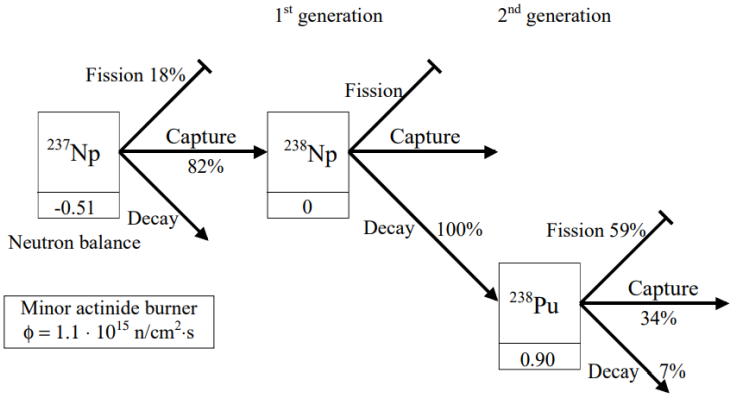

Figure 2.2: *Transformation chain for the transmutation of ^{237}Np in a minor actinide burner. The diagram shows the multi-generational process of neutron capture and decay reactions, eventually leading to fission. The neutron balance for each generation is indicated, showing an overall positive balance for this process in a fast spectrum.* [13]

Point 2 and 3 mean that, from a neutron balance perspective, TRU burners [1] can be designed as critical or sub-critical systems with any type of neutron spectrum, but dedicated minor actinides burners must be designed as critical or sub-critical fast reactors. Later it will be shown that critical and sub-critical feature similar overall neutron balances, and that the advantages of the sub-criticality are primarily a gain in core design and operation flexibility.

Since the primary mechanism for actinide transmutation (incineration) is fission reactions, the transmutation rate is obviously limited by the thermal power of the burner. Moreover, the fuel fraction which can be transmuted in a single pass of the fuel through the burner cannot exceed the fuel burn-up. For instance, solid fuels in fast reactors typically reach a burn-up limit of around 25%. This limitation means that an open fuel cycle where fuel passes through the reactor only once would be insufficient for effective, large-scale actinide reduction. Instead, a

[1] A transuranic (TRU) burner is a dedicated nuclear reactor concept designed to eliminate long-lived TRU elements (e.g., plutonium, americium, curium), which are one of the primary contributors to the long-term radiotoxicity of spent nuclear fuel. Instead of treating them as waste, a burner uses these elements as fuel, transmuting them via fission into fission products with significantly shorter half-lives.

comprehensive fuel cycle involving multiple recycles of the fuel is essential. Considering a scenario with a 25% maximum burn-up per cycle and a 6-year interval for each recycle, achieving a hundredfold reduction in the mass of actinide waste would necessitate approximately 96 years of continuous operation [14], which is a challenge for the sustainability of the technology. So, in conclusion:

- An effective transmutation system calls for a fully closed fuel cycle in which all actinides are recovered with a nearly 100% efficiency and recycled;
- To realize the potential of this system, it must be operated for an extended period of at least 100 years.

2.2.2 Fission products transmutation

For LLFPs, the goal is to transform them into shorter-lived or stable nuclides using neutron capture reactions. This process involves different challenges than actinide transmutation: FPs have to be separated individually from HLW with a high decontamination factor and processed to stable targets for irradiation, the necessity of isotopic separation and small reaction cross sections constrains the practical implementation of the process and, not least, the lack of producing net amounts of energy jeopardises the economic viability of the processes.
The traditional FP transmutation method is the irradiation of targets in a strong flux of neutrons produced by a fission reactor or a spallation neutron source. Neutron economy considerations prefers concepts where the fission product targets are irradiated in fast reactors. More recently, it has been suggested to use lead or lead-bismuth as coolant for fast-spectrum systems because the small energy loss of neutrons in collisions with the lead atoms enhances the probability for the slowing down neutrons to be captured in the resonances of the nuclide to be transmuted. Based on this principle, an accelerator-driven fission product transmuter, in which nearly every source neutron induces a transmutation reaction, is theoretically feasible.

So it appears that, at least in principle, the transmutation of LLFPs would be a useful method to mitigate the risk associated with geologic repositories. However, the practical feasibility of the required process has been established only for ^{99}Tc. In fact, the transmutation of a FP makes sense only if the reaction rate is higher than the natural decay rate of the nuclide. With the practically achievable neutron fluxes, this condition cannot be met for the most abundant ^{137}Cs,^{90} Sr with half-lives of only about 30 years. In few words: these FPs are not transmutable. Also for other FPs such as ^{135}Cs,^{129} I and ^{129}Sn, other methods have to be used (confinement in a stable matrix).

2.2.3 Implementation of transmutation

The neutrons used for transmutation can be produced in either a nuclear reactor or a sub-critical accelerator system. Also, depending on the radionuclides to be transmuted, the neutrons used can be fast or thermal, or both. Therefore, both a thermal reactor and a fast reactor present opportunities for transmutation.

Thermal reactors

As mentioned before ^{99}Tc and ^{129}I can be transmuted to stable isotopes by the neutron capture reaction. The capture cross-sections for these nuclides are extremely small for fast neutrons but reach the level of a few barns for ^{129}I and $\sim$ 14 b for ^{99}Tc. Although these values may not be large enough for efficient transmutation, use of a high neutron flux, i.e., increasing the number of neutrons, can make transmutation of these radionuclides feasible. Therefore, indirect transmutation through neutron capture can be pursued for long-lived actinides, such as ^{237}Np, ^{241}Am and ^{243}Am. Through capture, these actinides will form heavier nuclides, such as ^{238}Pu, ^{242}Am, ^{243}Cm, ^{239}Pu and ^{245}Cm. These new nuclides have sizable fission cross-sections and thus can be transmuted through fission.

Loading of minor actinides with high neutron capture cross-section into a reactor core for the purpose of transmutation will result in a decrease in the overall reactivity. In this case, the reactor core needs to contain higher fissile enrichment of the fuel to compensate for reactivity loss. During the life of the reactor core, the reactivity may also increase with the formation of fissile nuclides from radiative capture of actinides. This indicates the need for elaborate reactivity management during the core operating cycle. This option is not preferred.

Fast reactors

While all actinides can be fissioned with fast neutrons, the overall reaction cross sections (for both fission and capture) for fast neutrons are smaller than those for thermal neutrons. This lower cross section effect is compensated by having high neutron flux in a fast reactor which is 100–1000 times higher than thermal reactors. This is shown in Figure 2.3 for the case of a fast breeder reactor as an example.

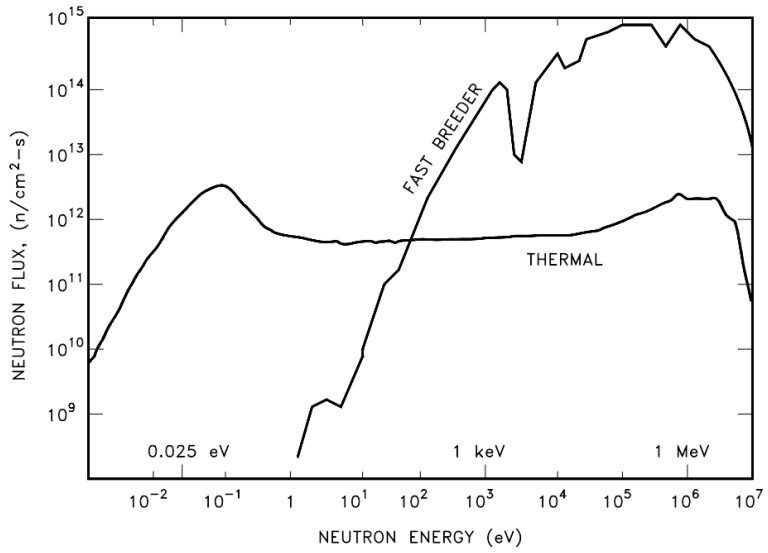

Figure 2.3: *Comparison of typical neutron flux spectra in a thermal reactor and a fast breeder reactor. The fast reactor operates with a much higher average neutron energy and a significantly higher total neutron flux (by 2-3 orders of magnitude), which is crucial for the effective transmutation of actinides.* [15]

Because of the differences in the cross sections, the build-up of actinides by neutron capture is much smaller in a fast reactor than in a thermal reactor. Also, with fast neutrons, the fission-to-capture ratio of actinides is higher in a fast reactor. Therefore transmutation of actinides is more effective in fast reactors. LLFPs, like ^{99}Tc and ^{129}I, can also be transmuted in fast reactors by the use of moderators in the system, thus by creating thermal neutrons through fast neutron moderation.

Accelerator Driven System

When a very high energy ($\sim$ 1 GeV) proton hits a target such as lead or tungsten, a spallation reaction takes place, disintegrating the nucleus of the target atom and releasing large number of neutrons. Therefore, use of a high-energy proton accelerator with a spallation target offers capability for transmutation using the resulting neutrons. The generated neutron from the spallation target reactions in the reactor core which contains the radionuclides to be transmuted. The heat from fission reactions is converted to generate electricity through the heat transfer using a coolant for steam production. Part of the generated electricity goes back to the system to support the operation of the accelerator. In the following Chapter this possibility will be explored in detail.

Chapter 3

Accelerator - Driven Systems

3.1 Concept of an ADS

The concept of an Accelerator-Driven System, also known as hybrid system, combines a particle accelerator with a subcritical fission reactor. Most proposals assume linear or circular proton accelerators, delivering continuous-wave beams with an energy around 1 GeV. A spallation target is hit by the protons in order to produce source neutrons for driving the subcritical core. The target is made of heavy metal in solid or liquid state. Spallation reactions in the target emit a few tens of neutrons per incident proton, which are introduced into the sub-critical core to induce further nuclear reactions. Except for the sub-critical state, the core is very similar to that of a critical reactor. It can be designed to operate either with a thermal or fast neutron spectrum.

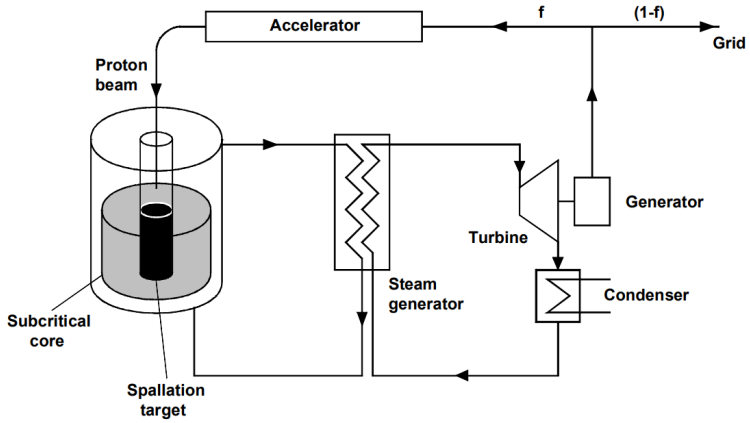

Figure 3.1: *Conceptual schematic of an Accelerator-Driven System (ADS). It shows the three core components: a high-power particle accelerator, a spallation target for neutron production, and a subcritical core that multiplies the source neutrons to sustain fission and generate energy.* [16]

In Figure 3.1 the concept of an ADS is shown.

When compared with critical reactors, ADSs have two specific characteristics:

- Given their design, they prevent any incident regarding criticality. In addition to that, because of their very small delayed neutron fractions, incineration of minor actinides appears to be feasible only with subcritical reactors;
- Subcriticality provides additional neutrons which can be used for increased breeding of ^{233}U and ^{239}Pu. It is even possible to breed them in the absence of any fissile element. Another possible use of the additional neutrons is to transmute long-lived fission products.

3.1.1 Safety advantages of ADSs

In principle, criticality accidents such as that of Chernobyl should be impossible for an ADS. However, this is true only as long as one can monitor the effective value of the reactivity. A important factor to keep in mind is that it requires more technical skill and good maintenance to keep them running than for critical reactors. Indeed, high-intensity accelerators are and will remain rather difficult to operate. Loss of expertise of the staff as well as poor maintenance will, inevitably, decrease the performance of the machine. In contrast, critical reactors are apt to run in rather bad shape and do not necessarily need the best team to be operated, with the dangers associated with such a situation.
Aside from criticality incidents, ADSs face the same risks associated with critical

reactors (core meltdown, radioactive leaks...).
For large subcriticality levels of more than a few per cent, the delayed neutron fraction has no influence on the safety of the reactor. This means that it becomes possible to use fuels with large minor actinide concentrations or plutonium without compensating for the small delayed fraction by the presence of ^{238}U. Similarly, the sign of the temperature and void coefficients have a reduced influence. However, they should be limited so that subcriticality should be guaranteed at all power levels of the reactor. In particular, overly negative coefficients should be avoided to prevent criticality in the case when an accelerator trip leads to a sharp fall of the reactor power. The high tolerance level of ADSs with respect to the fuel's neutronic properties should make them excellent tools to study new reactor concepts by relaxing many safety conditions. For example, the same accelerator could feed different subcritical systems like molten salt, gas or lead cooled reactors.

3.1.2 Additional neutrons

As will be shown later, the additional number of neutrons provided by an ADS is the number of initial spallation neutrons N_0 whatever the value of the multiplication coefficient k_s. It is then possible to estimate the additional transmutation capabilities of an accelerator. Typically an accelerator can provide 1.5×10^{25} neutrons/year/mA. Thus if all neutrons are captured in the nuclei to be transmuted, one gets an annual transmutation rate of 25 moles/year/mA. For example, the production rate of ^{233}U would be around 5 kg/year/mA. One sees that a 20 mA accelerator, as typically considered for ADSs, would produce 100 kg/year of ^{233}U, in addition to the production of the subcritical system, which in the best case amounts to 50 kg/GWe/year. Thus ADSs can increase quite significantly the ^{233}U breeding capabilities. Similarly around 2.5 kg of long-lived fission products could be transmuted each year per mA proton beam. Since a typical 1 GWe reactor produces each year 4 kg of ^{129}I, for example, a 20 mA accelerator could transmute annually the production of more than ten reactors.

However, important design and material challenges arise from the installation of the spallation target in the centre of a reactor: the interfacing of an accelerator with a reactor rises containment questions, and the target and surrounding structure materials are subjected to a complex degradation due to combined thermo-mechanical loads, high-energy particle irradiation and, in contact with liquid heavy metals, corrosion effects which are much more severe than those encountered in normal reactors.

3.2 Components of an ADS

3.2.1 Accelerator

In most hybrid reactor concepts, the external neutrons are provided by the interaction of accelerated charged particles with matter. The most widely proposed systems use high-energy protons. To accelerate a high intensity proton beam to an energy of the order of one GeV, two completely different accelerator schemes are possible, a linac or a cyclotron. The choice depends on many factors, but it is important to clarify from the beginning that, to fulfil the beam requirements for ADS applications (specifically a very low frequency of beam interrupts), both machine designs have to be modified and developed, to extend into a new dimension of complexity, cost, and size. The operating mode for ADS will most likely be CW (continuous). With a desired total thermal power of the ADS around 1 GWth, one sees that beam powers around 10 MW are needed (given an energy gain, the details of which will be later explored in detail, of 100). The requirements for a high-proton-intensity accelerator connected to an ADS can be summarized as follows:

- Proton energy should be higher than 600 MeV;
- Beam power should exceed 10 MW, which means that currents over 10 mA are needed;
- Beam losses should be made very small in order to minimize accelerator structure activation;
- High beam availability is required. This is obvious for long-lasting shutdowns due to equipment breakdown: this requirement is analogous to those for power reactors. Another event specific to accelerators is the occurrence of short-duration trips where the beam is lost. If the duration of a trip exceeds the time it takes for the subcritical system to reach thermal equilibrium after an input power variation the ADSR structures will be submitted to thermal stress and, thus, increased fatigue;
- The energy efficiency of the accelerator complex should be reasonably high.

One can see that these requirements are not easily met.

3.2.2 Spallation target

The most peculiar component of an ADS is the spallation target, which is designed to generate the maximum amount of neutrons while ensuring the removal of the

heat released in the spallation process. As the proton beam power being deposited in such a target attains several MW, even up to about 20 MW, very high power densities of several hundreds of kW per litre, occurring in the structure and in the spallation material, need to be safely removed. Heavy metals such as tungsten, tantalum, uranium, lead-bismuth, lead, mercury are considered as possible spallation materials for targets. Gas, heavy water or liquid metals are under consideration as coolants for these targets. The physical process of spallation will be explored later.
The target operates in a harsh mixed proton-neutron irradiation field, posing challenges for its design and material selection. Heavy metals like tungsten, tantalum, uranium, lead, lead-bismuth eutectic (LBE), and mercury are considered as spallation materials, with cooling options including gas, heavy water, or liquid metals. There are two main target options:

- **Solid Targets:** Commonly used in existing, lower-power, often pulsed spallation sources, typically made of W, U, or Ta cooled by heavy water. However, due to very high power densities in ADS applications, solid metal targets are generally excluded. Key challenges include radiation damage (swelling, reduced thermal conductivity), complex cooling, radiolysis/activation of water coolant, and decay heat removal after shutdown;

- **Liquid Metal Targets:** Preferred for ADS due to superior inherent cooling, higher tolerable power densities, and reduced irradiation damage to the target material itself. Lead and LBE are primary candidates. Mercury has also been proposed.

Liquid metal targets face several key challenges. Ensuring robust containment and the integrity of the beam window is critical, as this interface endures intense radiation, high temperatures (potentially up to 650°C), and corrosion, necessitating careful material selection and likely frequent replacement. Corrosion and erosion of structural materials by the hot, high-velocity liquid metal are persistent issues, with oxygen control in Pb/LBE systems being a vital mitigation strategy. The accumulation of spallation products can alter coolant chemistry, damage materials, and create radiological hazards. Furthermore, the mixed proton-neutron radiation field degrades the mechanical properties of the window and structures, an area where data for ADS-specific conditions remain scarce. Finally, beam trips pose a significant operational risk, causing thermal shocks and pressure waves that can lead to material fatigue and potential coolant solidification if not managed by emergency shutdown systems and careful thermal design.

3.2.3 Subcritical reactor

The main component of an ADS is the subcritical reactor. Its fundamental characteristic is of course that it's operating in a subcritical state, meaning its effective neutron multiplication factor (k_{eff}) is consistently maintained below 1. This condition implies that a self-sustaining nuclear fission chain reaction is not possible; the reactor relies on an external neutron source, provided by the spallation target, to maintain a steady fission rate.

Operating in subcriticality has significant implications for the system's safety and control. In principle, it eliminates the possibility of uncontrolled critical excursions typical of critical reactors and allows for greater flexibility in fuel composition, enabling the use of materials that would degrade the safety parameters of a critical core. The reactor's power control is directly linked to the intensity of the external neutron source, i.e., the accelerator beam current.

The core composition of an ADS reactor is designed according to specific objectives, such as waste transmutation or energy production:

- **Fuel:** various fuel types can be used, including MOX, but particularly innovative fuels designed for transmutation, such as those containing MAs or fuels dispersed in inert matrices. The choice depends on the adopted fuel cycle strategy;
- **Moderator:** its presence or absence defines the neutron spectrum. In fast-spectrum ADS, the moderator is absent or minimized to maximize the transmutation efficiency of actinides. In some thermal spectrum concepts, a moderator may be present for the transmutation of specific fission products;
- **Coolant:** it's responsible for efficient heat removal. Considered materials include heavy liquid metals like lead or LBE, gases (helium), or sodium, depending on the specific design and neutron spectrum.

The neutron spectrum is a crucial parameter:

- **Fast-spectrum** ADS are generally preferred for the efficient transmutation of minor actinides, due to their higher fission cross-sections in this energy range;
- **Thermal-spectrum** ADS have been proposed for the transmutation of specific long-lived fission products or for particular fuel cycles, such as the thorium cycle.

The external neutrons from the spallation target are essential to control the subcritical reactor. They compensate for the neutrons lost (by absorption or leakage) which are insufficient to maintain $k_{eff} \geq 1$, thus sustaining a significant

number of fissions in the core. The subcritical system acts as a multiplier for these source neutrons, leading to an energy gain.

3.3 Physics and neutronics of ADSs

In order to understand the physics of subcritical reactor, we start from the more general form of the Bolztmann equation, which express the variation in time of the number of neutrons present in an elementary volume V of surface S, to obtain the neutron balance equation:

$$\frac{d}{dt}\iiint n(\mathbf{r},v,t)d^3r = ((\text{entering - exiting}) + (\text{created - absorbed}) + (\text{inscattered - outscattered})) \text{ neutrons}, \tag{3.3.1}$$

where $n(\mathbf{r},v,t)$ is the neutronic density.
Analyzing in detail each term of the right-hand side of (3.3.1):

$$\text{entering - exiting} = -\int\limits_S\int \mathbf{J}(\mathbf{r},v,t)dS = -\int\limits_V\int \text{div } \mathbf{J}(\mathbf{r},v,t)d^3r \tag{3.3.2}$$

is the total net flow of neutrons entering the volume across the boundary surface V,

$$\text{created} = \int\limits_V\int\left[S(\mathbf{r},v,t) + \sum_i \nu_i\psi_f(v)\left(\int \varphi(\mathbf{r},v',t)\Sigma_f^{(i)}(\mathbf{r},v')dv'\right)\right]d^3r, \tag{3.3.3}$$

is the total neutrons generated inside the volume, both from the external source $S(\mathbf{r},v,t)$ and from the fission reaction. ν_i is the average number of neutrons emitted per fission, $\psi_f(v)$ is the velocity spectrum of the neutrons created in the fission, $\Sigma_f^{(i)}(\mathbf{r},v')$ is the macroscopic fission cross section per atom i and $\varphi(\mathbf{r},v',t)$ is the neutronic flux,

$$\int\limits_V\int\sum_j\left(\int \varphi(\mathbf{r},v',t)\Sigma_s^{(j)}(\mathbf{r},v'\to v)dv'\right)d^3r \tag{3.3.4}$$

is the rate at which neutrons enter the velocity group v as a result of being scattered from other velocities v'. $\Sigma_s^{(j)}(\mathbf{r},v'\to v)$ is the differential macroscopic scattering cross-section for nucleus j. It represents the probability that a neutron with initial velocity v' will have a final velocity v after scattering.

$$\text{outscattered + absorbed} = \int\limits_V\int \varphi(\mathbf{r},v,t)\sum_j \Sigma_T^{(j)}(\mathbf{r},v)d^3r \tag{3.3.5}$$

is the the total rate of removal of neutrons from the velocity group v due to all interactions (absorption and scattering). $\Sigma_T^{(j)}(\mathbf{r}, v)$ is the total macroscopic cross-section for nucleus j, defined as $\Sigma_T = \Sigma_s + \Sigma_a$. Therefore, we obtain the differential Boltzmann equation:

$$\begin{aligned}\frac{\partial\varphi(\boldsymbol{r}, v, t)}{v\partial t} =& -\operatorname{div}(\boldsymbol{J}(\boldsymbol{r}, v, t)) + S(\boldsymbol{r}, v, t) \\ &+ \int \varphi\left(\boldsymbol{r}, v', t\right) \sum_{i,j} \nu_i \psi_{\mathrm{f}}(v) \left(\Sigma_{\mathrm{f}}^{(i)}\left(\boldsymbol{r}, v'\right) + \Sigma_{\mathrm{s}}^{(j)}\left(\boldsymbol{r}, v' \rightarrow v\right)\right) \mathrm{d}v' \\ &- \varphi(\boldsymbol{r}, v, t) \sum_j \Sigma_{\mathrm{T}}^{(j)}(\boldsymbol{r}, v),\end{aligned} \tag{3.3.6}$$

where we used the relation $\varphi(\mathbf{r}, v, t) = vn(\mathbf{r}, v, t)$.
Then, making use of Fick's law, which relates the current $\mathbf{J}(\mathbf{r}, v, t)$ to the flux $\varphi(\mathbf{r}, v, t)$:

$$\mathbf{J}(\mathbf{r}, v, t) = -D\vec{\nabla}\varphi(\mathbf{r}, v, t), \tag{3.3.7}$$

where D is the diffusion coefficient, which can be expressed as $D = \frac{\Sigma_s}{3\Sigma_T^2}$. Finally, we obtain the neutron balance equation, assuming neutrons to be monoenergetic and dropping the integration over velocities in (3.3.6):

$$\frac{1}{v}\frac{\partial\varphi(\mathbf{r}, t)}{\partial t} = D\nabla^2\varphi(\mathbf{r}, t) + \varphi(\mathbf{r}, t)\sum_j \Sigma_a^{(j)}(\mathbf{r})(k_\infty - 1) + S(\mathbf{r}, t). \tag{3.3.8}$$

with:

- $\frac{1}{v}\frac{\partial\varphi(\mathbf{r},t)}{\partial t}$ is the neutron flux time-varying term, where $\varphi(\mathbf{r}, t)$ is the neutron flux and is defined as $\varphi(\mathbf{r}, t) = nv$, n is the neutron density and v is the velocity of neutrons. Thus, this term represents the variation in time of neutron density in an infinitesimal volume near $\mathbf{r}$.
- $D\nabla^2\varphi(\mathbf{r}, t)$ is the diffusion term, with D as the diffusion coefficient;
- $\varphi(\mathbf{r}, t)\sum_j \Sigma_a^{(j)}(\mathbf{r})(k_\infty - 1)$ is the neutron net production from fission and absorption, where $\sum_j \Sigma_a^{(j)}(\mathbf{r})$ is the macroscopic absorption cross section in $\mathbf{r}$ so $\varphi(\mathbf{r}, t)\sum_j \Sigma_a^{(j)}(\mathbf{r})$ is the absorption rate of neutrons per unit of volume. k_∞ is the already defined moltiplication factor;
- $S(\mathbf{r}, t)$ is the external neutron source.

In an infinite and homogeneous medium, with an evenly distributed neutron source, the neutron flux should be independent of $\mathbf{r}$, therefore the neutron balance equation

becomes:

$$\frac{1}{v}\frac{\partial\varphi(t)}{\partial t} = \varphi(t)\sum_j \Sigma_a^{(j)}(k_\infty - 1) + S(t). \tag{3.3.9}$$

Firstly, we consider the case of a system without external source. Therefore, per t>0, we have $S(t) = 0$ and $\varphi(0)$ is finite. The solution of the equation (3.3.9) reads:

$$\varphi(t) = \varphi(0)e^{v(k_\infty - 1)t\sum_j \Sigma_a^{(j)}} \tag{3.3.10}$$

which shows that if $k_\infty > 1$ the flux diverges, while it decreases to 0 for $k_\infty < 1$. It is time independent only if $k_\infty = 1$. This condition can never be met in reality. Rather, in critical reactors, a time-dependence of the absorption cross-sections is used, so that k_∞ fluctuates about 1.
We now consider the case of the ADSs, where $k_\infty < 1$ and $S(t) = S_0$ is time independent, but positive. The solution of (3.3.9) for stationary states is

$$\varphi = \frac{S_0}{(1 - k_\infty)\sum_j \Sigma_A^{(j)}} \tag{3.3.11}$$

and the number of absorption reactions per second is then

$$n_{reac} = \frac{S_0}{1 - k_\infty} \tag{3.3.12}$$

For a complete treatment of the neutron propagation problem, more can be found in [17].

3.3.1 Energy gain

Before introducing the energy gain definition, we have to make a distinction between k_{eff} and k_s. While k_{eff} is an intrinsic property of the system which measures the effective neutron multiplication capacity of the medium (in the absence of an external source), k_s measures the effective multiplication of neutrons from the specific source, taking into account spatial and energy distribution.

More formally, (3.3.9) can be written in the matrix form: $A\emptyset_{in} = P\emptyset_{in} + S_{in}$ (where A is the loss operator, P the fission production operator, and S_{in} the external source term), k_s can be derived from the formal balance condition $A\emptyset_{in} = \frac{1}{k_S}P\emptyset_{in}$. This leads to an explicit definition for k_s based on integrated reaction rates:

$$k_s = \frac{\langle P\emptyset_{in}\rangle}{\langle P\emptyset_{in}\rangle + \langle S_{in}\rangle} \tag{3.3.13}$$

where $\langle P\emptyset_{in}\rangle$ is the total rate of neutrons produced by fission and $\langle S_{in}\rangle$ is the total rate of neutrons supplied by the external source. k_s thus represents the fraction

of all neutrons appearing in the system that originate from fission. Consequently, $(1 - k_s)$ is the fraction originating directly from the source. It is k_s that directly determines the extent of source neutron multiplication and, therefore, the energy gain of the ADS.

We have seen that, if $k_{eff} < 1$, the chain reaction cannot be sustained. However, if a source of neutrons is introduced inside the multiplying medium, the initial neutron number is multiplied by a factor which can be very large. Since neutrons are associated with an equivalent number of fissions, a large energy could be produced with a subcritical system, provided a neutron source is available. If N_0 is the number of primary neutrons following, for example, the interaction of a proton with a target surrounded by a multiplying medium chacterized by a multiplication factor k_s,the total number of created neutrons, after multiplication, is

$$\frac{N_0}{1 - k_s} \tag{3.3.14}$$

and the number of secondary neutrons (produced after at least one multiplication) is

$$\frac{N_0 k_s}{1 - k_s} \tag{3.3.15}$$

Each of these neutrons is produced by a fission which itself produces ν neutrons. Thus, the number of secondary fissions in the system is $\frac{k_s N_0}{\nu(1-k_s)}$. Since each fission releases about $\epsilon_f = 0.18$ GeV energy (see Table 1.1, considering only kinetic energy) the thermal energy produced in the medium will be $\epsilon_f \frac{k_s N_0}{\nu(1-k_s)}$. We then define the energy gain of the system:

$$G = \epsilon_f \frac{k_s N_0}{\nu(1 - k_s) E_p} = G_0 \frac{k_s}{1 - k_s} \tag{3.3.16}$$

where E_p is the energy of the incident protons, ϵ_f is the energy per fission and $G_0 = \epsilon_f \frac{N_0}{\nu E_p}$. For example, according to the CERN FEAT experiment, $G_0 k_s = 3.1$ for proton energies larger than 1 GeV and for an uranium target [18].

The proton beam is produced with a finite efficiency which is the product of the thermodynamic efficiency for producing electricity from heat (typically 40%) by the acceleration efficiency. For high intensity accelerators, most of the power is used for the high-frequency cavities, at variance with low-intensity accelerators where most of the power is spent in the magnetic devices. High intensities, therefore, are expected to allow ∼40 % efficiencies. Finally, the total efficiency for proton acceleration is expected to be near 0.16. This leads to a minimum value of the multiplication factor for obtaining a positive energy production (ignition), $k_m = 0.69$. For a value $k_s = 0.98$ a net energy gain of 16 is achieved.

Neutron source importance

We introduce the concept of the importance of the neutron source. Understanding the behavior of a source-driven subcritical core depends largely on assessing the relative importance of source neutrons versus fission neutrons generated in the subcritical core.

The parameter ϕ^* is introduced as the ratio between the importance (intended as the ability to induce fissions in subsequent generations) of the source neutrons and the average importance of the fission neutrons. In simpler words, it quantifies how much the neutrons coming from the external source are effective in contributing to the neutron population (therefore the reactor power), compared to the neutrons generated internally by fissions.

It can be shown that this parameter ϕ^* is related to k_{eff} as

$$\phi^* = \frac{\frac{1}{k_{eff}} - 1}{\frac{\Gamma}{\nu}} \tag{3.3.17}$$

where Γ is the average number of source neutrons per fission. So, this equation shows how ϕ^* is related both the system sub-criticality and the fundamental neutronics characteristics of the fission process and of the production of neutrons from the source.

The ϕ^* parameter plays an important role in assessing the ADS performance parameters, as it basically takes the role of *linking* the particle accelerator to the reactor. In fact it is shown that it determines the requirements for the proton beam current [19]:

$$i_p = \frac{\nu W}{\phi^* Z \epsilon_f} \left(\frac{1 - k_{eff}}{k_{eff}} \right) \tag{3.3.18}$$

where i_p is the proton beam current, W is the power of the subcritical core and Z is the is the number of neutrons per incident proton.

In conclusion, a value $\phi^* > 1$ (i.e. the source neutrons are more "important" than the fission neutrons) can reduce proportionally the proton current beam required for a given level of subcriticality and for a given power of the subcritical core.

3.3.2 Delayed Neutrons and Power Control

As mentioned in Section 1.2.3, delayed neutrons play a significant role in thermal reactor. While they still maintain their importance, in ADS delayed neutrons don't play a fundamental role.

The equations which give the kinetic behaviour of a system driven by an external source are of the type:

$$\begin{cases} \dfrac{dW(t)}{dt} = \dfrac{\rho - \beta}{\tau} W(t) + \displaystyle\sum_i \lambda_i C_i + S(t) \\ \dfrac{dC_i}{dt} = \dfrac{\beta_i}{\tau} W(t) - \lambda_i C_i \end{cases} \tag{3.3.19}$$

where:

- $W(t)$ is the reactor power;
- $\beta_{eff} = \sum \beta_i$ is the total effective delayed neutron fraction;
- β_i is the delayed neutron fraction for the i-th group of precursors;
- λ_i is the decay constant for the i-th group of precursors;
- $C_i(t)$ is the concentration of the i-th group of delayed neutron precursors;
- $S(t)$ is the intensity of the external neutron source (neutrons/second);
- τ is the prompt neutron lifetime.

In steady state, i.e. $\frac{dW}{dt} = 0$ and $\frac{dC_i}{dt} = 0$, we have:

$$W = -\frac{\tau}{\rho} S \tag{3.3.20}$$

A decrease by a factor h of the reactivity or an increase by a factor h of the source induces an instantaneous increase in the power $W' = hW$. For example, if, the system is subcritical corresponding to -10β, a reactivity insertion of +5β causes a doubling of the power. This of course is totally different from the behaviour of a critical system, which becomes prompt critical, as can be clearly seen in Figure 3.2.

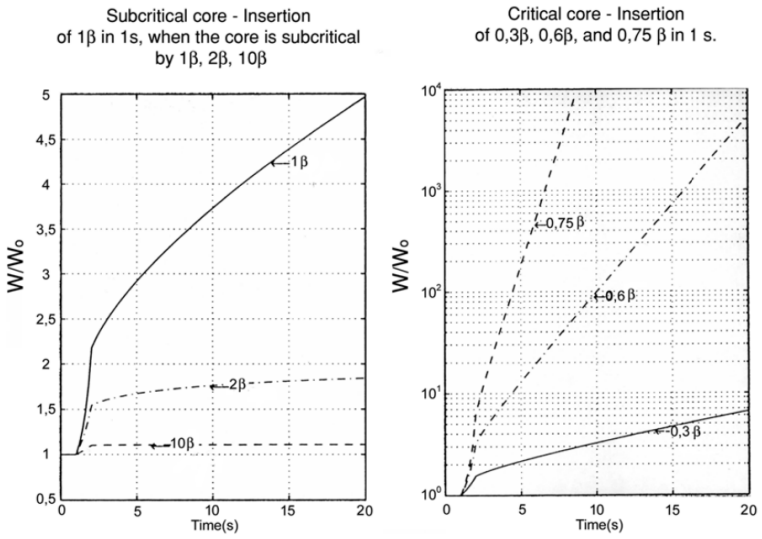

Figure 3.2: *Comparison of the kinetic behavior of power (W/W_0) in a sub-critical (left) and critical (right) core following a reactivity insertion. In the sub-critical system, the power increase is limited and stable, whereas in the critical system it leads to a rapid divergence.* [20]

In more general terms, the kinetic behaviour of a critical system is characterized by delayed neutrons and their time constants, while the kinetic behaviour of a subcritical core is determined by the time constants related to the external source, in the sense that an instantaneous variation of source has an effect on the time scale of the prompt neutron lifetime (typically of the order of microseconds).

The evolution of the power with time, and the related variation of the temperature, are related to the variation of the reactivity (effect such as those mentioned in Section 1.2.3). These feed-back reactivity effects are essential for the safety of a critical reactor. In a sub-critical core, the relevance of feedback reactivity effects varies according to the level of sub-criticality. In fact for a deeply sub-critical core, the dynamic behaviour is dominated by the external source and its variation in time. Closer to criticality, the feedback effects become more important and the behaviour of the core is approaching that of the corresponding critical core.

The response of an ADS to perturbations is characterized by two main time scales:

1. A **prompt response**, governed by the prompt neutron lifetime τ. Variations in the source intensity S_{ext} cause almost instantaneous changes in the prompt neutron population and thus in power;

2. A **slower response**, associated with the decay of delayed neutron precursors (with time constants $1/\lambda_i$, ranging from fractions of a second to about a

minute). This component becomes more relevant when considering long-term reactivity changes or when the system operates very close to criticality ($k_{eff} \approx 1$).

However, for most operations and rapid transients in a typically subcritical ADS (e.g., k_{eff} between 0.95 and 0.98), the dynamics are dominated by the source and the prompt neutron population.

3.3.3 Neutronic balance and breeding capacity

Aside from energy production, it is important to evaluate the potential of hybrid reactors for transmutation, i.e. to what extent they produce excess neutrons. A standard reactor can be viewed as a device producing energy and neutrons. Both energy and neutrons are primarily produced by fission, which releases about 200 MeV and 2.5 neutrons. It follows that one may say that 80 MeV are needed to produce one neutron. The spallation process requires only 30 MeV to produce one neutron. If the released energy in a fission were available for proton acceleration one would, then, get more than nine neutrons per fission (2.5 + 6.6), but of course no usable energy would be produced. More realistically, considering a thermodynamic efficiency of 40% and an accelerating efficiency of 40%, one finds that about 6 GeV are needed to accelerate a proton to 1 GeV. It would then be possible to obtain about 3.5 neutrons per fission, still without producing usable energy. For more realistic scenarios one sees that an accelerator allows an increase of the number of neutrons available for transmutation at the expense of usable energy. It is interesting to see if, as far as neutron availability is concerned, hybrid reactors are more or less efficient than the association of a critical reactor and an accelerator.

The number of neutrons produced in hybrid reactor is

$$N = \frac{N_0}{1 - k_s} \tag{3.3.21}$$

and the number of fissions is

$$N_F = \frac{N_0 k_s}{\nu(1 - k_s)} \tag{3.3.22}$$

On average a fission is produced by $\frac{\sigma_f + \sigma_c}{\sigma_f}$ neutrons. The total number of neutrons needed to produce N_F fissions is

$$N_{nf} = N_F \frac{\sigma_f + \sigma_c}{\sigma_f} = N_F(1 + \alpha) = \frac{N_0 k_s}{\eta(1 - k_s)} \tag{3.3.23}$$

where $\eta = \frac{\nu}{1+\alpha}$ has been used. Therefore, the total number of neutrons available for transmutation is, logically,

$$N_{Dhyb} = N - N_{nf} = \frac{N_0}{1-k_s}\left(1 - \frac{k_s}{\eta}\right) \tag{3.3.24}$$

We now consider a critical reactor coupled to an accelerator. N_{Dr} is the number of neutrons available when using a reactor producing N_F fissions, in addition to the N_0 spallation neutrons. The number of neutrons necessary for fission is

$$\frac{\sigma_f + \sigma_c}{\sigma_f} = (1+\alpha) \tag{3.3.25}$$

Therefore, the number of neutrons available per fission is $\nu - 1 - \alpha$. The total number of neutrons available in the reactor is then

$$N_{Df} = N_F(\nu - 1 - \alpha) = \frac{N_0 k_s}{\nu(1-k_s)}(\nu - 1 - \alpha) \tag{3.3.26}$$

and the total number of neutrons available for the reactor + accelerator is

$$N_{Dr} = N_0\left(1 + \frac{k_s}{\nu(1-k_s)(\nu-1-\alpha)}\right) = \frac{N_0}{1-k_s}\left(1 - \frac{k_s}{\eta}\right) \tag{3.3.27}$$

Clearly,

$$N_{Dhyb} = N_{Dr} \tag{3.3.28}$$

It follows that the choice of a specific value of k is irrelevant as far as the transmutation capabilities are concerned. Whatever the method of coupling between the fission reactor and the accelerator, the number of available neutrons is

$$N_D = N_0 + N_F(\nu - 1 - \alpha) \tag{3.3.29}$$

which easily states that the number of neutrons available for transmutation are the ones produced by the source plus the neutrons excess generated by the N_F fissions.

We now consider the breeding possibility of various fuels using an ADS. From the preceding, it is seen that using 10% of the available energy allows us to obtain about 0.1 additional neutrons per fission. Although small, this number has to be compared with the number of neutrons which are effectively available in reactors. However, in practice, the actual number is lower due to neutron captures in structural materials and transmutations of fertile nuclei. Let the number of such capture neutrons be ν_c. The number of available neutrons is then $\nu - 1 - \alpha - \nu_c$. Captures in structural materials typically consume at least 0.2 neutrons per fission,

particularly as reactivity changes are counterbalanced by the presence of consumable neutronic poisons. For each fissioning nucleus α fissile nuclei suffer neutron capture leading, in general, to a fertile nucleus. If one requires regeneration of the nuclear fuel, one sees that $\nu_c = 0.2 + 1 + \alpha$ at least. The number of available neutrons then amounts to $\nu - 2(1 + \alpha) - 0.2$. We consider four cases:

- **Thermal ^{238}U - ^{239}Pu:** $\nu = 2.871$ and $\alpha = 0.36$. The number of available neutrons is then -0.05. Regeneration is not possible and no neutrons are available for transmutation. The 0.1 additional neutrons made available by the use of an accelerator would allow regeneration;

- **Thermal ^{232}Th - ^{233}U:** $\nu = 2.492$ and $\alpha = 0.09$. The number of available neutrons is then 0.11. Regeneration is possible and 0.1 neutrons are available for transmutation. The additional number of neutrons that an accelerator would bring is significant;

- **Fast ^{238}U - ^{239}Pu:** $\nu = 2.98$ and $\alpha = 0.14$. The number of available neutrons is then 0.5. Regeneration is easy and therefore advantage of an accelerator is not compelling;

- **Fast ^{232}Th - ^{233}U:** $\nu = 2.492$ and $\alpha = 0.093$. The number of available neutrons is then 0.10. Regeneration is possible. The additional number of neutrons that an accelerator would bring is significant.

Therefore, it can be seen that the additional neutrons provided by the ADS prove significant in breeding capabilities of various fuel compositions.

3.3.4 Transmutation of minor actinides and LLFPs

Transmutation of MAs and LLFPs has already been explored in Section 3.3.4. We will now tackle the subject from the ADS point of view.

As already explained, the only practical way to dispose of actinides is to induce their fission. Both energy and neutrons are produced in fission, however several neutron captures may be necessary before fission occurs, so that the number necessary for actinide incineration will be $N_{cap} + (1 - \nu)$, where N_{cap} is the number of captures before fission. A fundamental parameter in MAs incineration is D (neutron excess paramter), which represents the number of neutrons necessary to incinerate a nuclei. A positive D value indicates that an actinide or an actinide mixture cannot be completely transmuted. Different values of D are shown in Table 3.1 and

$$D_i = \sum_{j,\alpha} R_\alpha P_j^{(\alpha)} y_i \tag{3.3.30}$$

where R_α is the neutron balance for reaction α (capture=1, fission=1-ν and decay=0), $P_j^{(\alpha)}$ is the probability that nuclide j, once it is formed, will undergo the specific reaction α and y_i is the total number of times a nucleus of species j is formed.

Isotope or fuel	**Fast spectrum**	**PWR**
^{232}Th	−0.39	−0.24
^{238}U	−0.62	0.07
^{238}Pu	−1.36	0.17
^{239}Pu	−1.46	−0.67
^{237}Np	−0.59	1.12
^{241}Am	−0.62	1.12
^{244}Cm	−1.39	−0.15

Table 3.1: *Values of D parameter for the incineration of different actinides.* [21]

It can be seen that most minor actinides cannot be completely transmuted in thermal systems and that fast systems offer more excess neutrons than thermal systems. In fact, incineration by fast neutrons is always a net neutron producer. This is due to the fact that fission cross-sections of fertile nuclei, which are very small or vanishing for thermal neutrons, are large for fast neutrons. Neutron balance is not the only parameter that should be considered for the choice of a particular system with the aim of waste incineration. The half-life of the waste in the neutron flux is also very important since it determines the inventory needed to reach a specified transmutation rate and the time it takes to get rid of the waste.

Regarding LLFPs, the neutron capture process is currently the only promising nuclear reaction for transmuting fission products. The capture process consumes neutrons, but fast reactors could deliver enough excess neutrons to allow the potentially troublesome long-lived fission products to be completely transmuted to shorter-lived or stable species. As already mentioned, only ^{99}Tc and ^{129}I are promising for transmutation. Therefore, the most efficient way to transmute fission products is to use neutrons which would be lost to captures in the structural elements or which would escape the reactor. That's why it has been proposed to capture neutrons in the resonances of fission fragments, whenever these display strong resonances. In this way, it is hoped that neutrons are captured by the fission products before they reach thermal energies where captures in structure materials are significant. We discuss these ideas in the case of a fast reactor using a lead reflector (which is a layer of material surrounding the core that reduces

neutron leakage by scattering escaping neutrons back into the core), taking into account the ^{99}Tc case, which is characterised by a strong resonance at $E_R = 5584$ MeV, with $\Gamma = 149.2$ MeV and $\sigma_a = 10^4$ (microscopic absorption cross-section) barns. The probability that the neutron survives an interaction, i.e. that it is not captured, is:

$$P_{surv} = e^{-\frac{\pi\Gamma}{\xi E_R}\left(\sqrt{1+\frac{\Sigma_a}{\Sigma_s}}-1\right)} \tag{3.3.31}$$

where Σ_a is the macroscopic absorption cross section, Σ_s is the macroscopic scattering cross section of lead and ξ is the average logarithmic energy loss per collision.

It is found that approximately 90% of neutrons encountering the relevant resonance energy are captured, if the concentration of ^{99}Tc nuclei relative to lead is 6×10^{-4}. In the context of the original energy amplifier design, about 6% of all neutrons were estimated to be captured within the lead, with roughly half of these captures occurring below 5 eV. This suggests that these lower-energy neutrons could indeed be captured by ^{99}Tc if it were present in the lead at the specified dilution. Based on an average of 2.5 neutrons produced per fission, this would mean about 7.5 neutrons could be absorbed in technetium for every 100 fissions occurring in the system. For an ADS with a 10 MW proton beam and $k_s = 0.98$, the annual number of neutrons captured by ^{99}Tc is $N_{Tc}^{(cap)} = 8.4 \times 10^{25}$. These captures correspond to the transmutation of about 14 kg of ^{99}Tc per year, resulting in an effective half-life for ^{99}Tc within this neutron flux of approximately 7.5 years. For fast reactors, even when using moderated assemblies, the shortest calculated ^{99}Tc half-life is 15 years, while in PWRs, this value is around 21 years. But the most crucial parameter for transmutation is the neutron flux level, as the effective lifetime of a nucleus in a neutron flux is inversely proportional to this flux value. In fact, the rate of transmutation of a nuclide j can be chacterized by the time T_j^{transm} needed to incinerate half of the initial mass:

$$T_j^{transm} = \frac{\ln 2}{\sigma_c^j \times 3.151 \times 10^7} \text{ years} \tag{3.3.32}$$

Therefore, transmutation of the toxic fission products in nuclear reactors and subcritical systems may be sensible if rates of nuclear interactions with neutrons are much higher than rates of natural decay $T_{\frac{1}{2}}$. That is, transmutation can be reasonable if $T_{\frac{1}{2}} \gg T_j^{transm}$. Values of $T_{\frac{1}{2}}$ and T_j^{transm} are shown in Table 3.2, while in Table 3.3 values of the parameter D for some LLFPs can be seen. Both parameters are fundamental in order to understand whether an isotope is qualifiable for transmutation.

Isotopes, j	$T_{1/2}$ (years)	T^{transm} (years)		**Recommendation from neutronic viewpoint**
		Fast spectrum	Thermal spectrum	
^{90}Sr	29	2.2×10^3	1.6×10^3	*Non-transmutable*
^{93}Zr	1.5×10^6	730	790	*Transmutable*
^{99}Tc	2.1×10^5	110	51	*Transmutable*
^{126}Sn	1×10^5	4.4×10^3	4.4×10^3	*Questionable*
^{129}I	1.6×10^7	160	51	*Transmutable*
^{135}Cs	2.3×10^6	310	170	*Transmutable*
^{137}Cs	30	2.2×10^3	1.1×10^4	*Non-transmutable*

Table 3.2: *Parameters of LLFPs to be eventually transmuted in a fast and thermal spectra with standard flux levels:* $\phi = 10^{15} \frac{n}{cm^s \cdot s}$ *and* $\phi = 10^{14} \frac{n}{cm^2 \cdot s}$ *respectively.* [22]

Transmutable isotopes	**D**
^{93}Zr	2.01
^{99}Tc	1.01
^{107}Pd	2.04
^{129}I	1.008
^{135}Cs	1.002

Table 3.3: *Paramter D of transmutable isotopes after 5 years of cooling time.* [23]

3.3.5 Spallation Process

The most peculiar process in an ADS is the spallation process. In Section 3.2.2 we described the spallation target, now we explore the physics behind the phenomena. Most of the hybrid reactor concepts use high energy protons, but other proposals use electrons or deuterons. We discuss the case of protons more thoroughly due to its importance.

Electronic range

Energetic protons and nuclei interact with matter mostly by collisions with electrons. These lead to progressive energy loss and, therefore, a slowing down of the protons. The energy loss due to electronic collisions is given by Bethe's formula which reads:

$$\frac{dE}{dx} = \frac{DZ\rho}{A} \left(\frac{Z_p}{\beta} \right)^2 \left(\ln \left(\frac{2m_e c^2 \gamma^2 \beta^2}{I(Z)} \right) - \beta^2 \right) \frac{\text{MeV}}{\text{cm}} \tag{3.3.33}$$

with A, Z and ρ respectively the mass number, the charge and the mass density of the target nucleus and Z_p, β and E respectively the charge, velocity and energy of the projectile. The constant D=0.3071 MeV cm^2/g; I is the average ionization potential of target atoms and $m_e c^2 = 0.511$ MeV. The projectile range, i.e. the distance it runs before losing all its kinetic energy, is, by approximation,

$$R_{el}(E) = \frac{E^{1.75}}{1.75 c_R}, \qquad \text{with } c_R = 496\rho\frac{Z}{A}Z_p A_p^{0.75} \tag{3.3.34}$$

Taking the examples of beryllium and lead we get, for a 1 GeV proton:

- **Beryllium:** R_{el}=250 cm;
- **Lead:** R_{el}=45 cm.

Nuclear range

While being slowed down, protons may undergo nuclear reactions. For proton energies larger than, typically, 100 MeV, the most violent reactions are called spallation. These account for most of the neutrons produced.

Using a black nucleus model, we arrive at the expression for the nuclear range:

$$R_{nuc} = \frac{31 A^{\frac{1}{3}}}{\rho} \tag{3.3.35}$$

Thus,

- **Beryllium:** R_{nuc}=35 cm;
- **Lead:** R_{nuc}=16 cm.

The probability that 1 GeV protons suffer nuclear reactions is very high both for beryllium and for lead. The nuclear range is smaller, relative to the electronic range, for light nuclei. On the other hand, the energy deposited in the target nucleus following a nuclear encounter is larger for heavy targets. This over-semplified considerations we have just made are only intended to give a feeling of the physics of the interaction of high-energy protons with nuclei. It showed that the proton energy should be chosen high enough that nuclear energy losses exceed electronic energy losses. A more detailed treatment requires nuclear cascade simulations. This interaction is complex and generally explained as a multi-stage process, leading to the emission of numerous nucleons (neutrons and protons), light charged particles, pions (if the incident energy is sufficiently high), and leaving behind a residual nucleus that is often different from the original target nucleus.

The spallation reaction can be broadly described in the following stages:

Intra-Nuclear Cascade (INC) Phase

This is the initial, fast stage of the reaction, occurring on a timescale of approximately 10^{-22} seconds. The incident high-energy particle (e.g., a proton) interacts with individual nucleons within the target nucleus through a sequence of quasi-free collisions. This creates a cascade of fast-moving nucleons and potentially pions within the nucleus. Some of these highly energetic "cascade" particles may gain enough energy to be directly ejected from the nucleus in a predominantly forward direction. The nucleus is left in a highly excited, non-equilibrium state.

Pre-Equilibrium (PE) Phase

Following the rapid INC phase, the nucleus enters an intermediate, pre-equilibrium stage. During this phase, the excitation energy is distributed among more nucleons, but the nucleus has not yet reached full statistical equilibrium. Additional energetic particles, mostly nucleons, can be emitted before complete thermalization is achieved.

Evaporation/Fission Phase

This is the final, slower stage of de-excitation (timescale $>\sim 10^{-16}$ s). The residual nucleus, now largely thermalized but still highly excited, unload the remaining excitation energy primarily by "evaporating" nucleons, predominantly low-energy neutrons, but also protons, alpha particles, and other light clusters. For very heavy target nuclei, high-energy fission can also compete with particle evaporation as a de-excitation channel, contributing further to the neutron yield and producing a range of fission fragments.

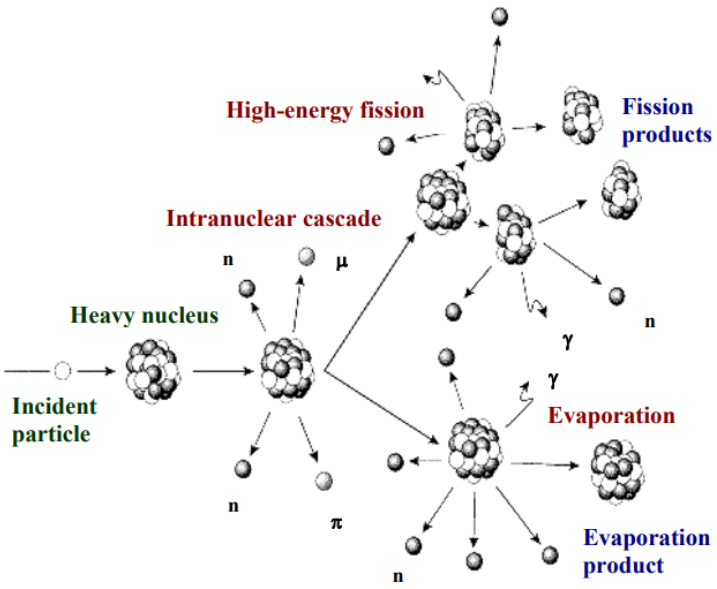

Figure 3.3: *Schematic representation of a spallation reaction. An incident high-energy particle strikes a heavy nucleus, triggering a multi-stage process that includes an intranuclear cascade, pre-equilibrium emission, and a final evaporation/fission phase, resulting in the emission of numerous neutrons and other particles.* [24]

The spallation process is a prolific source of neutrons. The neutron yield (N_0), defined as the average number of neutrons produced per incident proton, is a key parameter. It depends significantly on the incident proton's energy and the material and dimensions of the spallation target, but can be approximately described by the semi-empirical formula [25]:

$$M_n(E_p, A) = (0.0803 + 0.0336 \ln(E_p))A \tag{3.3.36}$$

gives better than 10% accuracy for A > 40.
For thick, heavy targets like lead or tungsten, and incident proton energies around 1 GeV, yields of 20 to 30 neutrons per proton are typically achieved. The energy spectrum of these spallation neutrons is broad, extending from the beam energy down to tenths of keV with the maximum around 2 MeV and differs so from fission neutron spectrum that reaches from thermal energies up to ≈10 MeV with the maximum around 1 MeV. Both the neutron fission and spallation spectrum and the neutron multiplicity are shown in Figure 3.4.

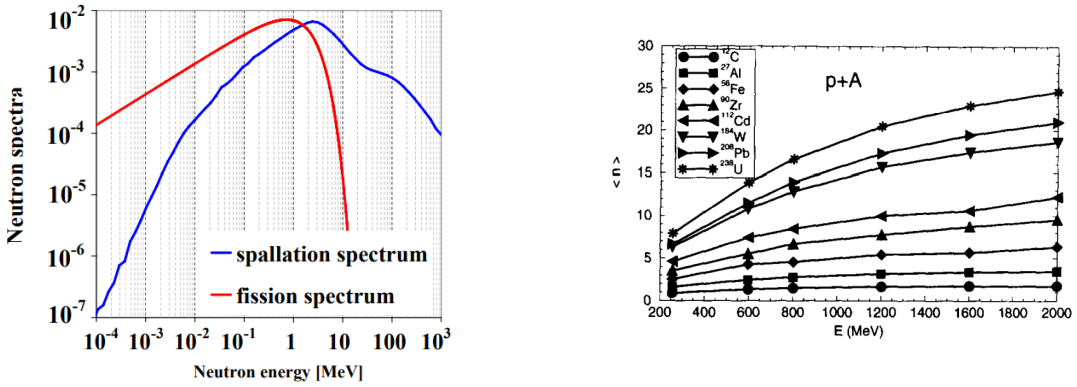

Figure 3.4: *Key characteristics of the spallation process. Left: Comparison of a typical spallation neutron energy spectrum with a fission spectrum, showing the presence of a high-energy tail in the spallation distribution. Right: Neutron multiplicity (average number of neutrons per proton) as a function of incident proton energy for various target materials, demonstrating a near-linear increase in yield.* [26]

A wide array of residual nuclei, or spallation products, are formed as a result of the spallation reaction. These products are the remnants of the target nucleus after the emission of multiple particles. They often include isotopes far from the line of beta stability and many are radioactive. The production of these spallation products leads to the activation of the target material and surrounding structures, which is a significant consideration for material damage, shielding requirements, remote handling, and long-term waste management of the target assembly. For example, the production of ^{210}Po (a highly radiotoxic alpha emitter) is a concern in LBE targets due to neutron capture on bismuth and spallation reactions.

Chapter 4

Current Developments, Projects and Future Prospects of ADS

After an overview of Nuclear Power, the problem of nuclear waste and the physics and components of ADSs, various projects and current developments of the ADS technology will be explored.

4.1 Role of ADSs in Nuclear Energy production

In the event of a massive use of nuclear power it is clear that any production system should be fuel breeding. Hybrid systems have very good characteristics in this respect. They would allow switching from a plutonium economy to a much less polluting thorium one. They could, in principle, allow the implementation of intrinsically safe reactors. They are also an attractive option for nuclear waste incineration, including minor actinides which would be difficult fuels for critical reactors. In their molten salt version they could allow fast plutonium incineration. Numerous issues have to be studied: reliability, safety, cost effectiveness, etc.

Hybrid systems require non-conventional technologies for the neutron multiplying assembly: molten salts, molten lead, natural convection, Th-U cycle. In principle such technologies could be used with critical reactors. The neutron surplus obtained from spallation is relatively small, especially for fast systems. The main asset of hybrid systems is their subcriticality which would allow building of reactors with deterministic safety and the use of fuels with unfavourable safety characteristics when used in critical reactors. They could also give a unique opportunity to improve the social acceptability of fission energy.

4.2 Ground laying proposals

The interest in ADSs originates essentially in the proposals made by Bowman and Rubbia. The first advocated a molten salt thermal neutron subcritical reactor, while the second advocated a solid fuel, lead cooled fast-neutron subcritical reactor. Almost all more recent work on ADSs elaborates on these two primordial proposals.

4.2.1 The Rubbia proposal

The proposal by Rubbia and his group, often referred to as the Energy Amplifier, is primarily aimed at massive energy production, though applications to nuclear waste incineration and transmutation were also considered. The system included an accelerator complex, consisting of three cyclotrons in series, to deliver a high-intensity proton beam (around 10 mA) at an energy of 1 GeV. This beam would strike a spallation target, and the produced neutrons would drive a subcritical assembly with a k_s of approximately 0.98. This configuration was projected to achieve a significant energy gain, potentially leading to a thermal power of 1500 MW and an electrical output of 600 MW.

The distinctive feature of the proposal was the use of liquid lead as a coolant for the fast neutron subcritical core. Lead was chosen for its favorable neutronic properties and safety characteristics, notably its high boiling point and chemical inertness compared to sodium. The design incorporated a large pool of molten lead (around 10,000 tons in a 30-meter high vessel), enabling primary cooling through natural convection and providing substantial thermal inertia.

A passive mechanism was proposed regarding safety. In case of significant overheating the lead was designed to overflow into the beam tube, thereby stopping the spallation neutron production, and into an inter-wall space to facilitate residual heat removal by air convection. This overflow would also introduce neutron absorbers to further reduce the system's reactivity.

The fuel cycle initially emphasized was the Th-U cycle, utilizing either oxide or metallic fuels. Reprocessing considerations included Thorex for oxide fuels or pyrochemical methods for metallic fuels. The neutronic design aimed for a relatively stable reactivity over the fuel cycle by balancing fission product poisoning with fissile breeding from thorium. [27]

4.2.2 The Bowman proposal

Professor Bowman's proposals for ADSs primarily centered on utilizing thermal-neutron molten salt subcritical reactors. His early concepts aimed at transuranic and fission product incineration, advocating for high thermal neutron fluxes (e.g., 10^{16} n/cm^2/s) to achieve rapid actinide burn-up with a small fissile inventory.

These systems often envisioned ^{233}U breeding from thorium, incorporating complex chemical reprocessing for ^{233}Pa and fission product extraction to maintain reactivity and neutron economy.

A more specific and later proposal, known as the TIER concept, focused on plutonium incineration with a emphasis on non-proliferation. The Tier-1 system was designed as a once-through transmuter using a molten NaF-ZrF_4 salt to carry plutonium and minor actinides from processed LWR spent fuel (uranium having been removed). Operating with $k_s \approx 0.96$ and a thermal flux around 2×10^{14} n/cm^2/s, it aimed to achieve significant actinide destruction in a single pass without reprocessing the transmuter salt itself, rendering the remnants less attractive for weapons use. An optional Tier-2 system, using Li-Be fluoride salts and chemical separations, was proposed for further reducing the remaining actinides from the Tier-1 output. The goal of the TIER approach was the immediate and irreversible elimination of the weapons proliferation risk associated with plutonium. Both approaches, however, highlighted the need for advanced high-power accelerators and posed significant challenges in materials science and chemical engineering for the molten salt systems. [28]

4.2.3 International Projects

A significant interest in ADSs exists in Europe, Japan and the US. Plans exist to demonstrate the interest of ADSs at a representative level.

Japan

In the early 2000s, Japan pursued significant research and development in ADSs for nuclear waste transmutation, primarily through institutions like JAERI and KEK. These efforts were often linked to national strategies such as the OMEGA program and the ambitious High Intensity Proton Accelerator Project.

The R&D landscape included conceptual designs for ADS, such as LBE cooled subcritical reactors utilizing nitride-based fuels. A key component was a multiphase plan for a high-power proton accelerator complex, intended to support an ADS experimental facility dedicated to transmutation studies. Experimental activities were wide, covering subcritical reactor physics, often using existing facilities like the Fast Critical Assembly for integral data. Concurrently, research addressed the challenges of LBE thermal-hydraulics, the development and testing of advanced nitride fuels suitable for transmutation, and the identification of robust materials for spallation targets and beam windows, critical for reliable ADS operation.

France

France is a key European contributor to ADS research and development, with activities largely coordinated through national programs like SPIN and collaborative groups such as GEDEON, which involved major institutions including CEA, CNRS, EdF, and Framatome. French efforts were significantly aligned with broader European initiatives, particularly in defining concepts for an Experimental ADS (XADS).

The main options considered for XADS typically included a fast neutron spectrum subcritical core, utilizing solid fuel, and driven by a proton beam in the 400 MeV to 1 GeV energy range via a spallation target. While lead-bismuth eutectic (LBE) was a common coolant choice in many international designs, French proposals also prominently featured gas-cooled (Helium) XADS concepts, offering an alternative to liquid metal systems. Experimental support for these conceptual studies was provided by neutronics experiments conducted at facilities like MASURCA in Cadarache (the MUSE program). Furthermore, France was actively involved in the development of critical accelerator technology, exemplified by the IPHI (Injecteur de Proton Haute Intensité) project at Saclay, which aimed to create a high-intensity proton injector suitable for future ADS applications, often in partnership with other European research bodies.

MYRRHA

The Multi-purpose Hybrid Research Reactor for High-tech Applications (MYRRHA), has been under development at SCK CEN in Belgium since 1998, and stands as the leading international project aimed at demonstrating the ADS concept at a pre-industrial scale. Its primary objectives include proving the efficiency of nuclear waste transmutation in an ADS, particularly minor actinides, to significantly reduce the volume and long-term radiotoxicity of SNF. Beyond transmutation, MYRRHA is designed to function as a multipurpose fast-spectrum irradiation facility, for instance serving as a materials testing reactor and securing the production of innovative medical radioisotopes.

The full MYRRHA facility, as shown in Figure 4.1 will feature a subcritical core fueled with highly enriched (typically around 30% Pu) MOX fuel. This core will be cooled within a pool-type primary circuit. The subcritical core is designed to be driven by a LINAC delivering a continuous wave proton beam of 600 MeV energy and up to 4 mA current. This proton beam will impinge on an LBE spallation target centrally located within the core to produce the necessary neutrons. The nominal design power of the reactor is 100 MWth, though in its primary subcritical mode (with a k_{eff} of approximately 0.95), the operational power is expected to be in the range of 65-70 MWth.

MYRRHA's implementation follows a phased approach. Phase 1, known as MINERVA, focuses on the construction and commissioning of the initial 100 MeV section of the LINAC, along with target stations for ISOL (Isotope Separation On-Line) based medical radioisotope production and for fusion materials research. The construction of MINERVA was planned to be completed by 2026, serving as a critical stage-gate for the subsequent phases. Phase 2 involves the extension of the accelerator from 100 MeV to its full 600 MeV energy, while Phase 3 encompasses the construction of the reactor itself. These latter phases can be executed in parallel, with the full MYRRHA facility anticipated to be operational around 2036.

The Belgian government has shown strong commitment to the project, approving its construction and operation until 2038 in September 2018. A significant budget was allocated, covering the construction of MINERVA (Phase 1), further R&D and licensing for Phases 2 and 3, and the operational costs of MINERVA, which has started construction in July 2024.

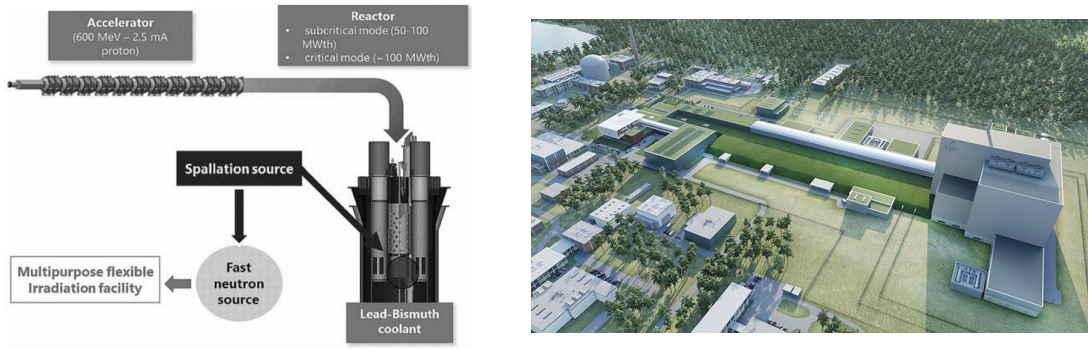

Figure 4.1: *The MYRRHA project. The left panel shows the three main interconnected components: the LINAC accelerator, the spallation source, and the lead-bismuth cooled subcritical reactor. The right panel provides an artist's concept of the complete research facility.* [29]

4.2.4 Final considerations

The transition of ADS technology from concept to widespread industrial application faces substantial hurdles. These primarily include ensuring the extreme reliability of high-power accelerators, developing materials and components for targets and cores that can withstand unique operational stresses, and establishing efficient, economically viable closed fuel cycles for advanced transmutation fuels. The overall cost-effectiveness of ADS, remains a critical factor for future deployment. Continued focused research, development, and successful demonstration are therefore essential to overcome these challenges and fully realize the potential of ADS to contribute to a more sustainable and safer nuclear energy future.

Conclusions

This thesis has embarked on a comprehensive exploration of Accelerator-Driven Systems, beginning with the fundamental principles of nuclear fission and the challenges posed by nuclear waste management, and culminating in a detailed analysis of ADS technology, its underlying physics, and its potential role in a sustainable nuclear energy future. The core concept of an ADS, which couples a high-energy particle accelerator to a subcritical nuclear reactor via a spallation target, has been thoroughly examined.

The primary motivation for developing ADS technology lies in its potential to transmute long-lived radioactive waste, particularly Minor Actinides (MAs) and selected Long-Lived Fission Products (LLFPs), thereby significantly reducing the long-term radiotoxicity and heat load of spent nuclear fuel destined for geological disposal. The subcritical nature ($k_{eff} < 1$) of the reactor core is a key feature, offering inherent safety advantages by precluding criticality accidents and allowing for greater flexibility in fuel composition. This flexibility is crucial for designing cores optimized for waste incineration, which might otherwise pose safety challenges in critical reactors.

The physics and neutronics of ADS were discussed in detail, covering aspects such as energy gain, the influence of the external neutron source on reactor kinetics, the modified role of delayed neutrons, and the neutron balance which dictates breeding and transmutation capabilities. The spallation process, responsible for generating the external neutrons, was also described, highlighting its efficiency in neutron production.

Key components of an ADS - the accelerator, spallation target, and subcritical core - were reviewed, along with the significant technological challenges that must be overcome. These include ensuring the high reliability and availability of powerful accelerators, developing materials capable of withstanding the harsh radiation and thermal environment of the spallation target and core, and establishing efficient and economical closed fuel cycles for advanced transmutation fuels.

The thesis also surveyed prominent historical proposals and ongoing international research and development efforts, such as the MYRRHA project in Belgium. These initiatives are crucial for demonstrating the viability of ADS technology and

for addressing the remaining technical hurdles.

In conclusion, Accelerator-Driven Systems represent a promising and innovative approach to mitigating the challenges of nuclear waste, potentially enhancing the safety and sustainability of nuclear power. While significant research and development are still required to bring ADS to industrial maturity, their unique capabilities, particularly in waste transmutation and inherent safety, warrant continued investigation. Successful demonstration through projects like MYRRHA will be pivotal in realizing the full potential of ADS to contribute to a cleaner and safer energy future.

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