



## Indicators for quantifying proliferation risk and nuclear waste issues in energy system models

*Master's Thesis in Engineering Physics*

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Cover: Illustration of some factors regarding nuclear reactor use.

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## **Abstract**

In order to better understand energy systems and to make effective, cost efficient decisions, models are built to evaluate different scenarios. When modelling energy systems, cost can easily be considered, while other more complicated side effects remain more difficult to model. To compare different nuclear technologies, models concerning proliferation and waste are sought. To this end, two model indicators were developed: unATTR - indicator of reactor waste storage and, ATTR - indicator of reactor waste fit for nuclear bomb construction. The model indicators were tested using different reactor technologies: normal light water reactor; a light water reactor with increased burn up; and, a light water reactor using MOX. Using unATTR and ATTR indicators across different reactor schemes and their waste enabled computer results in the form of code to single out best choice.



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Henrik Wall, Gothenburg December 17, 2012



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## List of abbreviations and terms

- Rouge state - A state which might act irrational or in a way which is not to their benefit.
- NED - Nuclear Explosive Device.
- Open cycle - Nuclear fuel usage scheme where no reprocessing occurs.
- Closed cycle - Nuclear fuel usage scheme when reprocessing is used.
- Availability factor - The amount of time that a power plant is able to produce electricity over a certain period, divided by the amount of the time in the period.
- MOX fuel - Mixed oxide fuel, a fuel type containing more than one type of fuel oxide. For instance natural uranium oxide mixed together with plutonium oxide obtained from reprocessed used nuclear fuel.
- LWR - Light Water Reactor. A reactor type using light water as coolant and moderator.
- Moderator - A medium used for slowing down neutrons.
- EOC - End Of Cycle.
- BOC - Beginning of Cycle.
- TRU - Trans Uranic Elements, for instance Np, Pu and Cm.
- Actinides - A group of radioactive elements within the periodic table with similar properties. Starts with  ${}_{89}\text{Ac}$  and ends with  ${}_{103}\text{Lr}$ .
- SF - Spontaneous Fission.
- IAEA - International Atomic Energy Agency.
- "Conversion time - The time required to convert different forms of nuclear material to the metallic components of a nuclear explosive device. Conversion time does not include the time required to transport diverted material to the conversion facility or to assemble the device, or any subsequent period. The diversion activity is assumed to be part of a planned sequence of actions chosen to give a high probability of success in manufacturing one or more nuclear explosive devices with minimal risk of discovery until at least one such device is manufactured" [1].
- IAEA timeliness detection goals - "The target detection times applicable to specific nuclear material categories. These goals are used for establishing the frequency of inspections and safeguards activities at a facility or a location outside facilities during a calendar year, in order to verify that no abrupt diversion has occurred" [1].

- Significant Quantity (SQ) - "The approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded. Significant quantities take into account unavoidable losses due to conversion and manufacturing processes and should not be confused with critical masses" [1].
- NPT - Non Proliferation Treaty.
- HEU - Highly Enriched Uranium





## 1 Introduction

Currently 436 nuclear reactors are in operation in 31 countries worldwide producing 370500MW<sub>e</sub>[2]. In early 2012 the nuclear produced electricity was 13.5% worldwide and in Sweden 37.5% [3]. 88.5% of the nuclear electricity is produced in LWRs (Light Water Reactors) which mostly consists of generation II reactors, which were build between the 1960s and 1990s, and were designed to have a life time of 40 years[4].

Most current operational reactors are more then 25 years old, see figure 1, while the average age of closed reactors are 22 years, despite being designed for a 40 year lifespan [5]. For several reasons many current day reactors have had their operational license extended to 60 years. By 2011, in the US, more than 58% had received a 20 year renewal on their licensed lifetimes [6]. According to figure 1, the bulk of current day nuclear reactors are over 20 years old, so if future energy systems are to contain nuclear power there is a need to build new reactors.

In May 2012 there were only two fast breeding reactors operating [2], fast breeding reactors are considered a generation IV concept considered to be reactors of the future. To determine whether to build new reactors, as for instance fast breeder reactors, it is necessary to compare and contextualise these with other energy sources in order to measure their competitiveness. The introduction of the European Unions' Emission Trading Scheme (EU ETS) places a price on CO<sub>2</sub> emissions for industry and the power sector [7]. This provides a framework for comparing different CO<sub>2</sub> emitting energy sources. Nuclear proliferation and long term waste are more complex aspects of nuclear power, which are more difficult to compare competitiveness in.

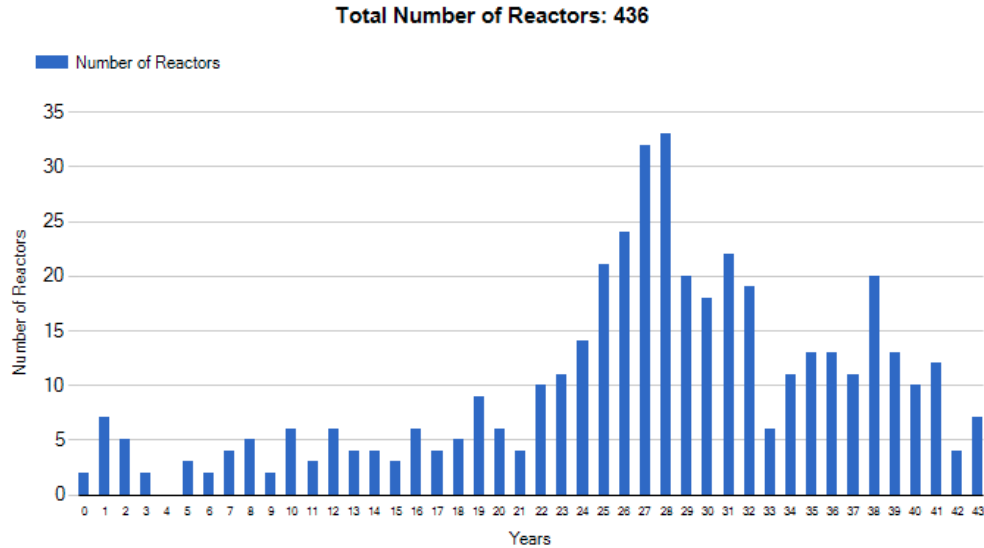


Figure 1: Distribution of current day reactor age [2].

## 1.1 Objective and scope of this thesis

Nuclear power is an energy form associated with many benefits as well as drawbacks. Among the most appealing aspects of nuclear energy are energy security. Due to high energy content in the fuel, low stockpile volumes are needed as well as low impact on electricity generation costs when it comes to fuel price changes [8]. Improving energy security can be achieved by reducing the vulnerability of economic activity in case of disruptions of energy supply [9]. For a country primarily using natural gas, disturbances in the Middle East have the potential to cause disruptions to the energy availability in the country. A better alternative may be Uranium which is a mineral naturally occurring across globe. Another major driving factor is the carbon emission free fission process. Current CO<sub>2</sub> emissions are about 1% of the total atmospheric concentration every year [10]. If one is to successfully meet policies to stabilize the atmospheric CO<sub>2</sub> concentration to 450ppm the need for CO<sub>2</sub> neutral energy sources, are vital to avoid an increase in atmospheric content. Nuclear power plays a large role in CO<sub>2</sub> neutral energy being produced in the world, but in order to increase this impact in future, more nuclear power plants needs to be built. However the benefits of nuclear power also come with numerous drawbacks, like nuclear proliferation and nuclear waste.

Quantifying a value for the produced radioactive waste, the by product from nuclear power generation, which might be dangerous tens of thousands years after the CO<sub>2</sub> neutral energy was harvested, and proliferation, are both major subjects concerning the use of nuclear energy. This involves the spreading of material, technologies or information that can be used to construct a nuclear explosive device (NED). For instance an atomic bomb in the hands of a "rouge state" or terrorist organization is a notion unacceptable for most people. The limited fuel reserves and risk of accident are two other oppositional arguments to nuclear energy.

The goal of this project is to develop indicators for the nuclear waste and the proliferation issue that can be used in energy system modelling. The aim is to compare different future energy scenarios using different indicator values. As the goal is to compare future energy scenarios the focus on proliferation, centres around Plutonium content in fuel, and not Uranium. As enrichment facilities might be the current day highest proliferation risk, the amount of enrichment facilities required are calculated using a simple model. The future aim of the developed indicators in this thesis are to evaluate future energy systems where fast breeder reactors might be in use and little to no enrichment will be required. When developing the indicators the arguments and decisions are scientifically based which means public or political opinion has been excluded. The risk then of shutting down a nuclear power plant prematurely, is not taken into account even though this type of action might produce high risk material in a proliferation sense.

## 2 Background

### 2.1 Nuclear history

In a document dated back to July 1934 Szilard explained experiments that, if they were successful would lead to power production on a scale and probably of such little cost that a sort of industrial revolution could be expected: "it appears doubtful for instance whether coal mining or oil production could survive after a couple of years" [11]. The first ever operating nuclear reactor was demonstrated in Chicago on December 2, 1942 led by Enrico Fermi. During World War II, the primary focus of the nuclear program was to support the military. The major goal for the American nuclear program was to produce  $^{239}\text{Pu}$ , which had already been recognized as an effective material for a nuclear bomb. The effect of the first reactor was 200 W, while less than two years later, a 200MW reactor began operating. This was a million fold increase of power in less than two years [12]. Notably, this 200MW reactor was the first full size reactor ever built and it was produced within 15 months [11].

The reactors built in the 1950s are considered first generation reactors, while most current day reactors are now Generation II, for instance, the CANDU (Canada Deuterium Uranium) reactor and the Swedish LWRs. Around the middle of the 1990s the progression to Generation III reactors was made. These models included improved reactor technology, improved passive safety systems, superior thermal efficiency, standardized design to decrease capital costs and reductions in maintenance costs [13]. Many of the world's nations both industrialized and developing believe that nuclear energy can be used now and in the future to meet their growing demands for energy, safely and economically [4]. To further enhance the future role of nuclear energy, Generation IV reactors are being researched and developed considering improved safety, economic efficiency, higher reliability and increased proliferation resistance [14]. Proliferation resistance is a new consideration being taken into account in the design of Generation IV reactor systems. Historically, a problem facing nuclear energy has been the connection between military nuclear use and civil use, since by reprocessing spent nuclear fuel one obtained the plutonium used in the early NEDs. In 1976 the US gave a clear signal that they wanted to limit reprocessing techniques when President Gerald Ford issued Presidential Directive, to suspend the commercial reprocessing and recycling of plutonium in the US. Further the American president Carter was advised by 21 scientists, economists and political scientists to abandon the US reprocessing program and postpone the breeder building program by 10, 20 or even more years [15]. The banning of reprocessing commercial reactor spent nuclear fuel due to the potential spread of nuclear weapons by President Jimmy Carter, aimed to encourage other nations to follow the US lead. However, the UK and France who had already developed reprocessing programs along with Russia, India, Pakistan and Japan who additionally had built reprocessing facilities did not abandon them [16]. Even though the US ban was lifted in 1981 by President Reagan, no industrial scale reprocessing plants have operated in the US since.

## 2.2 The nuclear power system

Lately some countries have decided to shut down their nuclear programs based around the central issues of nuclear power such as, resource limitation, non-proliferation and waste management. All are connected with the fuel cycle.

The 6 main components in a simplified fuel cycle are:

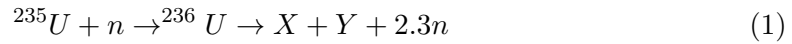
- The mills - Extracts uranium from the mined ore. This is a stage that produces the biggest waste streams since uranium ore is quite dilute.
- Enrichment facilities - In current reactor technology the fuel isotope of uranium is the  $^{235}\text{U}$  isotope, which in LWRs need to be present with at least 3% isotopic composition. Since natural uranium only contains 0.7%  $^{235}\text{U}$  the uranium needs to be enriched in  $^{235}\text{U}$  isotope. The enriched stream is used for fuel while the depleted uranium may either be stored for further recovery of  $^{235}\text{U}$ , used for fuel in fast neutron reactors or just be treated as waste. ( In some cases for military use in for instance warheads and armor in tanks)
- Fuel fabrics - The desired fuel is fabricated using the enriched uranium and in some cases recovered plutonium.
- The reactors - Here the fuel isotopes, mainly  $^{235}\text{U}$  and  $^{239}\text{Pu}$ , are burned and produces fission products together with breeding into heavier nucleus often referred to as trans uraniums (TRU), for instance  $^{238}\text{U} \rightarrow ^{238}\text{Np}$  which decays to  $^{239}\text{Pu}$ .
- Spent nuclear fuel storage - The spent fuel will be of different size and properties depending on reprocessing, but still there will be a need to store low active material and fission products. The spent fuel is often referred to as SNF- Spent Nuclear Fuel.
- Reprocessing facilities - This stage is dependent whether a open or closed fuel cycle is used. In case of a closed fuel cycle this is the stage where fissile materials can be recovered to use for production of new fuel, mainly even atomic number and odd neutron number e.g  $^{239}_{94}\text{Pu}$ . If the conversion ratio(see section 2.3) is high enough, there is a possibility of recover sufficient fissile material for producing new fuel. This scenario would make a possibility of phasing out much of the enrichment facilities and milling facilities in the long run.

By 2008 there were 5 commercial reprocessing plants in use, for the purpose of recovering uranium and plutonium. The process used today is called PUREX (Plutonium Uranium Extraction). The first known application for nuclear reprocessing was the Manhattan Project which supplied the military with weapon material [17]. Reprocessing can easily be defended from a resource point of view, since a open (once through) fuel cycle only make use of a small amount of the energy content in the fuel. If one defines nuclear resource utilization as (fuel fissioned)/(resource input), one reaches a 0.75% utilization for a CANDU reactor at 7,5 MWd/tHM burnup, while the number is even

lower for a LWR, 0.55%, due to a 5.5 times higher uranium ore usage because of the enrichment process [18]. So the potential for unused energy in spent nuclear fuel is very high since not even one percentage of the energy in the fuel is used. However even due to the very low utilization in an open fuel cycle there is yet to be proved economic benefits with reprocessing. Further the tight connection between separation and weapon grade plutonium makes a big proliferation issue.

### 2.3 Reactor Physics

The energy in the fission process can be calculated via the famous formula  $E = mc^2$ . The main process in a LWR using Uranium as fuel can be described as in equation 1 where the difference in mass between the right hand side and left hand side is the mass that is transferred to energy according to Einstein's formula.



What is really happening is that  $^{235}\text{U}$  absorbs a neutron, creating the compound nuclei  $^{236}\text{U}$ , which is a nuclei containing too much energy to be stable so it splits. The two resulting elements (fission products and neutrons) will have a lower total mass than the fissioned  $^{235}\text{U}$ , which according to Einstein's formula is proportional to the released energy. This energy will be in the form of kinetic energy in the fission products and the neutrons.

Fissile nuclei, which amongst others refers to  $^{235}\text{U}$  and  $^{239}\text{Pu}$ , have an unstable compound nuclei created after a neutron absorption which means that the nuclei will split. Fissile nuclei are what is used in today's nuclear reactors as fuel. Fissionable means that just by neutron absorption the compound nuclei will not contain enough energy for fission, however extra energy can be added if the neutron is absorbed while having high speed, i.e. high kinetic energy. The drawback with fissionable nuclei versus fissile nuclei is that the cross section (probability) to absorb a neutron decreases with increased neutron energy which means a much higher neutron flux is needed in order to compensate for the lower cross section if the total number of fissions are to be the same, i.e. the power level.

Today's nuclear reactors are operated in a critical regime, which means that every fissioned nuclei will induce one more fission. This is referred to as  $k_{eff} = 1$ , where  $k = \frac{\text{number of fission neutrons in generation } n}{\text{number of fission neutrons in generation } n-1}$ . In reactor physics a simplified version of  $k$  can be expressed as in equation 2.

$$k = \eta f p \epsilon \quad (2)$$

In equation 2,  $\eta$  is average neutrons released by fission,  $f$  the probability to get absorbed in fuel,  $p$  probability to escape the resonance peaks and  $\epsilon$  the fast fission factor. There are three defined regimes depending on the  $k$  value,  $k < 1$  is subcritical,  $k = 1$  critical and  $k > 1$  is super critical.

Another category are fertile nuclei which means that by absorbing a neutron the result is a fissile nuclei. Examples of fertile nuclei are  $^{238}\text{U}$  which by neutron absorption and a following beta decay will result in  $^{239}\text{Pu}$ , see figure 4. By deciding the average

production of fissile nuclei per fissile fuel nuclei consumed one can define a conversion ratio as  $\frac{\text{production rate}}{\text{burn rate}}$ . When the conversion ratio exceeds unity there will actually be more fuel produced than what was used, this requires low  $p$  and high  $\eta$ . High  $\eta$  is often seen in the transuranic elements.

## 2.4 Nuclear Reactors

### 2.4.1 Light Water Reactors

A current day LWR are operated at around 290 degrees Celsius with an efficiency of approximate 33% [18]. This can be compared with the assumed efficiencies of more than 40% in future design reactors as well as much higher operation temperatures [4]. Higher outlet temperatures in certain designs can also enable for instance production of hydrogen or other high temperature processes to be integrated within the power plant for increased economics.

Light water reactors uses a thermal spectrum of neutrons, i.e the produced neutrons (see equation 1) are slowed down in a moderator in order to have higher probability for fissioning a  $^{235}\text{U}$  nuclei. This means that during the slowing down the neutron can be lost in numerous ways, absorbed in the moderator, absorbed in the fertile  $^{238}\text{U}$  or by escaping the reactor without interacting with any material. To some extent the loss of neutrons must occur, since from  $^{235}\text{U}$  the neutron release per fission is on average 2.3 and without loss there would be 2.3 times as many fissions in the end of the next neutron generation. In a swedish LWR there is on average 0.6 fissioned  $^{239}\text{Pu}$  on every fissioned  $^{235}\text{U}$ , which means that the at first glance useless  $^{238}\text{U}$  stands for a substantial amount of the produced energy. If one assumes the released energy from a fissioned plutonium nuclei and uranium nuclei would be the same, then the amount of energy produced from fissioned plutonium would be  $\frac{0.6}{1.6} \approx \frac{1}{3}$ . The conversion factor of such a plant is 0.6. If the conversion factor reaches unity, then a reactor would produce as much fuel as it uses up. Such conditions would only last as long as the conversion factor is actually unity or above, for instance decreasing  $^{235}\text{U}$  in the core could decrease the conversion factor in a reactor.

In a light water reactor a small fraction of the neutrons released by fission are emitted as delayed neutrons with a delay time up to 57 seconds. For  $^{235}\text{U}$  0.65% of the neutrons are delayed neutrons which means that if  $k > 1.0065$  the reactor is in a prompt critical state, where the reactor is critical even without the delayed neutrons. This must never happened, since going from the critical state to the prompt critical state changes the time to sustain the chain process is decreased from 57 seconds to 0.1 milliseconds, which is the average neutron life time in a light water reactor. Due to the delayed neutrons the actual effective neutron life time is around 0.1 seconds instead of 0.1 milliseconds. The reactor power is proportional to the neutron flux and the number of neutrons in a reactor can be described by  $N(t) = N_0 e^{\frac{kt}{T}}$  where  $T$  is the effective neutron life time. So when going from  $T = 0.1$  to  $T = 0.1 \cdot 10^{-3}$ , a factor of  $10^{-3}$  less, one reaches the new neutron flux as  $N(t) = N_0 e^{\frac{1000kt}{T}}$  which corresponds to a reactor power many thousand times higher.

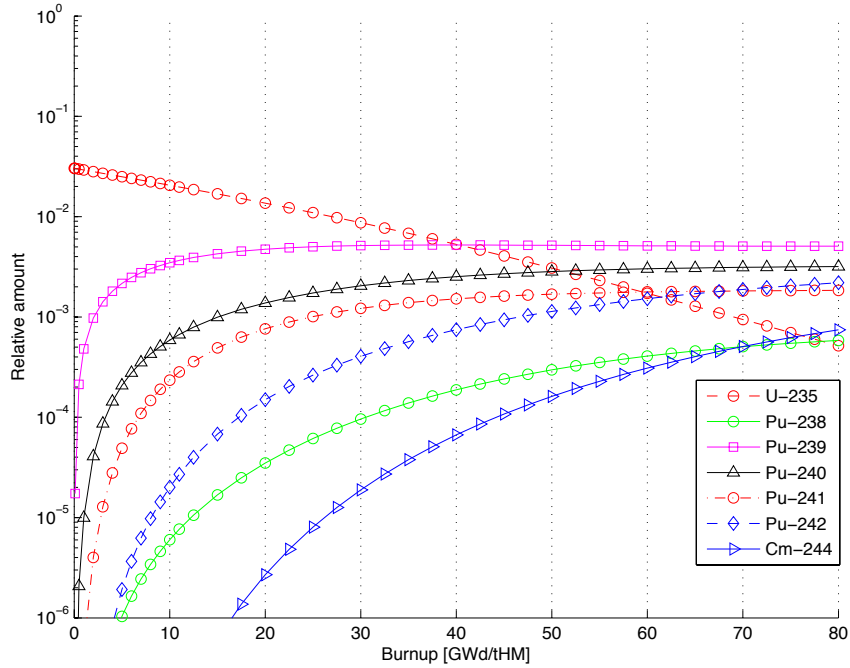


Figure 2: Percentage amount of certain isotope build up in a LWR.

When operating a nuclear power plant the chain process is sustained over long time. Due to the sustained chain process a lot of different isotopes will be produced, both due to transmutation and as fission products.

In figure figure 2 the amount of isotopes, relative to each other, that are important to the radio toxicity shown in figure 5 can be seen. What one can see is that with increased burnup the amount of higher actinides (Pu and Cm) will increase, since for instance  $^{244}\text{Cm}$  occurs first after a burnup of 15GWd/tHM. Burnup is the thermal power multiplied by operation-days divided by initial fuel loading. So for instance a burnup level of 5GWd/tHM in 1GW<sub>e</sub> reactor loaded with 24t fuel, corresponds to  $5 * 24/3 = 40$ days. The shape of figure 2 is quite obvious since the actinides are created via breeding processes and the longer the reactor is critical, the more breeding events will occur. For instance the relative amount of  $^{244}\text{Cm}$  increases with higher burnup, which might be good from proliferation perspective since  $^{244}\text{Cm}$  is an isotope with high spontaneous fission grade which makes the reactor waste less attractive as bomb material for reasons explained in section 3, while it might the waste to dangerous to be kept near human activities for a long time due to its long half life and high radio toxicity.

In figure 3 one can see how the relative amount of  $^{239}\text{Pu}$  compared to all other plutonium isotopes depends on the burnup. This shape of the curve in figure 3 also fits with results of Mark et al. in [19]. So since relative  $^{239}\text{Pu}$  amount decreases with burnup

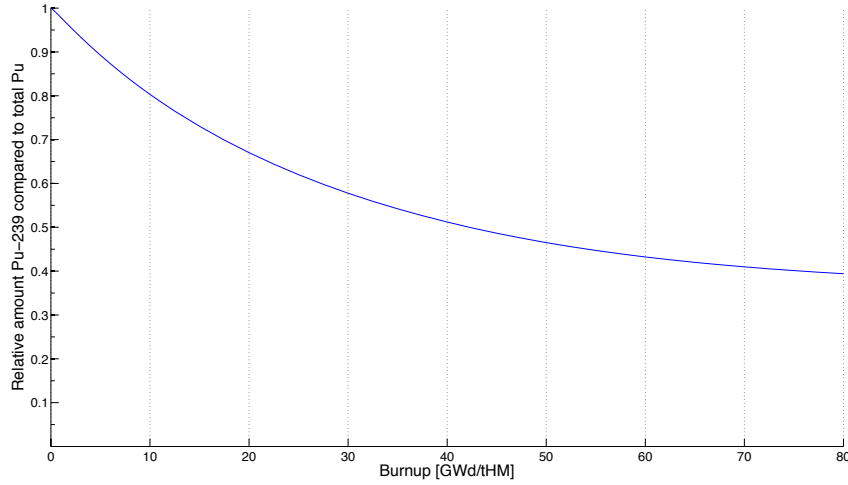


Figure 3: Relative amount of  $^{239}\text{Pu}$  compared to total amount of plutonium with respect to burnup, the data used is produced by the computer code called CASMO 4e.

a longer burnup is important when it comes to proliferation issues, see section 2.6, since for bomb making one would like as pure  $^{239}\text{Pu}$  as possible. For instance stopping a  $1\text{GW}_e$  reactor within 40 days (corresponding to burnup level of  $5\text{GWd/tHM}$ ), the produced plutonium would be of good enough quality to make a good reliable bomb since as can be seen in figure 3 the waste would contain 94%  $^{239}\text{Pu}$  which is categorized as weapon grade [19] and no isotopic separation would be needed. As stated in section 1.1 this is something that is not taken into consideration in this thesis, instead calculations are performed for designed burnup levels.

Even though higher burnup levels are attractive from proliferation perspectives it is usually harder to maintain certain safety issues with increased burnup. One example being the pellet-cladding interaction(PCI), which occurs when the fuel swells and come into contact with the cladding surrounding the fuel. Higher burnup causes higher cladding temperatures due to increased heat generation within the fuel, risking the melting/cracking of the cladding. This must be avoided since the cladding is part of the barrier system to block the passage of radio toxic elements from within the fuel [18].

#### 2.4.2 Fast Breeder Reactor

A different strategy than thermal reactor is a fast spectrum reactor. One difference is that the neutrons are not slowed down in a moderator, so a moderator is not necessarily at all. Because fast spectrum reactors do not depend on thermal neutrons, one can use the more abundant non fissile  $^{238}\text{U}$  and convert it via breeding processes to fissile  $^{239}\text{Pu}$  which can be used as fuel. This is possible only with a good neutron economy making  $^{239}\text{Pu}$  a much better suited fuel than  $^{235}\text{U}$  since  $^{239}\text{Pu}$  releases 2.9 neutrons on



average per fast neutron induced fission while  $^{235}\text{U}$  releases 2.4 on average [18]. Worth mentioning is the possibility of breeding  $^{233}\text{U}$  from Thorium which would avoid the breeding of  $^{239}\text{Pu}$ . This idea is considered by different countries having large supplies of Thorium. However this track involves different difficulties involving safety issues and will produce  $^{233}\text{U}$  which is also an isotope requiring government under the IAEA (International Atomic Energy Agency) safeguard system, see table 4a.

## 2.5 Fuel

Current day nuclear reactors uses  $^{235}\text{U}$  as the fuel loaded into the core. However once the chain reactions starts there are some additions of fuel via breeding processes that converts  $^{238}\text{U}$  to plutonium isotopes, see figure 4. When a nuclear core is critical many different nuclei reactions occur, fission, (n,2n), neutron capture followed by beta decay etc. All those processes contributes to the isotopic composition of the spent nuclear fuel. Natural uranium contains 0.7204% fissile uranium and 99.2742% fertile  $^{238}\text{U}$ . And the current inventory of uranium reserves recoverable at the maximized cost of 130 USD/kg U is 5,404,000 tones [20]. There have been interests in making use of reprocessing techniques to utilize more of the uranium content since an open fuel cycle only utilize approximately 0.6% of the available energy[18]. There are different views here though, some are of the opinion that there is enough uranium to support all nuclear mankind would possible use, while others prioritize efficient resource usage. For instance making use of a breeder technique and reprocessing today's uranium resources would much longer, since instead of utilizing mostly  $^{235}\text{U}$  which only exist to 0.72% in nature, one can make use of much of the other 99.3%. The commercialized reprocessing techniques used today are however based on a military technique in which pure plutonium streams were sought for. With this in mind it is not hard to understand concerns raised about nuclear proliferation issues when discussing reprocessing. However, there is research done and possibilities of new separation techniques developed for a commercial purposes [21]. Benefits of such process would be the possibility of shattering the link between reprocessing and nuclear bombs since no pure plutonium would be handled. Instead all reactor fuel would be handled as one unit leaving separation of plutonium, which is indeed a complicated process for a potential misuser. What is left then is the economic side. For instance the Electrical Power Research Institute (EPRI) did an investigation for fuel cycle cost comparisons between open cycle and a closed cycle with the main result being that with \$312/kgU the price for a closed cycle would be \$750/kgHM. But in the case where the unit Uranium price would increase with a factor of five, then reprocessed fuel cycle would cost as much the open one [21]. The fuel produced by reprocessing plants is referred to as MOX fuel.

### 2.5.1 Waste

Nuclear power plants produces three different types of radio active waste, which is classified according to the activity level. Low activity waste includes machines, clothes, filters etc which is about 90% of the total waste volume [22]. This waste is according to IAEA

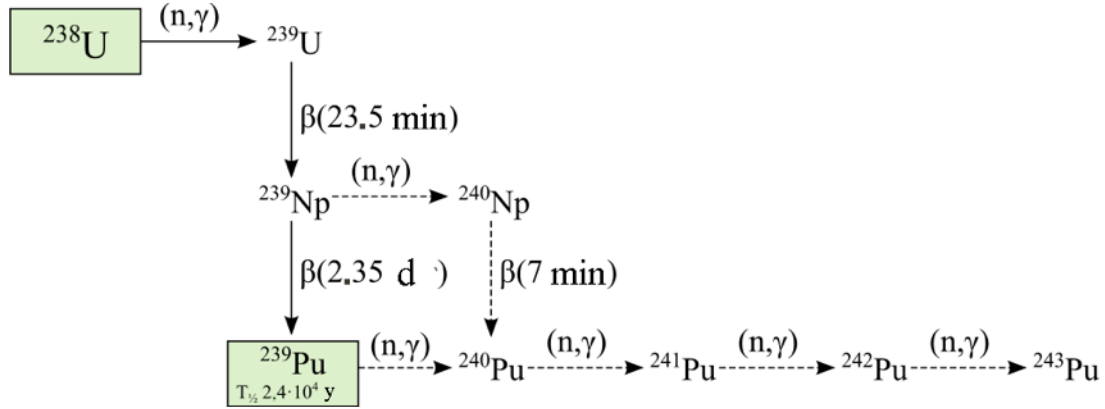


Figure 4: Figure showing the steps in which plutonium isotopes are created within a nuclear reactor.

suitable to be stored in engineered near surface facilities [23]. The intermediate level waste which typically is chemicals and fuel cladding is about 7% of the waste volume [22]. Intermediate level waste should be stored in order of tens of metres below surface level [23]. This leaves the high active waste, which is the actual spent fuel. Those 3% of the volumetric waste contains approximate 95% of the total waste radioactivity and should for example be stored in deep, stable geological formations on depths of at least several hundred metres or even more.

### 2.5.2 Spent Fuel

The radio toxicity in spent nuclear fuel is made up of different factors. Most accountable for the longterm toxicity are plutonium isotopes and minor actinides while the short term toxicity is due to the fission products (FP), see figure 5. Worth mentioning here is that even though the minor actinides have fairly low cross sections for fission, the numbers of neutrons released in a fission event is quite high [24]. The same goes for plutonium, which creates a possibility to reprocess spent nuclear fuel and extract those elements and create new nuclear fuel since a high number of released neutrons in fission events are important for a good neutron economy in a core. The radio toxic material left for storage would then only be the fission products. By looking at figure 5 one can see that the time scale fission products (FP in the figure) are dangerous are much shorter then the time of minor actinides (U, Np, Pu, Am and Cm). This creates the possibility of shortening the deep repository storage time for spent nuclear fuel if one can successfully separate away the fission products from the minor actinides and only store the fission products while recycling the minor actinides into new reactor fuel.

In table 1 the plutonium composition in fresh MOX fuel can be seen. As one can see the total amount of plutonium is increasing in every generation, but one can also see that the amount of  $^{239}\text{Pu}$ , the plutonium used to create bombs are decreasing. The

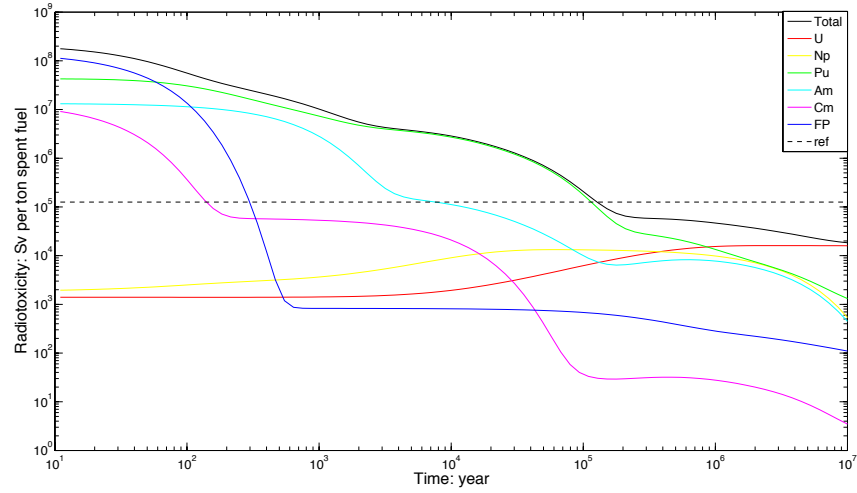


Figure 5: Radio toxicity in non reprocessed UOX fuel with initially 4.0%  $^{235}\text{U}$  enrichment. Burned to 45GW/tHM and 10 years of cooling in onsite pools. The reference level is the radio toxic level of natural uranium in equilibrium with its daughters for the amount of natural uranium needed to produce 1 ton of enriched uranium.

increase of heavier plutonium isotopes happens since by exposing the reprocessed fuel for longer times in the reactor the breeding process seen in figure 4 will act over longer times causing higher buildup of heavier isotopes.

Nuclide	1 st	2nd	3rd
$^{238}\text{Pu}$	3.5	4.2	4.7
$^{239}\text{Pu}$	51.9	43.9	40.5
$^{240}\text{Pu}$	23.8	27.4	28.8
$^{241}\text{Pu}$	12.9	13.6	13.1
$^{242}\text{Pu}$	7.9	10.8	12.8
Pu[% of total]	9.1	12.8	14.3

Table 1: Change of Plutonium composition (weight percent) in fresh MOX fuel in the  $n_{th}$  generation, where  $n = 1, 2$  or  $3$  [25].

Isotope	Content(weight %)	Isotope	Content(weight %)
$^{235}\text{U}$	0.77	$^{241}\text{Pu}$	0.16
$^{236}\text{U}$	0.55	$^{242}\text{Pu}$	0.10
$^{238}\text{U}$	92.19	$^{241}\text{Am}$	0.04
$^{237}\text{Np}$	0.07	$^{243}\text{Am}$	0.03
$^{238}\text{Pu}$	0.04	$^{244}\text{Cm}$	0.01
$^{239}\text{Pu}$	0.62	$^{243}\text{Cm}$	0.001
$^{240}\text{Pu}$	0.29	Fission products	5.15

Table 2: Isotopic composition in weight percentage of spent PWR fuel 4.2% enrichment after 4 years of cooling [25].

If one instead look on the open fuel cycle, i.e no reprocessing, the total composition can be seen in table 2. When it comes to radio toxicity one can see in figure 5 that the total toxicity is built up by different components. The reason for the long term radiotoxicity are the higher number nuclides, for instance plutonium. So by just using MOX fuel in a one time reprocessing cycle will increase the storage time required due to higher buildups of plutonium as could be seen in table 1.

## 2.6 Nuclear explosive device (NED)

In order to build a NED, fissile material in the metallic form needed, for instance  $^{235}\text{U}$  or  $^{239}\text{Pu}$ . The basics of a NED is to bring together a super critical mass fissile material at will, causing an increased number of fissions in every neutron generation, i.e  $k > 1$ . The basic NEDs that were designed and built during world war 2 was built on two principles. The first design is called a gun-type and utilizes the idea to merge two slightly sub critical masses together to create a super critical mass. This design however is often discarded when it comes to plutonium based bombs and considered better for Uranium based bombs.

In table 3 the bare critical-mass (the mass required to achieve  $k_{eff} = 1$  in a spherical geometry) for different plutonium isotope metals can be seen. The basis of conclusions,

	$^{238}\text{Pu}$	$^{239}\text{Pu}$	$^{240}\text{Pu}$	$^{241}\text{Pu}$	$^{242}\text{Pu}$
$\alpha$ - heat power [W/kg]	567	1.93	7.06	3.4	0.12
Spontaneous fission [n/g s]	2660	0.0226	1030	0.0493	1720
Bare critical mass of plutonium metal [kg]	13.1	14.8	44.8	17.6	87.8

Table 3: Characteristic data of different Pu-isotopes [27]

that NEDs could be designed using any isotopic composition of plutonium, is often based on the low critical mass values seen in table 3 according to Kessler et al. [26]. Kessler et al. further states that this conclusion neglects that a  $^{238}\text{Pu}$  content of several percent makes such a NEDs technically unfeasible due to reasons such as alpha-heating and spontaneous fission (SF).

The approach from IAEA, the International Atomic Energy Agency, is very concrete: All plutonium compositions, excepts  $^{238}\text{Pu}$  content exceeding 80%, being useable for a NED and that 8kg of plutonium the significant quantity (SQ), see table 4a. The meaning of SQ is the amount material needed to create one nuclear bomb. This means that there will be no differentiation between reactor grade plutonium and weapon grade plutonium. Those restrictions have been in place since 1972 [26]. Recent reports and results tell a story about proliferation proof and resistant plutonium due to high alpha heating and spontaneous fission rates (SFR), which is not taken into account in IAEA safeguard system [26], [28].

Nuclear Material Type	Significant Quantity
Pu(<80% $^{238}\text{Pu}$ )	8kg Pu
$^{233}\text{U}$	8kg $^{233}\text{U}$
HEU (>20% $^{235}\text{U}$ )	25kg $^{235}\text{U}$
LEU (<20% $^{235}\text{U}$ )	75kg $^{235}\text{U}$
natural U	10t natural U
depleted U	20t depleted U
Thorium	20t Thorium

(a) Definition of significant quantities for IAEA nuclear material types [29].

Nuclear Material	Material Form	Conversion Time	IAEA Timeliness Goals
Pu, HEU or $^{233}\text{U}$	Metal	7-10 days	1 month
Pure Pu components	Oxide	1-3 weeks	1 month
Pure HEU or $^{233}\text{U}$ compounds	oxide	1-3 weeks	1 month
MOX	Nonirradiated fresh fuel	1-3 weeks	1 month
Pu, HEU or $^{233}\text{U}$	in scrap	1-3 weeks	1 month
Pu, HEU or $^{233}\text{U}$	in irradiated fuel	1-3 months	
LEU, Nat U, Depleted U and Th	Unirradiated fresh fuel	1 year	1 year

(b) Definition of conversion times and timeliness goals for IAEA nuclear material types [29]. With conversion time one mean the time required to convert different form of nuclear material to metallic components of a NED and timeliness goal is the target detection time which is used to determine the frequency of inspections.

Table 4

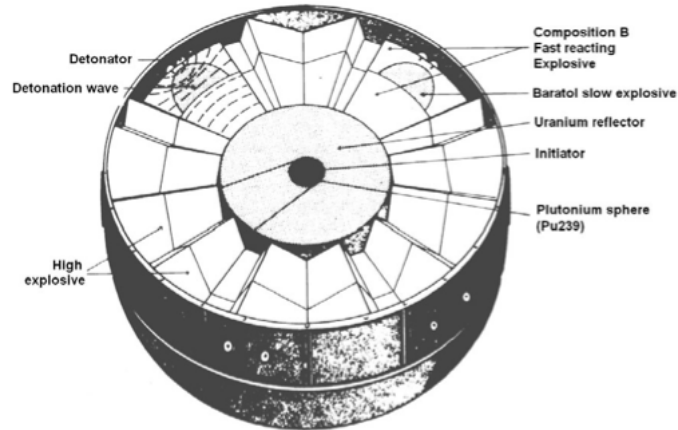


Figure 6: Design of the first Plutonium nuclear explosives, Fat Man [12].

The first successful plutonium bombs were made out of an implosion design and that design is the one considered when investigating feasibility for creating a NED out of reactor grade plutonium. The design of the first implosion bomb can be seen in figure 6 and uses chemical explosives to create fast traveling/high pressure waves into the spherical fissile core compressing it. Reason for this is that  $k$ -value depends on density, so with increased density one can achieve criticality with a normal subcritical mass as long as one manages to decrease the volume. One problem with this approach is the risk of pre detonation. Pre detonation occurs in the case of high neutron background (i.e for high SPF rates), which can initiate the explosion before the pressure waves has compressed the whole plutonium sphere causing the neutron multiplication to be greater the unity in only a small fraction of the fissile core. The explosion yield from such a pre detonation might be significantly lower, for instance Kessler et al. calculates one nuclear explosive yield to be 120 t of TNT which can be compared with the 20 000t of TNT Fatman had [26].

## 2.7 Different Technology level

When analyzing feasibility for building a Hypothetical Nuclear Explosive Device (HNED) one can divide the analysis in three different scenarios, weather the bomb maker has access to low, medium or high technology. Low technology refers to that of the earliest NEDs, which had an explosive lens made of Baratol and Composition B [12]. With medium technology one is capable of producing the NED using explosives with higher thermal conductivities, higher melting points etc creating faster and higher pressure waves utilizing explosives of higher technology. High technology refers to nuclear weapon states which have acquired their know-how with research and experiments, as for instance USA, France which are assumed to be able utilize even better explosives than those of a medium technology country [26]. The three different technology scenarios is

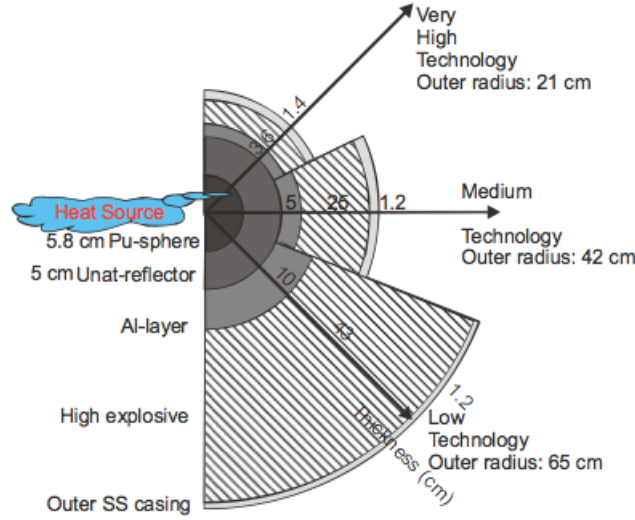


Figure 7: Schematic visualization of technology levels [26].

schematically be shown as in figure 7.

### 2.7.1 Heat source limits

For the different technologies defined in figure 7 Kessler et al. reaches the conclusion that reactor plutonium with more than 1.8%  $^{238}\text{Pu}$  corresponds to a heat source of 120W in a low technology NED due to alpha heating. Kessler et al. further shows that levels above 3.5%  $^{238}\text{Pu}$  corresponds to a heat source of more than 240W in a medium technology NED [26]. The reason those heat sources would make the plutonium proliferation proof is that such high heat generation causes the chemical explosive lenses surrounding the plutonium to partially melt or initiate self ignition. Due to the high requirements on simultaneous ignition of the chemical lenses (within  $10^{-6}\text{s}$  [26]) one can understand that a self ignition of the chemical lens would result in a much smaller yield. Together with the very high precision required in placement and composition of the chemical explosives in order to reach good yields, the effect of partially melted and malfunctioning lens would affect the explosive yield negatively.

## 2.8 Proliferators

Many of the steps in the nuclear fuel chain are of proliferation risk with different nature. For instance Uranium found in Venezuela that might be shipped to Iran may be seen as a major proliferation risk, while uranium found in Gabon might be of less concern while shipped to France and of bigger concern if destined for Iran or India [30]. However for mined uranium to be of risk, there is need for enrichment facilities. For instance were the Iraqi case in late 1980s and early 2000s under heavy surveillance since they had



pursuit their own enrichment facilities [30]. However, the biggest proliferation risks when it comes to the mining of uranium lay within "rogue states" having their own enrichment program. Which means that pure mining of uranium is not really one of the main proliferation risks. One of the most common used techniques when it comes to uranium enrichment is the gas centrifuge method, which utilize the differences in mass between  $^{235}\text{U}$  and  $^{238}\text{U}$ . To create an enrichment facility using this method, many centrifuges are connected together in a cascade connection. For optimal high enriched uranium the configuration of centrifuges are a bit different from standard configuration for commercial nuclear fuel. But the same configuration used for low enriched uranium ( $< 20\%$ ) which is used in commercial nuclear reactors could be used to create highly enriched uranium if one uses the product of a low enrich process to re-feed a low enrichment process again and again. One of the big drawbacks with highly enriched uranium bombs are the share size and weight. For instance it has been argued that Iraq, which got close to finishing a bomb using their own designs and enrichment efforts (as close as 3 years [31]) would probably ended up with a NED with a weight of at least one ton. Iraq devoted its effort to produce highly enriched uranium (HEU) bombs and began building complex research and production facilities which can easily cost 10 to 50 times more money then a plutonium weapon path [31]. If HEU is produced from a site, aiming to produce at least HEU for 10 or more bombs a year, it is believed that the produced heat will be observable, at least when using energy-inefficient technologies such as for instance gaseous diffusion. This is however not the case if higher technology would be applied, such as centrifuges [31].

When it comes to Nuclear fuel facilities Braun [30] particularly mentions proliferation concern about small national fuel cycle facilities which might not be justified on economic grounds. Such small facilities might attempt breakouts from the NPT (The Nuclear Non-proliferation Treaty) according to Braun. NPT is a treaty that entered into force 1970, which requires non-nuclear weapon member states to place all their nuclear material under safeguards, which is a system of materials accountancy, containment and surveillance administered by IAEA. The nuclear states declare to not in anyway assist non-nuclear weapon states to acquire nuclear weapons. Further by signing the treaty research on peaceful nuclear applications can freely be exchanged in between NPT states which can act as a driving force for non nuclear states, that has an interest in going nuclear to sign the treaty and therefor receive aid from already existing nuclear states. One of the major risks concerning proliferation is at the back end of the fuel cycle with reprocessing plants. In a reprocessing plant plutonium is separated from the highly active fission products which in a way acts as a barrier against theft, both due to heat production and lethal radiation levels. Those are under tight surveillance of multilayered materials protection control and accounting systems (MPC&A) safeguarding developed by IAEA. So the two main sources of proliferation risks lies within enrichment and reprocessing facilities. Another proliferation concern is early shutdown of a nuclear power plant, however the nuclear power plants is considered to be quite safe from proliferation view due to good surveillance and accountancy of booth spent and fresh fuel. Still there are fundamental limitations in the NPT, for instance as mentioned earlier, a state could

be of the NPT and amass nuclear weapon material under the safeguard system and then withdraw from the NPT. Another limitation worth mentioning is that even though member states declare all of their nuclear facilities and place them under safeguards, it does not give IAEA the right to hunt for undeclared facilities or the power to request special inspections and declared or undeclared facilities. For instance IAEA requested for a special inspection in North Korea 1993 due to suspicions of more separated plutonium than the revealed 100g, but this request was refused after which IAEA declared North Korea to be in violation of the NPT [31]. However, towards the end of the 90 days withdrawal period North Korea announced that they would "suspend the effectuation" of that withdrawal. And in the end of the year 2002, North Korea started to disable surveillance cameras and the 27 of december IAEA inspectors was ordered to leave the country and in 2003 North Korea left the NPT.

### 3 Method - Indicators

#### 3.1 Storage time - unAttractiveness

The reason for putting spent nuclear fuel in deep geological repositories is to isolate it from the environment while the radio toxicity decreases due to natural nuclear decay processes. In figure 5 the reference level is the radio toxic level of all uranium ore used in order to produce the fuel which was burned in order to achieve the nuclear waste. So by considering figure 5 again one can see the high total radio toxicity consist of different components. Depending on efficiencies in reprocessing plants together with the neutronics of the nuclear core, the isotopic content (see table 2) looks differently. Different isotopic composition results in different storage time. In order to calculate the isotopic evolution of the waste RadTox has been used on different waste. RadTox is a computer code which calculates radiotoxicity curves of spent nuclear fuel as a function of efficiencies of partitioning and transmutation. Partitioning is the process of separating the spent nuclear fuel, and since it is mostly plutonium isotopes which decides the storage times, see figure 5, even very small occurrences of plutonium can cause big differences in the storage time. The transmutation efficiency is the efficiency with which transuranium elements are burned in the reactor. Since it is the final value of transuranium elements in the waste that decides the time required for storage, the transmutation efficiency has a minor effect on the total radiotoxicity compared to the partitioning efficiency. In a way inferior transmutation efficiency can be compensated for, since if one can separate away much of the transuranium elements before storing the waste, one can mix them into another fuel batch. So this parameter has not been changed through the cases but kept at 20% efficiency. Within RadTox there are certain limitations of which reactor schemes that can be used. Because of those limitations four common reactor schemes has been chosen:

- 4% initial  $^{235}\text{U}$  enrichment burned to 45GWd/tHM that is kept 10 years in onsite storage pools for cooling.

- 4.2% initial  $^{235}\text{U}$  enrichment burned to 45GWd/tHM that is kept 10 years in onsite storage pools for cooling.
- 4% initial  $^{235}\text{U}$  enrichment burned to 65GWd/tHM that is kept 10 years in onsite storage pools for cooling.
- 1% initial  $^{235}\text{U}$  enrichment and 3%  $^{239}\text{Pu}$  burned to 45GWd/tHM that is kept 10 years in onsite storage pools for cooling.

The three considered waste handling cases which the fuel schemes are tested on are the following:

- Without any reprocessing at all.
- A prediction based on an expert (Professor Christian Ekberg) using 99% partitioning efficiency [32].
- A best case scenario with 99.9% efficiency in the partitioning process.

In figure 8 the radio toxicity evolution can be seen for all the covered cases based on 4% initial enrichment and a burnup at 45GWd/tHM.

Storage of radioactive waste should be according to the nine principles of radioactive waste management stated by IAEA [33]. They are as following:

- Protection of human health - Radioactive waste shall be managed in such a way as to secure an acceptable level of protection for human health.
- Protection of the environment - Radioactive waste shall be managed in such a way as to provide an acceptable level of protection of the environment.
- Protection beyond national borders - Radioactive waste shall be managed in such a way as to assure that possible effects on human health and the environment beyond national borders will be taken into account.
- Protection of future generations - Radioactive waste shall be managed in such a way that predicted impacts on the health of future generations will not be greater than relevant levels of impact that are acceptable today.
- Burdens on future generations - Radioactive waste shall be managed in such a way that will not impose undue burdens on future generations.
- National legal framework - Radioactive waste shall be managed within an appropriate national legal framework including clear allocation of responsibilities and provision for independent regulatory functions.
- Control of radioactive waste generation - Generation of radioactive waste shall be kept to the minimum practicable.

- Radioactive waste generation and management interdependencies - Interdependencies among all steps in radioactive waste generation and management shall be appropriately taken into account.
- Safety of facilities - The safety of facilities for radioactive waste management shall be appropriately assured during their lifetime.

The developed indicator for the storage time can be seen in equation (3), where  $R(0)$  is the ingestion toxicity at beginning of storage,  $R_{ref}(0)$  is the ingestion toxicity from 45GWd/tHM 4% LWR waste,  $t_{\text{reference level}}$  is the storage time needed to reach reference level,  $t_{LWR}$  the time it takes for 45GWd/tHM 4% LWR waste to reach reference level and  $f_1$  and  $f_2$  are functions which should mirror how the time perspective should be taken into account compared to the radiotoxicity.

$$unATTR = \frac{R(0)}{R_{ref}(0)} \cdot f_1(t_{\text{reference level}}) \cdot f_2(t_{LWR}) \quad (3)$$

In this thesis the indicator for the storage of waste, unATTR, is calculated according to equation 4 where  $W$  a weight function who reflects how we treat the responsibility of waste storage in the future. So if we actually think we want reactor waste in the future, this weight function should be increasing for longer storage times and decreasing for lower. The reason for taking the logarithms is to place changes in the time it takes to reach reference level comparable with the changes in radiotoxicity, since radiotoxicity levels changes with a few percentages while  $t_{\text{reference level}}$  changes with millions of years causing the time perspective to be dominant in all cases. The idea that the indicators catches the fundamentals in how hard the waste is to be managed properly. So it takes into account the initial radiotoxicity(after 10 years of cooling in on-site pools) and the total time the waste would need to be stored for until it reached the radiotoxicity of the reference level. This is made in a way where they are of about the same importance, i.e a change in the initial radiotoxicity should be comparable with an equal change in storage time. Reference level is the radiotoxicity level of the uranium ore before it was processed into fuel. The initial radiotoxicity is normalized with respect to the level of the waste from 45GWd/tHM 4% enrichment LWR fuel and the storage time is normalized to that of the time it takes for 45GWd/tHM 4% enrichment LWR fuel to reach reference level.

$$unATTR = \frac{R(0)}{R_{ref}(0)} \cdot \frac{\log(t_{\text{reference level}})}{\log(t_{LWR})} \cdot W(t_{\text{reference level}}) \quad (4)$$

The shape of the weight functions,  $W(t_{\text{reference level}})$ , is  $W(t) = e^{-rt}$  where  $r$  takes values 0.05, 0 and -0.05 as an attempt to catch whether reactor waste should be considered more or less attractive in the future.

### 3.2 Plutonium quality - Attractiveness

Spontaneous fission can cause a NED to pre detonate. Pre detonation occurs when chain reaction starts prematurely, for instance already in the compression phase when

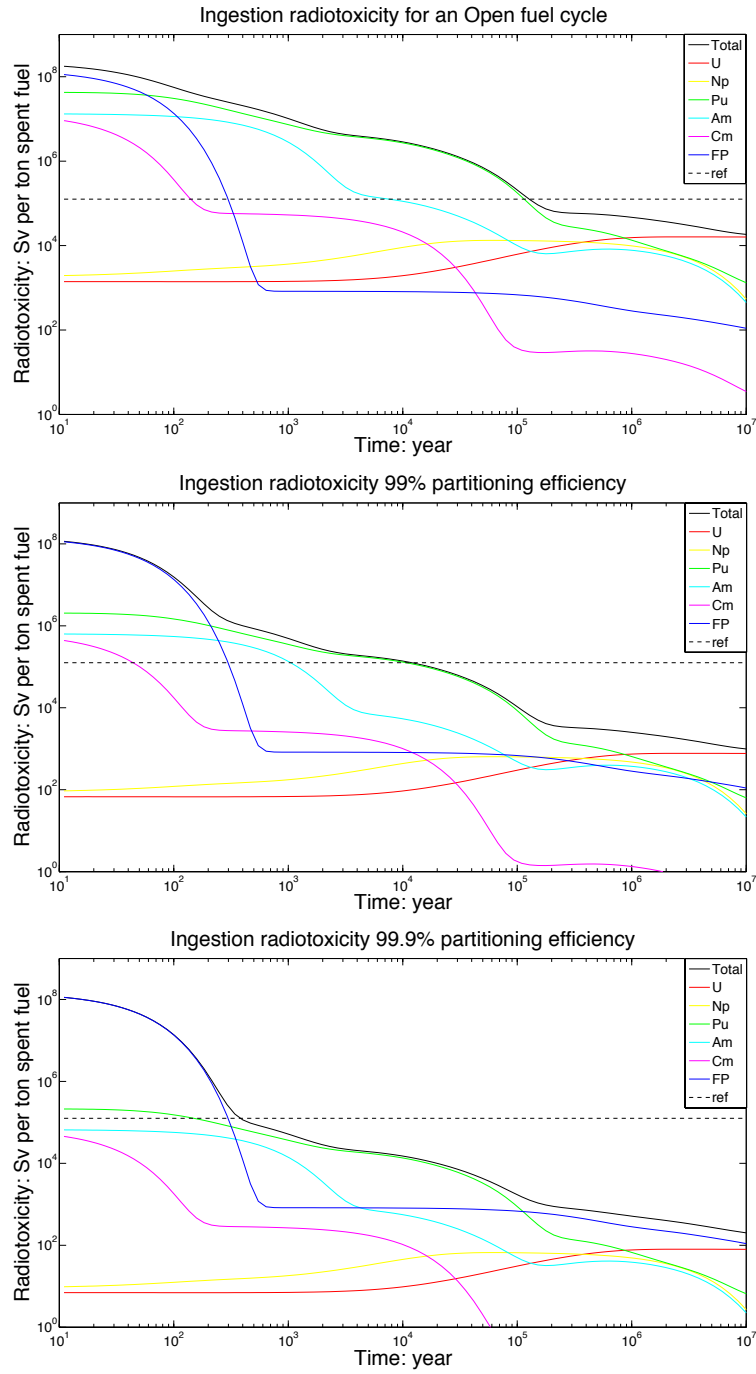


Figure 8: Figures showing ingestion radiotoxicity for different fuel schemes.

the pressure waves from the chemical explosives are traveling inwards to compress the plutonium sphere. If  $k$  denotes the number of direct descendant neutrons from an original fission event that did not escape the system, then  $\frac{(k-1)}{\tau}$  will be the rate of change of neutrons, if  $\tau$  is the mean lifetime of a neutron until it gets absorbed. So if  $N$  is the number of neutrons at time  $t$  the change of neutrons at time  $t$  would be  $\frac{dN}{dt} = \frac{(k-1)}{\tau} N = \alpha N$ . This differential equation gives the solution  $N(t) = N(0)e^{\alpha(t)}$ , where  $\alpha$  is commonly known as "Rossi alpha". The final explosion yield ( $Y$ ) has been shown to be proportional to  $\alpha_{max}^2 \cdot \Delta k_{max}$  at the instant the explosion starts, i.e  $Y \propto \alpha^3$  [34].

The excursion of a NED can be said to occur in two phases. The first phase ends with the boiling of plutonium at approximately 3255 degrees Celsius. This phase is usually considered to end after some 40-45 consecutive fission events releasing. With about  $e^{35}$  fissions the energy released is about one calorie per gram in 10kg of plutonium. This energy is only enough to increase the plutonium metal temperature by some 30 degrees [19]. At about  $e^{42}$  fissions the energy is about 1 kilocalorie per gram, which is that of an conventional explosives. Even though there are no way of exactly determine when phase two is initiated, it is commonly approximated to be at around  $e^{40} - e^{45}$  fissions [26], [19], [28], [35]. When the second phase is initiated the increased pressure due to plutonium gas and expanding core volume causes the reactivity of the core to decrease until it finally gets subcritical again. If one tries to solve an analytical model based on basic physics without even taking pre detonation into account, a third order ODE (Ordinary Differential Equation) arises. Which needs to be solved numerically to provide relevant information for explosive yield [36]. As can be understood the explosive yield from a NED is a very complicated thing and covers many variables.

Y. Kimura et al. states the non pre detonation probability to be according to equation (5).

$$P_{non-predet} = \exp\left[\frac{S_M}{\nu\Gamma_2}(-\Delta k_{max}T + 2N\tau)\right] \quad (5)$$

In equation (5)  $\Delta k_{max}$  is the maximum super criticality (no delayed neutron emissions needed to reach criticality) which occurs just before phase two starts which happens during a compression time  $T$  and  $\tau$  is the prompt neutron life time.  $N$  here is the number of consecutive fission events that need to occur before the explosion starts, which is often assumed to be 40-45 and  $S_M$  the neutron multiplication in a medium. The formula for the neutron multiplication is known from  $S_M = \frac{1}{1-k_{eff}} \cdot S$ ,  $\nu$  is the average number of neutrons released in a fission event and  $\Gamma_2$  the Diven factor which in a weak neutron source assumption is given as  $\Gamma_2 = \frac{\nu(\bar{\nu}-1)}{\bar{\nu}^2} = 0.714 + 0.035\nu$ . For a evaluation of the weak neutron source assumption see section A. Whats valuable for the indicator of the spontaneous fission rate here is that the pre detonation probability( $1 - P_{non-predet}$ ), is a function with the parameter  $S$ =spontaneous fission neutrons per second, as the most important parameter. Even though the compression time, which can be seen as a insertion of reactivity, and prompt neutron lifetime  $\tau$  are of some importance when one are to calculate the pre detonation probability [28].

Depending on technology level and plutonium quality Kessler et al. reaches different explosive yields for different technologies according to table 5. The plutonium grading

Technology	Yield (kt) of TNT equivalent
Medium technology	0.12
Very high technology	0.35
Fat Man	22

Table 5: Nuclear explosion yields for very high and medium technology for a reactor grade plutonium case with 5.5%  $^{238}\text{Pu}$  [26]. The explosive yield of Fat Man is shown as well for comparison.

Technology	Maximum alpha heating before melting, [W]
Low Technology	144
Medium technology	240
Very high technology	377-562

Table 6: Onsets for melting of the explosive lens surrounding a NED for different technological levels [26].

is based on the definition of United States Department of Energy and United States Nuclear Regulatory Commission [37]. The calculations made by Kessler et al. are based on absolutely perfect symmetry etc which one might argue is not achievable. Still those are significantly lower than the 22 kton explosion of the Fat Man bomb.

### 3.2.1 Alpha heating

Alpha heating is something that causes trouble when trying to create a bomb. Alpha heating mainly occurs with the presence of  $^{238}\text{Pu}$  see table 3, which will cause the fissile core will act as a heat source due to the decay heat generated by the alpha decays. When this heat source is too great the explosive lens surrounding the NED will partially melt or auto ignite. Using the results of Kessler et al. [26] with different technology level defined as in figure 7, the limits for the different levels can be seen in table 6. Worth to mention is that those limits are calculated with their own dimensions on the NED according to figure 7.

### 3.2.2 Attractiveness Factor

Lately there has been an increase effort in trying to grade and categorize plutonium. The general definition of Attractiveness(ATTR) is given in equation (6) which was proposed by Saito et al. 2007 [38].

$$ATTR = \frac{\text{Characteristics of Potential Explosive Energy}}{\text{Technical Difficulty for Fission Explosive Device Use}} \quad (6)$$

It was earlier in this thesis mentioned that explosive yield was proportional to  $\alpha^3$ , i.e

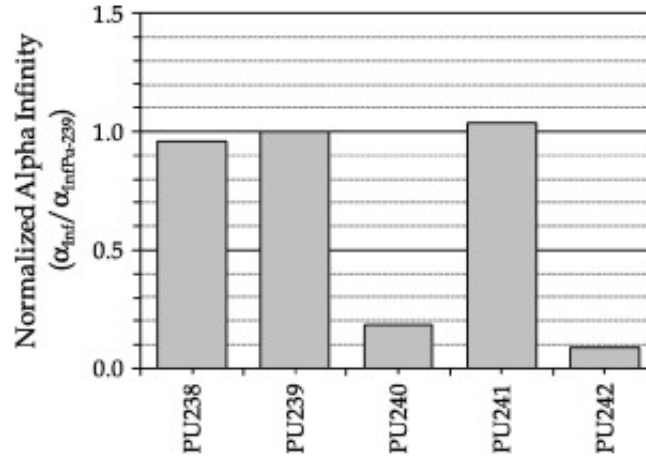


Figure 9: Values for normalized Rossi alpha [28].

the Rossi-alpha can be used as a representation for explosive properties which is in the nominator of (6). Furthermore the Rossi-alpha will be normalized to that of the best possible material when it comes to building an explosive,  $^{239}\text{Pu}$ . The Rossi-alpha for different compositions is calculated using a linear dependence with respect to percentage of a given isotope. This is not completely correct but has via personal contact with Kimura been concluded to be a good enough approximation. Normalized values of the infinite medium Rossi is shown in figure 9 and those relative values are used together with the linear dependence approximation to calculate the Rossi-alpha of a given composition. The Technical Difficulty for Fission Explosive Device use, the denominator in equation (6), is represented by two parts. First the spontaneous fission rate in a bare critical mass which will be normalized with respect to that of  $^{238}\text{Pu}$ . While the other part is the decay heat in a bare critical mass, this value is calculated using values from table 3. In equation (7) the full formula for how attractiveness is calculated throughout the result section. In equation (7) the index composition stands for whatever isotopic composition the waste would have.

$$ATTR = \frac{\left(\frac{\alpha_{\text{composition}}}{\alpha_{^{239}\text{Pu}}}\right)^3}{\frac{SNF_{\text{composition}}}{SNF_{^{238}\text{Pu}}} \cdot DH_{\text{composition}}} \quad (7)$$

In table 7 can the Attractiveness factor for 4 different classifications be seen. Those values can be used to be compared with against the Attractiveness of the spent nuclear fuel, so for instance if the Attractiveness of the spent nuclear fuel would be below  $1.9 \cdot 10^{-4}$  one could deem it to be safe according to the IAEA limit, even if this limit is calculated to another unit compared to that one given by IAEA.



Plutonium grade	Attractiveness factor
Super Weapon Grade	0.64
Weapon Grade	0.24
70% $^{239}\text{Pu}$ and 30% $^{240}\text{Pu}$	0.11
IAEA deems safe	$1.9 * 10^{-4}$

Table 7: Attractiveness for different plutonium gradation.

Scenario for waste	Open fuel cycle			
	r=0.05	r=0	r=-0.05	time in repository (Million years)
4% enrichment 45GWd/tHm 10y cooling	0.56	1	1.80	0.13
4.2% enrichment 45GWd/tHm 10y cooling	0.55	0.98	1.77	0.12
4% enrichment 65GWd/tHm 10y cooling	0.97	1.76	3.20	0.16
MOX fuel 4% enrichment 45GWd/tHm 10y cooling	1.60	3.21	6.45	1.1

Table 8: Table showing indicator values for open fuel cycle.

### 3.3 Support ratio

The ratio at which TRU elements changes with respect to each year can be calculated as  $S.R = \frac{\text{Mass of TRU in waste}}{\text{Mass of TRU in fuel}}$ . One can also use the same equation for calculating the net change of  $^{239}\text{Pu}$ . With the for equation support ratio one can calculated the change of transuranic material, so if this value would be less than unity, the amount of transuranic isotopes are decreasing. In this thesis the support ratio are used for the amount of plutonium.

## 4 Results and Discussions

### 4.1 Storage time

The results for calculating the unAttractiveness factor (unATTR) for an open fuel cycle scheme, case 1, can be seen in table 8. The boxes marked with gray in table 8 are the best options due to being the lowest value. The values are very similar between the 4% enrichment low burnup scheme, and the 4.2% enrichment scheme. But they are clearly lower than the 4% high burnup and the MOX fuel schemes. Therefore for an open fuel cycle, the indicator would separate 2 fuel schemes as superior.

For case 2, the 99% partitioning efficiency case (deemed to be most probable by Professor Christian Ekberg), table 9 demonstrates that MOX fuel receives the lowest

Scenario for waste	Closed fuel cycle 99% efficiency			
	r=0.05	r=0	r=-0.05	time in repository (years)
4% enrichment 45GWd/tHm 10y cooling	0.29	0.52	0.93	12000
4.2% enrichment 45GWd/tHm 10y cooling	0.29	0.52	0.93	16000
4% enrichment 65GWd/tHm 10y cooling	0.40	0.72	1.32	11000
MOX fuel 4% enrichment 45GWd/tHm 10y cooling	0.24	0.49	0.98	33000

Table 9: Table showing indicator values for closed cycle 99% partitioning efficiency.

Scenario for waste	Closed fuel cycle 99.9% efficiency			
	r=0.05	r=0	r=-0.05	time in repository (years)
4% enrichment 45GWd/tHm 10y cooling	0.18	0.32	0.58	380
4.2% enrichment 45GWd/tHm 10y cooling	0.18	0.32	0.58	430
4% enrichment 65GWd/tHm 10y cooling	0.24	0.43	0.79	370
MOX fuel 4% enrichment 45GWd/tHm 10y cooling	0.14	0.28	0.57	1500

Table 10: Table showing indicator values for closed cycle 99.9% partitioning efficiency.

indicator values of  $r=0.05$  and  $r=0$  while for  $r=-0.05$  low burnup, 4% enrichment and 4.2% enrichment receives lower values. Those lower values indicate that extended time in repositories are not beneficial (which is somewhat expected due to the increasing weight put to longterm radiotoxicity with  $r=-0.5$ ), so the MOX fuel scheme would not be the best option. However, the differences are very small for  $r = 0$  and  $r = -0.05$ . The only fuel scheme that stands out with larger differences in indicator values is the 65GWd/tHm burnup scheme.

For case 3, the best possible partitioning efficiencies, the results can be seen in table 10. Here the MOX fuel scheme is the best choice independent on how we want to treat the repository time. But in the case were long term storage is treated as an increasing negative, the indicator value are more or less the same at 4% enrichment 45GWd/tHm, 4.2% enrichment 45GWd/tHm and MOX.

Storing nuclear reactor waste includes many difficulties and costs, however unATTR is a simpler means with which to make comparisons to determine which reactor waste would be most preferable to store, taking into consideration storage time and radio toxicity on an equal basis. The precise form of this indicator is still somewhat unclear,

with regards to the weighting of time and radiotoxicity. It could be argued that with improved technology some radiotoxic waste could be more easily dealt with, or even desirable. If, for example long term waste is desirable, then the weighting function could be altered to meet the needs of this scenario.

To be considered, is a stated principle by IAEA, see section 3.1, that responsibility passed on to future generations should be kept at a minimum. So if the weight function increases for longer storage times yet another scenario might be achieved. Tables 8-10 demonstrate concrete examples using equation (4) with different values of "r" mentioned in section 3.1. The best choice is indicated by a grey box. Of interest is that the best choice changes depending on fuel cycle. With an open cycle the 4.2% enrichment is the best case across all three approaches for weighting function. But when reprocessing comes into play one can see that MOX fuel is considered better in the case where we consider nuclear waste a resource in the future as well as in the neutral case. But for the case when we really want to punish long term waste, i.e  $r < 0$ , it is normal LWR waste and 4.2% enrichment fuel that performs best. In a best case scenario with very high efficiency in partitioning, one can see that MOX fuel is the best choice in all three scenarios.

For sensitivity analysis a test was carried out where  $r$  was increased with a factor of 10. This does increase the value of the indicator, but for an open fuel cycle the 4.2% enrichment scheme is still among the best choices. For  $r = -0.5$ , where long term storage is undesirable, then the differences are quite small between 4.2% enrichment and 4% enrichment low burnup options, while long burnup option for 4% enrichment as well as MOX fuel are one order of magnitude larger i.e undesirable. When  $r = 0.5$ , which means reactor waste is considered desirable in a future, then an open cycle MOX fuel would be considered equal to a low burnup 4% enrichment and 4.2% enrichment. For a closed fuel cycle, with  $r = -0.5$  the 4.2% enrichment is favorable and for increasing  $r$  values a transition is made towards MOX fuel which can be understood since MOX fuel leaves waste with longest storage time. For higher values of  $r$ , the difference increases.

In the current form unATTR comparisons between values provide information on how the fuel schemes relate themselves to an open cycle with LWR waste. A value lower than unity means the fuel scheme performs better than an open fuel cycle for LWR waste. The performance is strictly determined by radiotoxicity and storage time. In order to make good use of unATTR the precise form to compare radiotoxicity to storage time should be established. This thesis explores how unATTR can be used generally. A better evaluated form is needed in order to make more consistent judgements. This thesis does not for instance, consider the difficulties related to storage of higher activities or the need for cooling such containers.

## 4.2 Spontaneous fission, Decay heat and Support ratio

Calculation of the spontaneous fission rates, normalized Rossi-alpha and decay heat for different waste scenarios, table 11, are used to calculate the ATTR values which can be seen in table 12.

If one compares the amount of decay heat (table 11) with the results from Kessler

Fuel scheme	Spontaneous fission rate ( $n/(g \cdot s)$ )	normalized Rossi alpha	Decay heat in bare critical mass [W]	rate of $^{239}\text{Pu}$	kg $^{239}\text{Pu}$ /ton waste at end of cycle
4% enrichment, 45GW/tHM, 10y cooling	355	0.81	331.4	66%	6.89
4.2% enrichment, 45GW/tHM 10 years cooling	342	0.81	316.8	67%	6.78
4% enrichment, 65GW/tHM 10 years cooling	526	0.74	634.6	58%	8.01
MOX 3% $^{239}\text{Pu}$ , 45GW/tHM 10 years cooling	827	0.59	885.6	34%	19.86
Super weapon grade Plutonium	27	0.98	37.2	> 97%	
Weapon grade plutonium	53	0.95	45.8	$\approx$ 94%	
70% $^{239}\text{Pu}$ 30% $^{240}\text{Pu}$	273	0.75	115	70%	
Safe plutonium (> 80% $^{238}\text{Pu}$ )	2080	0.96	5948	< 20%	

Table 11: Results of spontaneous fission rates, decay heat and rate of  $^{239}\text{Pu}$  mass.

et al. (table 6) it is clear that it would be possible to create a NED using 4% enriched uranium and 4.2% enriched uranium while using a burnup level 45GWd/tHM if one has access to very high technology. While the higher burnup case and MOX fuel generates too much decay heat making the construction of a NED unfeasible.

For a MOX fuel scenario with 3%  $^{239}\text{Pu}$ , 45GW/tHM and 10 years of cooling the TRU change according to the equation given in section 3.3 would be TRU change=0.46. For this result the indicator tells us that the change in transuranium elements are decreasing at a rate of almost 50% each year. This is a preferable scenario since transuraniums are of big concern when it comes to proliferation. Difficulties arise when considering this result with the indicator for storage time, unATTR. The MOX fuel cycle is not always the ideal choice. An open cycle, for instance, with 4.2% enriched fuel is best from a storage point of view, while MOX fuel would be the best choice from a proliferation point of view due to the destruction of transuraniums, validity is only maintained if the separation processes used to create MOX fuel are not considered (i.e ignore the fact that the separation process is closely connected to proliferation issues). If one only looks to the change in  $^{239}\text{Pu}$  one receives a  $^{239}\text{Pu}$  change of 0.22 indicating that the amount of  $^{239}\text{Pu}$  is reduced to a fifth of its original mass in every reactor year.

### 4.3 Scenario testing

For testing ATTR in context, four scenarios have been constructed which assume a constant linearly growth in nuclear reactor numbers. Initially there are 356 low burnup

Scenario	ATTR	Decay heat verdict
4% enrichment, 45GW/tHM, 10y cooling	$8.5 * 10^{-3}$	Possible with high technology
4.2% enrichment, 45GW/tHM 10 years cooling	$9.6 * 10^{-3}$	Possible with high technology
4% enrichment, 65GW/tHM 10 years cooling	$1.9 * 10^{-3}$	Impossible
MOX 3% $^{239}\text{Pu}$ , 45GW/tHM 10 years cooling	$3.0 * 10^{-4}$	Impossible
70% $^{239}\text{Pu}$ 30% $^{240}\text{Pu}$	0.011	Impossible
Super weapon grade Plutonium	0.64	Possible with low technology
Weapon grade plutonium	0.24	Possible with low technology
Safe plutonium ( $> 80\% ^{238}\text{Pu}$ )	$1.9 * 10^{-4}$	Impossible

Table 12: Attractiveness for different plutonium gradation.

LWRs. Due to lack of data on fast breeder reactors they have not been included.

- Scenario 1: The number of low burnup LWRs increase from present number 356 to 1000 by year 2100. High burnup LWRs increase from 0 to 1000 by 2100. Number of MOX reactors stays at 0.
- Scenario 2: The number of low burnup LWRs increase from present number 356 to 600 by year 2100. The number of high burnup LWR increase from 0 to 1200 by year 2100. MOX reactors increase from 0 200.
- Scenario 3: The number of low burnup LWR stay constant at present number 356 to year 2100. The number of high burnup LWR increase from 0 to 1244 by year 2100. MOX reactors increase from 0 400.

The results from the three mentioned scenarios can be seen in figures 10 - 15. One can see that plutonium inventories are increasing over the whole period of time, which is expected since there are no plutonium burning reactors in use (figure 10). It can also be noted that the number of reactors are increasing every year, along with the number of needed enrichment plants (figure 11). Since no reprocessing is needed for Scenario 1, no reprocessing plants are built over the period. For scenario 2 one can see that the plutonium inventories will be slightly lower, which is expected due to the MOX reactors which are using plutonium as fuel. However, it is evident that 200 MOX reactors built over 90 years will not be sufficient to decrease plutonium levels which will be increasing over the 90 years modeled.

In scenario 3 (figure 15), one can see that in 2055 all plutonium in the system will have been converted to the quality of MOX fuel plutonium. Which is the lowest ATTR value the plutonium can reach in this model. This is however not low enough to be below the indirect limit set by IAEA acquired by converting plutonium containing more than 80%  $^{238}\text{Pu}$  to ATTR. It is also shown in figure 15 that at around 2055 the amount of  $^{239}\text{Pu}$  starts to decrease, and by the end of the modeled time the  $^{239}\text{Pu}$  amount is returned to the starting amount. Additionally, no  $^{239}\text{Pu}$  has been generated if one progresses through the full time model.

Even though no results represent an ATTR lower than the limit provided by the IAEA, keep in mind that the reactors used in this thesis are old type reactors where proliferation issues has not been focused on during the design. Furthermore, the MOX fuel has been treated the same at every step, but according to Walenius, the plutonium vector, changes with respect to which reprocessing generation the MOX fuel exists in [25]. Equation (8) is a ATTR calculation using the plutonium content shown in the third generation Pu vector (table 1), the normalized Rossi alpha values (table 9), the SFN rates, decay heat and bare critical masses (table 3):

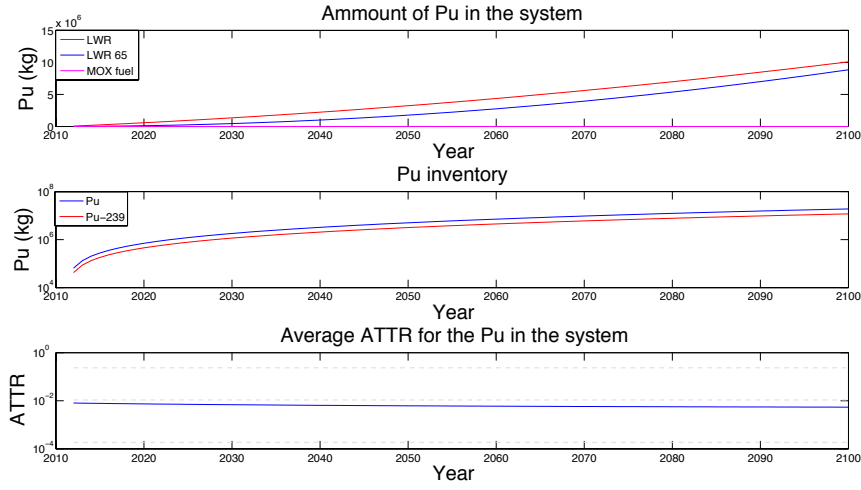


Figure 10: Graphs showing how plutonium inventories from different reactors, total Pu mass and the average ATTR changes in scenario 1.

Normalized Rossi alpha :

$$0.047 \cdot 0.95 + 0.405 \cdot 1 + 0.288 \cdot 0.19 + 0.131 \cdot 1.05 + 0.128 \cdot 0.1 = 0.65$$

BCM :

$$0.047 \cdot 13.1 + 0.405 \cdot 14.8 + 0.288 \cdot 44.8 + 0.131 \cdot 17.6 + 0.128 \cdot 87.8 = 33.06 \text{ kg}$$

SFN :

$$0.047 \cdot 2660 + 0.405 \cdot 0.0226 + 0.288 \cdot 1030 + 0.131 \cdot 0.0493 + 0.128 \cdot 1720 = 641.8 \text{ n/kgs}$$

DH :

$$33.06 \cdot (0.047 \cdot 567 + 0.405 \cdot 1.93 + 0.288 \cdot 7.06 + 0.131 \cdot 3.4 + 0.128 \cdot 0.12) = 989.3 \text{ W}$$

$$\text{ATTR} = \frac{0.65^3}{\frac{33.06 \cdot 641.8}{13.1 \cdot 2660} \cdot 989.3} = 0.00046 \quad (8)$$

The decay heat generated is higher than the limit given for very high technology (table 6), and the ATTR value is higher than the IAEA limit. However, if one calculates the ATTR for the Pu vector of the first generation (table 1) according to what was done on the third generation, one reaches  $\text{ATTR}_{1th} = 0.0014$ . This indicates that going from first to third generation, the attractiveness on the MOX fuel (table 1), decreased from 0.014 to 0.00046. Due to lack of data and limitations in RadTox, the ATTR of different recycle generations has been kept constant despite deducing from the above made calculations that this is not the case. So even though no technology has been investigated in this thesis that was able to fall below the IAEA limit, there might still exist the possibility for waste to reach below the IAEA limit.

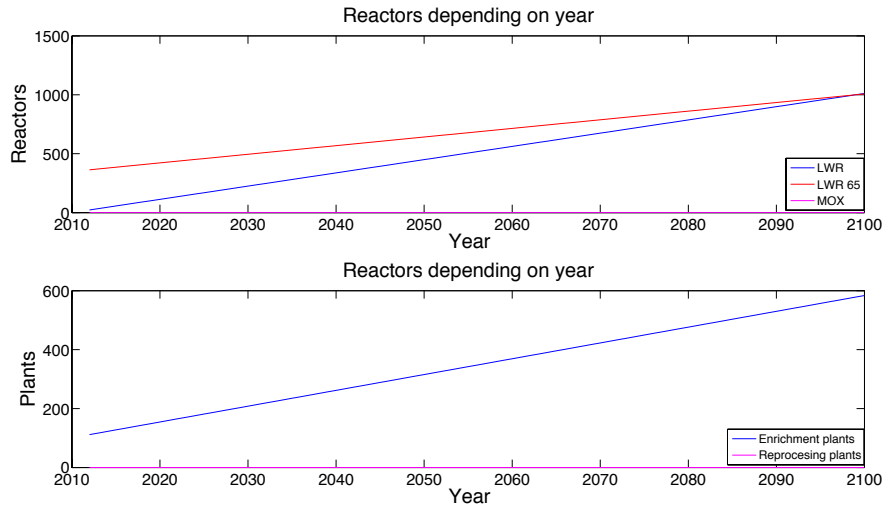


Figure 11: Graphs showing how reactors and enrichment plants will be built in scenario 1.

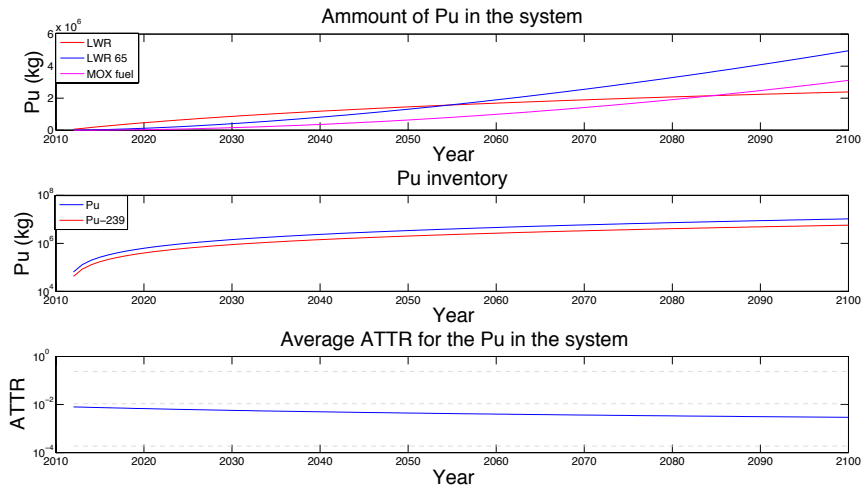


Figure 12: Graphs showing how plutonium inventories from different reactors, total Pu mass and the average ATTR changes in scenario 2.



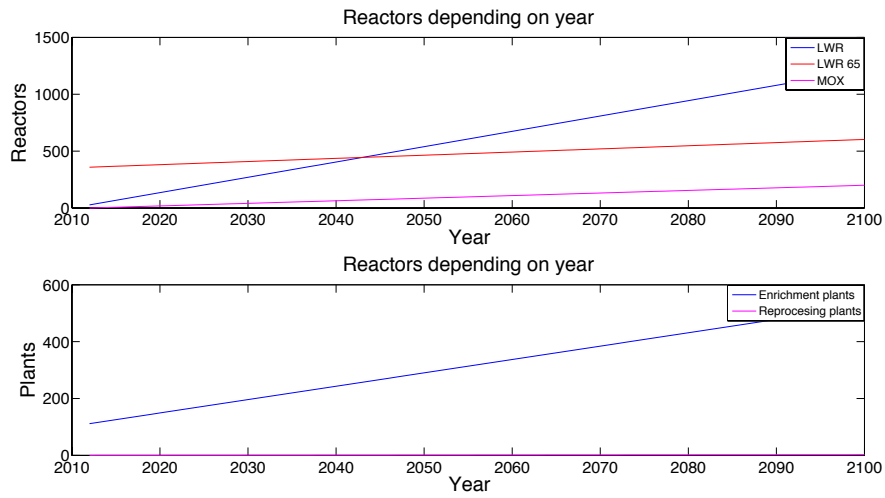


Figure 13: Graphs showing how reactors and enrichment plants will be built in scenario 2.

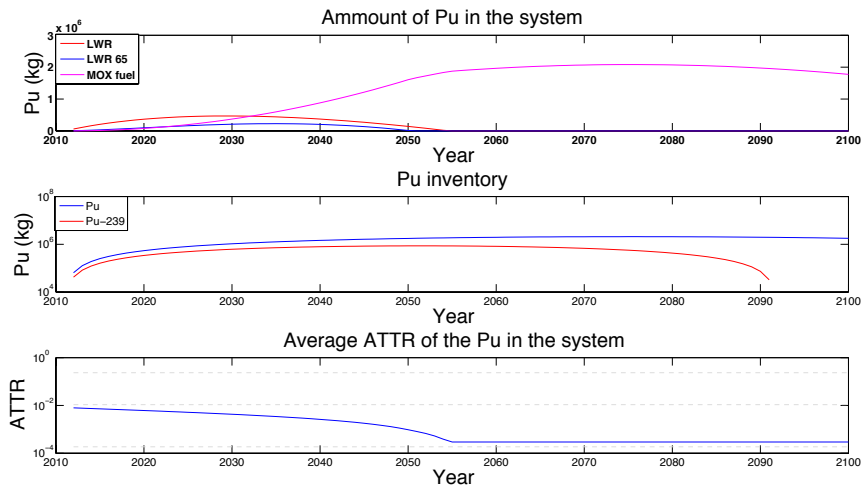


Figure 14: Graphs showing how plutonium inventories from different reactors, total Pu mass and the average ATTR changes in scenario 3.

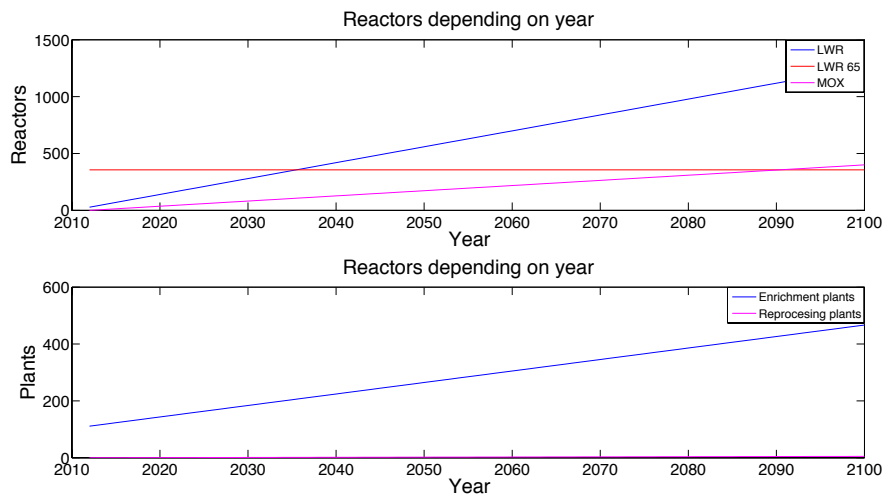


Figure 15: Graphs showing how reactors and enrichment plants will be built in scenario 3.

To use limits of usability for plutonium created in different types of reactors with respect to decay heat, only very high technology states, which are states already having NEDs, could construct a bomb using such plutonium (table 12). In the models used in this thesis there have been simplifications made, for example, that Rossi-alpha is linear with respect to composition and that the bare critical mass is linear with respect to composition, which is not accurate. These assumptions are made in agreement with experts of the area, PhD Yoshiki Kimura of the Japan Atomic Energy Agency for the linear mixture behavior of the Rossi-alpha and Professor Gunnar Skarnemark of the Nuclear Chemistry department of Chalmers University of Technology for the linear behavior of the bare critical mass. In order to decide whether plutonium created in different types of reactors is usable, potentially more precise models may be required. Another issue is the uncertainty around waste composition. Nuclear engineers are struggling to model and code how it will evaluate over time inside the nuclear core. With more precise data the indicator values would be more trustworthy.

Indicators should be further tested on more types of waste. Compositions and values for a generation IV reactor waste might generate lower values on ATTR. Unfortunately, during this thesis there was no such data obtained.

## 5 Conclusions

- To compare different reactor wastes in order to decide which type of reactors produces least attractive waste in the eyes of a proliferator one can calculate ATTR.
- MOX fuel was the modeled fuel which produced the lowest ATTR while higher burnup produces highest ATTR.
- In this thesis standard LWRs was used and none of them could produce a reactor waste with an ATTR low enough to be regarded as of non proliferation concern.
- When taking a more narrow approach, special limits of alpha heating into account, higher burnup and MOX fuel could be considered as impossible to use for a NED.
- When treating radiotoxicity and storage time to be of equal importance MOX fuel is the worst type of waste to store from an open fuel cycle.
- For a closed fuel cycle low burnup fuel and MOX fuel are considered to be equal.

### 5.1 Outlook

In order to improve the work done in this thesis concerning ATTR more reactor types should be evaluated. More advanced and specialized LWRs should be used instead of a basic category of LWRs since there might be a difference between pressurized water reactors and boiling water reactors. Further generation 3 and generation 4 reactors should be evaluated in order to be able to model future energy systems in which those types of reactor might exist in.

To improve unATTR one should at first determine the importance of the time perspective versus radiotoxicity since in this thesis they have been considered to be of about the same importance. Even here there is need to acquire data for more reactor types, as for instance different categories of LWRs, generation 3 and generation 4 reactors since the radiotoxicity and the storage time depends very much on the waste composition.

## A     Weak Neutron Source

This section is about the weak neutron source approximation which is an approximation used in some calculations. Equation 5 is for instance based on the assumption of weak neutron source, were the limit for a weak source is given by Hansen in equation 9. In equation 9  $\Gamma_2 = \frac{\nu(\nu-1)}{\bar{\nu}^2} = 0.714 + 0.035\nu$  is the Diven factor and  $\nu$  average neutron released by fission [39]. The formula for the Diven factor works when the emitted neutrons are binomial distributed but equals 1 for a high neutron background when neutrons are assumed to be emitted according to a poisson distribution instead [40].

$$2S\tau << \nu\Gamma_2 \tag{9}$$

As an example for what is a weak source, the limit can be calculated by considering that an average neutron travels  $\approx 15cm$  from birth to absorption in a fissile core containing plutonium [19]. Such a neutron has a kinetic energy of around 1Mev [26]  $E = \frac{mv^2}{2} \rightarrow 1.602 * 10^{-13} = \frac{1.66*10^{-27}*v^2}{2} \rightarrow v \approx 1.4 * 10^9 cm/s$ . Which give an average neutron life time as  $t = \frac{s}{v} \rightarrow \approx 10^{-8}s$ . This used together with equation 9 gives the limit for a weak neutron source as  $S = \frac{\nu\Gamma_2}{2*\tau} = \frac{2.3*(0.714+0.035*2.3)}{2*10^{-8}} = 9.13 * 10^7 n/s$ .

Typical critical mass for reactor plutonium is around 10kg [41] which needs to be taken into consideration when checking if a resulting reactor plutonium composition even for fills the weak neutron assumption. If not there is even wrong assumptions used assessing the pre detonation probability. Results for checking if the weak neutron approximation can be used can be seen in table 13. So strange enough it seems like the weak neutron approximation can not be used in any scenario trying to evaluate reactor waste.

Scenario	ratio to weak neutron source ( $\frac{S_M}{\text{limit weak source}}$ )
4% enrichment, 45GW/tHM, 10y cooling	1.94
4.2% enrichment, 45GW/tHM, 10y cooling	1.87
4% enrichment, 65GW/tHM 10 years cooling	2.88
MOX 3% $^{239}\text{Pu}$ , 45GW/tHM 10 years cooling	4.53
Super weapon grade Plutonium	0.15
Weapon grade pluto- nium	0.29
Safe plutonium ( $> 80\% ^{238}\text{Pu}$ )	11.39

Table 13: ratios to the weak neutron source approximation

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