Studies of Accelerator-Driven Systems for Transmutation of Nuclear Waste

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Abstract

Accelerator-driven systems for transmutation of nuclear waste have been suggested as a means for dealing with spent fuel components that pose potential radiological hazard for long periods of time. While not entirely removing the need for underground waste repositories, this nuclear waste incineration technology provides a viable method for reducing both waste volumes and storage times. Potentially, the time spans could be diminished from hundreds of thousand years to merely 1,000 years or even less. A central aspect for accelerator-driven systems design is the prediction of safety parameters and fuel economy. The simulations performed rely heavily on nuclear data and especially on the precision of the neutron cross section representations of essential nuclides over a wide energy range, from the thermal to the fast energy regime. In combination with a more demanding neutron flux distribution as compared with ordinary light-water reactors, the expanded nuclear data energy regime makes exploration of the cross section sensitivity for simulations of accelerator-driven systems a necessity. This fact was observed throughout the work and a significant portion of the study is devoted to investigations of nuclear data related effects. The computer code package EA-MC, based on 3-D Monte Carlo techniques, is the main computational tool employed for the analyses presented. Directly related to the development of the code is the extensive IAEA ADS Benchmark 3.2, and an account of the results of the benchmark exercises as implemented with EA-MC is given. CERN's Energy Amplifier prototype is studied from the perspectives of neutron source types, nuclear data sensitivity and transmutation. The commissioning of the n_TOF experiment, which is a neutron cross section measurement project at CERN, is also described.

Keywords: transmutation, accelerator-driven systems, Energy Amplifier, subcritical reactors, nuclear waste, Monte Carlo simulation, burnup, actinides, fission products, benchmark, nuclear data, neutron cross sections, sensitivity analysis, neutron source, spallation target, n_TOF experiment, neutron time-of-flight, cross section measurement, FLUKA, EA-MC, MCNP, MCNP-X, EADF, TRADE

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For my wife Guðrún
List of Papers

This thesis is based on the following papers, which are referred to in the text by their Roman numerals.


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**Author’s Contribution and Comments**

Papers I, II, III, VI and VII are based on analyses made by the thesis author and were also written primarily by the thesis author.

Paper IV was produced by the then existing Emerging Energy Technologies (EET) group of the SL Division at CERN, primarily written by C. Borcea. The thesis author participated in planning and data collection activities during the commissioning period for the n_TOF experiment.

Paper V was primarily written by A. Herrera-Martínez, and based on the methodology developed during the work on Papers II and III. The thesis author also contributed with proofreading and feedback during the preparation of the paper.

Papers II, IV and V are refereed, whereas Paper VII is currently subject to scrutiny by reviewers of the Elsevier Journal Annals of Nuclear Energy. The essentials of Paper VI are also planned for publication in a scientific journal.

Paper III, which is not refereed, offers a more comprehensive background to the analysis presented in Paper II. Some of the conclusions drawn in Paper VII exclusively rely on the more detailed parts of Paper III. On these grounds, Paper III is included in the thesis, although it essentially covers the same topics as Paper II.

The papers are ordered chronologically according to their respective publication dates.
Complementary Works Not Included in The Thesis

In addition to the work presented in the enclosed papers, the author has within the context of the nTOF experiment at CERN participated in the following publications.


C. Borcea et al., First Results from the Neutron (nTOF) facility at CERN. *CERN Report, CERN-SL-2001-070*.


C. Borcea et al., Commissioning Measurements of the n_TOF Spallation Neutron Source at CERN. *CERN Report, EET Note 2002-001*.


C. Borcea et al., Results from the Commissioning of the nTOF Spallation Neutron Source at CERN. *CERN Report, CERN-SL-2002-051*.  

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C. Borcea et al., First results from the neutron (nTOF) facility at CERN, Appl. Phys. A74 [Suppl.], S55-S57 (2002).


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1. Introduction

In light of recent development in energy pricing and availability, it is not difficult to understand the appearance of renewed interest for nuclear energy. As a serious candidate for supplying important contributions to the energy mix of the future, present-day nuclear power production would greatly benefit from the development of more comprehensive alternatives for dealing with long-term radioactive waste. Accelerator-driven systems for transmutation of nuclear waste have thus been suggested as a means for dealing with spent fuel components that pose potential radiological hazard for long periods of time. While not removing the need for underground waste repositories, this nuclear waste incineration technology provides a viable method for reducing both waste volumes and storage times. Potentially, the time spans could be diminished from hundreds of thousand years to merely 1,000 years or even less.

The research carried out within the framework of this thesis is part of the ongoing international development effort in the technology field of accelerator-driven systems for transmutation of nuclear waste. A central theme in most of the work that has been performed is nuclear data. Here the emphasis has been put on neutron cross section representations of essential nuclides in the fast energy regime.

Nuclear data is the foundation for calculation of the vital characteristics of any system based on multiplicative nuclear reactions, be it a conventional light-water reactor, a fast breeder reactor or an accelerator-driven system. Flaws in nuclear data will be reflected in the predictive accuracy for vital system parameters, which determine the economical and safety characteristics of the device. Therefore, the quantification and examination of nuclear data related errors should be paid due attention in the evaluation of multiplicative systems. The novel subcritical, source-driven systems also require nuclear data of high quality for wider energy ranges to produce reliable simulation results than what is the case for conventional, thermal reactor calculations. In combination with a more complex neutron flux distribution arising due to the presence of a high-yielding neutron source, the expanded nuclear data energy regime makes exploration of the cross section sensitivity for simulations of accelerator-driven systems a necessity. This fact was observed throughout the work and a significant portion of the study is devoted to investigations of the
neutron cross section sensitivity in different geometrical and computational configurations.

The computer code package EA-MC, developed by C. Rubbia and his group at CERN, is the main computational tool employed for the analyses presented within the thesis. Directly related to the development of the code is the extensive Yalina benchmark, or more formally, the IAEA ADS Benchmark 3.2, and an account of the results of the benchmark exercises as implemented with EA-MC is given in the thesis. Situated in Minsk, Belarus, the Yalina experiment forms the basis for the benchmark calculations. The hardware consists of a subcritical thermal assembly coupled to a neutron generator providing source neutrons from deuteron-deuteron and deuteron-triton fusion.

A simulation study of a neutron generator yielding source neutron distributions similar to the ones of the Yalina setup, but coupled to CERN’s Energy Amplifier prototype, is also described. Although such a configuration in reality is unphysical, since neutron generator technology is not capable of delivering the needed source fluxes, simulations thereof are feasible. The results enable assessment of the validity of conclusions drawn from experiments based on systems driven by fusion neutrons to those systems that employ proton-induced spallation source neutrons.

An important European effort within the transmutation field consists in the select nuclear data measurement projects funded within European Union Framework Programmes 5 and 6. One of these is the n_TOF experiment at CERN, the commissioning of which is described in part of this thesis. The goal of the n_TOF-ND-ADS project is to produce, evaluate and disseminate high precision cross sections for the majority of the isotopes relevant to nuclear waste incineration and accelerator-driven systems design. Directly associated with n_TOF, serving as supporting and advisory documentation, are also the cross section sensitivity studies described in the thesis.
2. World Energy Situation

2.1 Energy Consumption and Production

The International Energy Agency (IEA) predicts that the world’s energy consumption will increase by two thirds until 2030 and that electricity use will grow faster than any other energy end-use [1], see Figure 2.1. It is thus urgent to make plans for how this demand could be met with as sustainable energy forms as possible.

![Figure 2.1: World installed electricity generation capacity and expected increase until 2030 [1].](image)

The energy forms that dominate the market and currently are utilised for large-scale energy production are quite readily extracted and thus come at a reasonable cost; fossil fuel-based (coal, oil and gas), hydroelectric and nuclear energy. The timescale for the cost of each is different, the initial investment being larger for hydroelectric and nuclear, whereas the fuel cost itself dominates the cost for fossil fuel-based energy. The long-term alternatives are hydroelectric and nuclear power. Hydroelectric has the benefits of low maintenance and non-existent fuel expenses, whereas nuclear energy characteristically shows low fuel cost and very high power generation reliability.
2.2 Fuel Resources

2.2.1 Conventional Fuels

There are evident limitations in the availability of the fuel resources. The cheapest energy production form, hydroelectric, is in many areas a stable electricity supplier as long as precipitation stays reasonably stable from year to year. However, there are only a finite amount of rivers that can be exploited, so in this respect there is a limit for how much hydroelectric power can be produced, even if the fuel resource itself is inexhaustible. In fact, the maximum expansion capability has in practice already been attained in most of the industrialised countries.

The availability limitations particularly apply to the case of fossil fuels. Worldwide recoverable coal reserves are estimated to last for another 200 years at current rates of exploitation [2]. Concerning natural gas and oil, the values given are 60 and 40 years, respectively. It should be noted that these values are estimates, formed by simple division of the amount of reserves with the current production rate, i.e. no attention is paid to future growth in demand. Ultimately, as production declines and cost goes up, there will have to exist a replacement for oil as the most important energy source.

2.2.2 Nuclear Fuels

A similar line of argumentation as the above can be followed concerning the availability of uranium resources, but there are several reservations that need to be made. Firstly, uranium is abundant on many locations, forming about two parts per million of the Earth’s crust, which makes it 500 times more abundant than gold, 40 times as common as silver and slightly more abundant than tin. At a roughly estimated (and optimistic) average rate of consumption at 75,000 tonnes of uranium per year for the period between 2002 and 2041, it could be assessed that low-price reserves would only suffice until the end of the same period [3]. Still undiscovered resources have the potential of extending this period with a factor of three. However, it needs to be pointed out that the uranium price does not significantly affect the production costs of nuclear power, and thus even lower-grade uranium ore at higher effective cost can be utilised without significant economic consequences.

Secondly, the fissile isotope $^{235}$U is only present to 0.7% in natural uranium ($U_{\text{nat}}$), the rest mostly consisting of the fertile $^{238}$U, and the uranium must be enriched up to 3-5% in $^{235}$U for use in conventional light-water reactors (LWR)$^1$. In practical terms, this means that about six units of $U_{\text{nat}}$ is needed

$^1$The term fissile refers to isotopes that undergo fission by thermal neutrons, whereas fertile implies those isotopes that fission by fast neutrons only. Fertile materials can also be transmuted into fissile isotopes by means of neutron capture. The latter process is referred to as breeding.
for one unit of reactor grade uranium. However, if fertile isotopes were to be utilised in power production, the potential nuclear fuel resources become vast indeed.

Significant research and development efforts have been and are invested in systems that use fast neutrons, which has produced a flora of promising new concepts for fast reactors (FR) and accelerator-driven systems (ADS), see Chapter 4. It has been estimated that nuclear fuel resources could be extended by a factor 100 if a strategy including these systems were to be implemented [4]. Thirdly and lastly, there is an important alternative fuel, thorium, that could be employed for nuclear energy production[5, 6]. Thorium is three times more abundant than uranium in the earth’s rocks and soil, with economically recoverable world thorium reserves even surpassing uranium reserves.

2.3 Environmental and Social Considerations

When environmental issues and social acceptance are taken into consideration, the issue of future energy sources becomes more complex. Large-scale hydroelectric plants have an impact on local ecosystems and in some cases also on human habitation. Even smaller-scale plants affect local lake, river and/or marine ecosystems; particularly local fishing industry may suffer.

Common concern for carbon dioxide releases, with global warming as a probable consequence, has already incited world leaders to sign international agreements regulating carbon dioxide releases and the decrease thereof, most importantly the Kyoto Protocol that stipulates greenhouse gas emission limitations and reduction commitments for the signing countries. The climate policy adopted in the Kyoto Protocol is now a fact. It is thus clear that developed countries will not be able to unconditionally extend their energy use in the future with fossil fuel-based energy.

2.4 Nuclear Energy

The already existing option is nuclear energy, which offers the only long-term solution allowing for an economically sound expansion of large-scale energy production. Nuclear power in its turn has suffered from public distrust since the 1970’s, largely initiated by the reactor incident at Three Mile Island in 1979. The effect of the negative public opinion has perhaps in the end proved to be, if not useful, but essential to the nuclear industry. The safety standards adapted and implemented at modern nuclear installations by far supersede those of most other industrial sectors, and a culture of openness towards the public is actively cultivated. Thus the nuclear power industry of today is mature to take on a larger responsibility for the world’s energy production.
If it is proven that new generations of nuclear reactors can be operated under exceedingly safe conditions, e.g. by proven passive safety measures and multiple protection measures, then only one major obstacle remains from gaining thorough public acceptance, namely the issue of high-level nuclear waste. The predominant solution that countries like e.g. Finland, Sweden, Switzerland and the US have opted for, is the geological disposal alternative. While the method is well supported by extensive research and candidate sites undergo thorough geological survey, the general public tends to remain sceptical. Apart from such considerations, the proliferation issue is also of importance; the storage needs to be protected from future human intrusion, both intentional and unintentional.

The problem has lately been addressed by revived interest for accelerator-driven systems (ADS) for transmutation of nuclear waste, which are dedicated nuclear waste incineration systems having the potential to efficiently reduce waste volumes and shorten storage times from a million years to some hundreds of years. ADS addresses the safety aspects of burning nuclear waste since the system is source-driven and therefore can be operated in a subcritical mode with larger reactivity margins, cf. Section 4.3.4. Many of the suggested ADS concepts also employ passive safety systems concerning coolant flow and accelerator beam shutoff. In order for nuclear energy to provide an important contribution to the sustainable energy development of the future, successful implementation of ADS technology for nuclear waste treatment may prove essential, if not from a technical but from a public acceptance point-of-view.

\[^{2}\text{When using minor actinides as fuel, the reactivity margins are smaller. This is a cause for concern if fuels containing large amounts of minor actinides are introduced into critical systems.}\]
3. Nuclear Waste

3.1 The Composition of Nuclear Waste

The primary purpose of accelerator-driven systems is to destroy the most radiotoxic (cf. Section 3.2) and long-lived components of nuclear waste. Therefore, the composition of nuclear waste will be briefly reviewed.

Nuclear waste can primarily be classified as high-level (HLW) and low-level waste (LLW). HLW consists of highly radioactive fission and capture products arising in the nuclear fuel during operation, whereas LLW represents waste produced during operation of nuclear facilities, e.g. activated machine parts, structural materials, protectional clothing and residues from medical and industrial use of radionuclides. According to some classification schemes, a third category, intermediate-level waste, is also defined. Activated machinery and structure materials are in that case normally assigned to this intermediate class.

The concept of accelerator-driven transmutation of nuclear waste applies to HLW, or more specifically, to the radioactive products contained in the spent fuel. The constituents of HLW in their turn are often classified in two groups: transuranic elements (TRU) and fission products.

3.1.1 Transuranic Elements

TRU are products that arise from uranium and, if used, thorium isotopes by means of transmutation processes that occur in any operational multiplicative assembly. The most significant contribution of long-lived radiotoxicity to nuclear waste is yielded by TRU. Representative for the TRU category are the Pu isotopes and the minor actinides (MA) Np, Am and Cm. Table 3.1 gives an overview of the TRU masses in LWR spent fuel after 15 years of cooling time.

TRU are produced by way of transmutation-decay chains involving neutron capture as well as $\alpha$- and $\beta$-decay. Figure 3.1 schematically shows the main transmutation processes responsible for TRU buildup, (a) presents the chain beginning with the fertile $^{232}$Th and (b) the equivalent but starting from fertile $^{230}$Th.

1Some of the TRU are sometimes regarded as forming their own group of nuclear waste, due to the fact that they are not radioactive enough to strictly qualify as HLW.
Table 3.1: Transuranics in LWR spent fuel (40 GWd/ton U) after 15 years decay, from [7].

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Amount [g/ton]</th>
<th>Nuclide</th>
<th>Amount [g/ton]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Np-236</td>
<td>5.30E-04</td>
<td>Am-242m</td>
<td>2.50E+00</td>
</tr>
<tr>
<td>Np-237</td>
<td>6.50E+02</td>
<td>Am-243</td>
<td>1.40E+02</td>
</tr>
<tr>
<td>Pu-238</td>
<td>2.30E+02</td>
<td>Cm-242</td>
<td>5.90E-03</td>
</tr>
<tr>
<td>Pu-239</td>
<td>5.90E+03</td>
<td>Cm-243</td>
<td>4.30E-01</td>
</tr>
<tr>
<td>Pu-240</td>
<td>2.60E+03</td>
<td>Cm-244</td>
<td>3.10E+01</td>
</tr>
<tr>
<td>Pu-241</td>
<td>6.80E+02</td>
<td>Cm-245</td>
<td>2.30E+00</td>
</tr>
<tr>
<td>Pu-242</td>
<td>6.00E+02</td>
<td>Cm-246</td>
<td>3.20E-01</td>
</tr>
<tr>
<td>Pu-244</td>
<td>4.20E-02</td>
<td>Cm-247</td>
<td>3.70E-03</td>
</tr>
<tr>
<td>Am-241</td>
<td>7.70E+02</td>
<td>Cm-248</td>
<td>2.40E-04</td>
</tr>
</tbody>
</table>

$^{238}$U, while (c) depicts the beginning of the MA chain starting from $^{241}$Am and $^{243}$Am. Fission and capture thermal neutron cross sections are indicated, as well as $\beta$-decay half-lives. Although otherwise omitted, $\alpha$-decay is indicated in the cases of $^{242}$Cm, $^{243}$Cm and $^{244}$Cm due to the particular importance of these decay processes for the production of Pu isotopes (cf. Paper VII).

3.1.2 Long-Lived Fission Products

The two nuclei resulting instantly from nuclear fission are referred to as fission fragments. Figure 3.2 shows the distribution according to mass number of the fragments arising from fission, i.e. the fission yield, of some important fissile nuclei. The majority of the fission fragments are reasonably short-lived and rapidly decay into stable or long-lived nuclides, and these are referred to as fission products. In addition, the fission fragments that decay slowly may be classified as fission products.

Obviously, stable end-products are unproblematic from a radiotoxicity point of view. The long-lived fission products (LLFP), on the other hand, significantly add to the challenges of waste confinement technology, particularly due to the high solubility of some of their species. Out of the LLFP, the two major contributors to the long-term risk of HLW repositories can be transmuted, namely $^{129}$I and $^{99}$Tc. These two nuclides represent 95% of the LLFP volumes requiring long-term storage [9].

8
Figure 3.1: Main transmutation and decay chains starting from (a) $^{232}$Th, (b) $^{238}$U, as well as (c) the MA chains from $^{241}$Am and $^{243}$Am. Thermal neutron capture and fission cross sections, $\sigma_{th}$ [b], are given to the extent nuclear data admit. $\beta$-decay half-lives, $T_{1/2}$ with units given inside the figure, are also indicated. (For reasons of graphical clarity, $\alpha$-decay is not represented other than for the important Cm $\alpha$-decay.)
Figure 3.2: Fission yields of $^{233}$U, $^{235}$U and $^{239}$Pu at thermal (0.0253 eV) and fast (400 keV) neutron energies [8].
3.2 Radiotoxicity and Radiotoxicity Effects

The notion radiotoxicity has already been used in the above text. In fact, even without a definition the term is rather intuitive; in contrast to the purely physical measure radioactivity, it refers to the toxicity to living organisms of a particular radionuclide. It is defined as

\[ \text{Radiotoxicity} = A \cdot e, \tag{3.1} \]

where \( A \) stands for the activity and \( e \) for the effective dose coefficient. The activity is simply the number of disintegrations per second measured in units of Bq (1 Becquerel = 1 Bq = 1 disintegration per second = 1 s\(^{-1}\)). The damage caused to biological tissue by ionising radiation associated with the radioactivity of an isotope is quantified by means of \( e \) and measured in units of Sv/Bq.

The unit Sv, or Sievert, applies to the dose arising from the ionisation energy absorbed and the measure is referred to as equivalent dose. In other words, it includes a quality factor that describes the biological effect of the particular type of radiation deposited in a living organism. The Annual Limit of Intake (ALI) of an isotope is defined as the activity required to give a particular annual dose. An ALI value that has become something of an internationally recognised convention is the maximum annual dose for radiation workers, 20 mSv (0.02 Sv), i.e. \( \text{ALI} = (0.02 \text{ Sv})/e \).

The potential hazard index is defined as the ratio of the amount of a nuclide to its respective ALI value. Figure 3.3 shows the potential hazard index over time for HLW recovered from PWR spent fuel with a burnup of 33 GWd/t and 3 years of cooling [10]. The underlying calculations included the assumption of 99.5% uranium and plutonium recovery. In the figure, the potential hazard index of uranium ore required for production of the corresponding amount of fuel, 5 tonnes, is plotted for reference.

3.3 Options for High-Level Waste Disposal

Several techniques for disposal of HLW have been suggested over the years. Some early ideas even involved dispersion of the material into the atmosphere and oceans, a thought that today appears absurd, but that – considering the practices employed in fossil fuel handling – probably seemed viable at the time. Other ideas include burial in deep sea trenches, disposal in outer space, geological repositories, as well as partitioning and transmutation.
Figure 3.3: Potential hazard index of the HLW from PWR spent fuel as a function of time [10].
3.3.1 Storage in Underground Repositories

As was touched upon in Section 2.4, the most studied and widespread concept is the deep underground repository. The principles of the technique are quite independent of what kind of matrix is employed for the waste products. In countries employing a once-through fuel cycle (e.g. the US, Finland, Sweden, Switzerland) spent fuel in unmanipulated condition, i.e. as fuel bundles, is foreseen to be directly deposited. Countries that reprocess their waste will normally dispose of the waste in a vitrified or glass-like form.

A key factor to the method of HLW disposal in underground repositories coming to dominate the field is that it generally has been seen as the economically and technologically most readily available alternative. However, the waste only generates expense whilst in storage and deep underground facilities require significant initial investment costs, which implies that the economical aspect is far from clear-cut. Technological issues like confinement of highly volatile (water-soluble) LLFP during geological time perspectives represent considerable challenges for the technique, particularly in cases where the HLW is foreseen to be buried in an oxidising environment. Another issue vital to waste repositories is cooling of the waste materials; if the decay heat produced in the HLW can not be removed efficiently enough, the integrity of components and facilities may be threatened. Due to non-proliferation considerations and radiological safety, it will further be of high importance to protect repositories from human intrusion, both in the present and far into the future.

The farthest political and practical progress for building a final disposal facility for spent nuclear fuel has at the moment of writing been reached in Finland, cf. Figure 3.4. The concept is based on the KBS-3 method [11], developed by the Swedish Nuclear Fuel management Company, SKB. Construction of an underground research facility for rock characterisation for the final disposal began in 2004, with excavation work down to 420 m planned to be finished by 2008. Final disposal operation is foreseen to commence by 2020.

There is also another motivation to conduct extensive research in the field of deep underground HLW storages. Even if a fuel reprocessing and/or partitioning and transmutation (see Sections 3.3.2 and 3.3.3 below) policy is opted for, there will in the end be a need for storage of final waste, albeit that the volumes that need be buried are smaller and required confinement periods are shorter. A common misconception is that transmutation of HLW would entirely remove the need for underground repositories; the two technologies are rather to be seen as complementary to each other.

3.3.2 Fuel reprocessing

The HLW is contained in the spent fuel, which also is composed to more than 90 % of $^{238}\text{U}$. It is, from a radiotoxicity point-of-view, of interest to separate
the depleted uranium from the HLW, since its share of the volume is great and it contributes only modestly to the potential radiotoxicity, i.e. it would not need to be stored as rigorously as the HLW, at the same time as it is fertile and a possible fuel material for the future. This fact has led some countries, in particular France, Japan, the UK and Germany, to choose a fuel reprocessing strategy for their nuclear waste treatment, i.e. the uranium and plutonium is recovered from the fuel while the remaining HLW (consisting mostly of fission products and MA) is isolated in liquid and solid fuel reprocessing products.

Fuel reprocessing reduces the potential long-term radiotoxicity by a factor of 10 due to removal of plutonium and the waste volume to an even larger extent due to removal of uranium [13]. However, it then follows that the plutonium should be burnt, otherwise the procedure would only separate one type of waste from another. Currently, the world’s plutonium stockpile is only growing, to a large extent due to worldwide reductions of nuclear arsenals. Hence, the demand for waste incineration – and in particular plutonium burning – techniques is also increasing.
3.3.3 Transmutation and ADS

Transmutation is technically defined as any change of one nuclide into another. That is, it implies nuclear reactions, which change the number of and/or the identity of nucleons in a nucleus. In the particular context where transmutation is applied to nuclear waste, the signification is rather the one of nuclear reactions induced within human-made devices to produce stable (or more short-lived) nuclei from radioactive nuclei. A fuel cycle scheme employing dedicated transmutation devices is thus the HLW disposal option that has the potential of addressing both the issues of long storage times and HLW incineration.

One of the questions is then what type of transmutation device would be most suited for the task. It is possible to recycle plutonium in conventional LWR, but the advantages of plutonium incineration in LWR are limited [14]. Neither the natural uranium requirement nor the final radiotoxicity reduction are significantly improved. Also, MA nuclides show low fission cross sections at thermal energies (even the fissile ones). With a low fission-to-capture ratio, the LWR transmuter would rather be facing a buildup of MA than a reduction.

Fast reactors (FR) can be utilised to close the fuel cycle, but the number of FR needed to handle the MA amounts is large and hence a costly alternative for MA burning. Another drawback concerns safety; FR inherently operate with a short reactor period, and the insertion of MA fuel that has a small delayed neutron fraction further deteriorates control margins. FR were originally conceived for the purpose of Pu breeding. However, if the priority rather is to incinerate Pu, they can be optimised for this purpose instead. FR may thus be chosen as an integral part of an efficient transmutation strategy.

For MA transmutation, the most viable alternative is an implementation of accelerator-driven systems (ADS) technology, both from a safety and an economical perspective. Figure 3.5 shows the evolution of the potential hazard index (cf. also Figure 3.3 and Section 3.2) of HLW waste before and after transmutation [10]. Several variants of fuel cycles involving ADS burners exist: it is possible to run regular once-through cycles in base LWR stations and consequently separate the HLW and burn it in ADS, although the most economically appealing alternatives at present involve a FR stage as well (the Double Strata approach).

The techniques employed for separation of waste nuclides from the fuel and each other is generally referred to as partitioning. Essentially, partitioning resembles traditional fuel reprocessing, but extends to chemical processes specially designed for the purpose of further extraction of materials that are of interest for transmutation. These methods are still being developed [15] and are of utmost importance to the successful implementation of the transmutation technology, since the chemical losses in the partitioning step of the fuel cycle significantly affect the efficiency of any transmutation scheme.
Figure 3.5: Potential hazard index before and after transmutation of HLW as a function of time [10].
4. Accelerator-Driven Systems

4.1 A Brief History of ADS

The concept of transmutation dates back as early as 1919, when Rutherford first transmuted $^{14}\text{N}$ to $^{17}\text{O}$ using energetic $\alpha$-particles. Following the development of high power accelerators in the 1940’s, the first large-scale proposal for producing neutrons by spallation with an accelerator was made by Lawrence in 1950. The project was code-named Material Testing Accelerator (MTA) [16], but the actual aim was to produce plutonium from depleted uranium. The MTA project was abandoned after four years and after the 60’s, spallation-driven transmutation received little attention until the late 70’s and early 80’s, when interest was renewed, only then with rather the opposite objective: to reduce nuclear waste, i.e. to burn plutonium and minor actinides. A series of studies on partitioning and transmutation (P&T) were carried out at Oak Ridge National Laboratory [17]. The findings and recommendations of the ORNL studies are largely valid even today, and may in such a sense be seen as the foundation for modern P&T and ADS research activities.

In the early 90’s, particularly in the US and Japan, the ADS field received attention once again. The driving forces were growing plutonium and defense waste stockpiles, the evolution of high-power accelerators, as well as studies pointing towards issues with water-soluble radionuclide migration in the oxidising environment of the US Yucca Mountain repository. Since a group of CERN scientists led by Carlo Rubbia in 1993 proposed the first Energy Amplifier concept based on the thorium cycle [18], a number of research groups around the world have worked intensely within the field of accelerator-driven systems (ADS), accelerator-driven transmutation of waste (ATW) and hybrid systems, which are all different variants of systems based on an accelerator-driven spallation source coupled with a subcritical core.

4.2 The ADS Concept

In the exploratory phase of ADS development, both thermal and fast neutron systems were suggested. However, as simulation tools improved and the interest in waste incineration grew (with energy production becoming a benefit rather than primary target), systems employing fast neutrons have been estab-
lished as standard design, since only modest radiotoxicity reductions can be reached with a thermal system, cf. Section 3.3.3. Another specialty is the core of an ADS, which is *subcritical* (cf. Section 4.3.4) and must be driven by externally produced neutrons. As the name indicates, an ADS makes use of an accelerator to drive the multiplicative processes. The accelerator is used for delivering high-energy projectile particles, normally protons (with a kinetic energy of typically 1 GeV), which in turn are capable of producing source neutrons via nuclear intranuclear cascade processes in a *spallation* target, thus providing the external neutron source. The basic concept scheme for an ADS is depicted in Figure 4.1.

![Figure 4.1: Basic concept of an accelerator-driven system](image)

As is seen from Figure 4.1, the ADS power production scheme is fairly similar to the one of conventional electricity generation. The heat produced in the fuel is transported with the coolant to heat exchangers, where steam is produced to drive a turbine and consequently an electricity generator. The difference is that part of the electricity must be fed to the proton accelerator to keep the external neutron source running. The core structure itself is also fairly similar to classic ones; many ADS concepts envisage the fuel loaded according to well-known and proven technology, in fuel rods.

The fuel composition, with high Pu and/or MA content, naturally distinguishes the system from conventional nuclear reactors. Another vital difference is the coolant material, which must be sufficiently transparent to neutrons. Since the system is desired to work with fast neutrons, the neutrons need to retain a large part of their energy after each collision. The most commonly suggested coolant medium owning such properties is a lead-bismuth eutectic mixture (LBE), although some proposals have involved sodium-cooled and
gas-cooled systems.

In this work, the fast systems studied employ only LBE, and it is henceforth the coolant material that will be accounted for unless otherwise stated. The thermal systems of TRADE and Yalina (cf. Papers V and VI) are an obvious exception. The majority of the work presented within this thesis is based on the particular ADS reference configuration known as the Energy Amplifier Demonstration Facility\(^1\) (EADF) [19]. The system was developed with the Energy Amplifier (EA) as the conceptual basis [5]. The EADF is described in detail in the papers and the references.

4.3 ADS Physics

The physics of an ADS is distinguished from that of critical reactors by the presence of an external spallation target acting as a neutron source and the subcriticality of its core. In principle, other neutron sources than spallation sources can be employed for driving subcritical cores. Fusion neutron sources based on deuteron-deuteron (DD) and deuteron-triton (DT) fusion reactions, so-called neutron generators, are commonly used alternatives, cf. Papers I and VI. These sources are, however, limited with respect to neutron yield and hence also with respect to the source flux they are able to produce (within reasonable economical constraints), although they may be perfectly suited to drive smaller-scale experimental configurations. The scope in this summary will be limited to spallation neutron sources.

4.3.1 The Spallation Process

The concept of nuclear spallation is not a clear-cut physical process and thus lends itself only to a somewhat ambiguous definition. It implies a collection of nuclear reactions, in which the energy of every incoming particle is so high that more than two or three particles are expelled from the target nuclei under the change of both their masses and atom numbers. Figure 4.2 gives a schematic representation of the spallation-fission process according to modern understanding [20].

The initial collision is followed by an \emph{intranuclear cascade}, which implies that individual nucleons or small nucleon groups are ejected. Subsequently to the cascade, the excited nucleus emits further nucleons to reach its ground state. Virtually any nucleus of smaller mass number than the target nucleus situated on the neutron-poor side of the line of stability, and most of the lighter nuclei, can be produced by spallation [21]. These remains of the target nucleus, the stripped residual nucleus, are referred to as a \emph{spallation product}.

\(^1\)The EADF is sometimes also referred to as the Energy Amplifier Prototype (EAP-80).
Depending on target material and the kinetic energy of the incoming particle, the number of emitted particles – particularly neutrons – may be large. The ratio of emitted neutrons to protons impinging on the spallation target is referred to as spallation yield and is, insofar that it dictates the required accelerator power, of utmost significance to the economy of an ADS. By calculating the ratio between spallation yield and proton kinetic energy, an optimum proton kinetic energy for a target type may be obtained. The optimum is in the vicinity of 1 GeV for LBE targets.

4.3.2 The Spallation Target

The spallation target and its surrounding structural materials are the components exposed to the heaviest strain in an ADS. The target needs to be designed for efficient heat removal, since it receives a proton beam of several MW and the subsequent spallation processes release significant amounts of energy. In order to solve this issue, external cooling by means of liquid metals or gas has been envisioned. Furthermore, supportive structures for the target has to be highly resistant to high-energy proton and neutron irradiation.
As an example of the stress imposed on the target, the accelerator current required to drive a full-scale ADS design (~1500 MW\textsubscript{th}) – with a 1-GeV proton beam, as suggested in Section 4.3.1 – would typically be 10-100 mA, depending on the point in the production cycle and the exact power output of the facility. This implies a beam power of tens of MW impinging on the spallation target.

While these requirements must be fulfilled, the target also has the task to reliably provide as high a spallation yield as possible. The materials suggested for spallation targets include lead-bismuth, lead, tungsten, mercury, uranium and tantalum in liquid form, of which the main candidates are lead and lead-bismuth. Liquid metal has been chosen as a spallation target material for high-power conditions (in the MW range) owing to excellent heat transfer capabilities and reduced mechanical constraints. The main challenges in spallation target design are the corrosive/erosive properties of liquid metals, the intersection between beamguide and target, as well as removal systems for spallation products.

The radiation damage conveyed to surrounding structural materials is directly determined by the spallation neutron source spectrum, cf. Paper I. It should be noted that there is a marked difference between the two terms spallation neutron source spectrum and spallation neutron spectrum, the former refers to the energy spectrum in the source material, after moderation in the target material, and the latter refers to the spectrum of neutrons emitted in the spallation process. In principle, the harder the source spectrum is, the more damage the neutrons will cause. The spallation neutrons are produced at a relatively high average energy (3-4 MeV), with the highest energies ranging up to almost that of the incoming protons. The spallation neutron source spectrum is considerably harder than the one in the ADS fuel, due to the fact that fission neutrons dominate in the fuel and these are produced with a considerably lower average kinetic energy (around 2 MeV) than spallation neutrons, cf. spectra in Paper III. Further on, Paper I shows what impact the source spectrum has on system parameters and neutron balance.

4.3.3 The Subcritical Core

In a subcritical core, the neutrons produced by fission are too few to sustain a nuclear chain reaction. In other words, the exact balance between produced and absorbed neutrons found in a critical device is not present, i.e. the system will in comparison be deficient in fission neutrons. This deficit of neutrons must be compensated by the spallation neutron source, the intensity of which will be dependent of how deeply subcritical the core is.

The central aspect of ADS is its subcriticality feature, which allows for larger reactivity margins during operation, independently of the delayed neu-
tron fractions, $\beta$, of the fuel material. In general, MA and plutonium both share the property of having markedly smaller $\beta$ values than uranium; reactor safety considerations would seriously limit the loading of MA in a critical fast reactor (FR) core, cf. the discussion in Section 3.3.3. Due to this fact and the FR’s generally positive reactivity coefficients without $^{238}\text{U}$, i.e. with an absence of significant resonance broadening effects in the capture cross-section of $^{238}\text{U}$, it is virtually impossible to burn MA in the FR unless adding a significant quantity of $^{238}\text{U}$. Such a measure should be avoided, however, since it would mean further buildup of MA and breeding of more plutonium from $^{238}\text{U}$. With an ADS, on the other hand, such problems can be avoided, since its safety is guaranteed by the design subcriticality margin. It can thus be generally stated that in order to handle uranium-free transmutation fuels, ADS implementation is a prerequisite.

The neutron energies in the ADS fuel are high, with average and median neutron energies of 150-200 keV, although depending on the fuel matrix these values may vary. It is vital to obtain a hard spectrum in the fuel in order to provide favourable conditions for the direct fission of even neutron-number (fissionable) TRU nuclides like $^{237}\text{Np}$, $^{240}\text{Pu}$ and $^{242}\text{Pu}$. The fission neutron cross sections of these are in the range of a few barns over 500 keV, similar to the one of $^{235}\text{U}$ in this energy region.

### 4.3.4 Multiplication Factors and Source Importance

The effective neutron multiplication factor, $k_{\text{eff}}$, is generally employed for measuring the criticality in a given system. It indicates whether a nuclear chain reaction occurring in the system will tend to decrease or increase (as well as at what rate this will happen). If $k_{\text{eff}} < 1$, less than one neutron per fission (on average) survives to cause another fission and the system is subcritical. When more than one neutron survives, then $k_{\text{eff}} > 1$ and the system is said to be supercritical. Only when $k_{\text{eff}} = 1$, exactly one fission neutron survives to produce another fission, the system is critical, which is representative of the situation maintained during operation in current-day commercial reactors.

Thus, $k_{\text{eff}}$ is an intrinsic property of any multiplicative system and applicable to ADS. For ADS, it is appropriate for description of variations in the configuration of the system, such as in geometrical setup and material composition. However, it is not sufficient for describing the multiplication of the subcritical system in its source-driven mode.

Furthermore, if the fraction of neutrons with energy below 10 keV shrinks, the enhanced neutron capture due to resonance broadening, i.e. the Doppler effect, becomes much less effective [22].
Instead, we may define the net multiplication factor during \( m \) generations

\[
M_{\text{src}} = 1 + k_1 + k_1 \cdot k_2 + \ldots + \prod_{i=1}^{m} k_i ,
\]

(4.1)

where \( k_i (i > 0) \) is the ratio of the number of neutrons between neutron generations \( i \) and \( i - 1 \). In this manner, the impact of the source neutrons is also included, i.e. the multiplication characteristics of the system is described in terms of the number of neutrons released in multiplication reactions per source neutron.

If the multiplicative system is close enough to criticality and its multiplication consequently fission-dominated, the approximation

\[
k_1 \approx k_2 \approx \ldots \approx k_i \approx \ldots \approx k_m \approx k_{\text{src}}
\]

is valid. Furthermore, if the system is subcritical, then \( |k_i| < 1 \) holds true, and Eq. 4.1 may then be interpreted as a geometric series, and hence,

\[
M_{\text{src}} = \frac{1}{1 - k_{\text{src}}} .
\]

(4.2)

A redistribution of the terms yields

\[
k_{\text{src}} = 1 - \frac{1}{M_{\text{src}}} ,
\]

(4.3)

and a definition of the source multiplication factor, which accounts for the position and energy spectrum of the source neutrons, is found.

By definition, a constant power operation requires \( \bar{\nu}/k_{\text{eff}} \) neutrons per fission, where \( \bar{\nu} \) denotes the average number of neutrons released per fission. This means that an external source has to provide a number of neutrons per fission that is

\[
\mu_{\text{eff}} = \bar{\nu} \cdot \left( \frac{1}{k_{\text{eff}}} - 1 \right) = \frac{\bar{\nu}}{M_{\text{eff}} - 1} ,
\]

(4.4)

where \( M_{\text{eff}} \) is the net multiplication factor due to fission. In the case of an arbitrary external source, this number becomes

\[
\mu_{\text{src}} = \bar{\nu} \cdot \left( \frac{1}{k_{\text{src}}} - 1 \right) = \frac{\bar{\nu}}{M_{\text{src}} - 1} .
\]

(4.5)

The ratio

\[
\phi^* = \frac{\mu_{\text{eff}}}{\mu_{\text{src}}}
\]

(4.6)

is known as the neutron source importance or efficiency, cf. Paper VI. \( \phi^* \) gives an effective number of neutrons per fission and thus contains a correction
for non-fission multiplicative processes such as (n,xn) reactions, which are of
great importance in lead-bismuth cooled fast reactors.

4.3.5 Numerical Calculations

Numerical calculations of ADS system parameters and simulation of the sys-
tem behaviour requires special tools. A conventional nuclear reactor simula-
tion code is not directly applicable for a number of reasons [23], of which
perhaps the most important is the spatial distribution of the neutron flux de-
creasing in an exponential manner in the radial direction out from the spalla-
tion source in the centre of the core. In a critical reactor, the flux distribution
is instead essentially of a cosine form and determined by the geometrical con-
straints of the setup [24]. The neutron source, its geometrical form and the
associated spallation processes have to be simulated correctly in order to de-
termine the flux distributions, and therefore deterministic few-group diffusion
theory-based codes are generally not sufficient for the task.

The most suitable simulation method for the purpose of exploratory and yet
accurate calculations of ADS is provided by Monte Carlo (MC) techniques.
MC methods are statistical to their nature, and their basic principles are not
difficult to fathom. Given that a system and the processes present therein can
be described by probability density distributions, MC techniques can be ap-
plied for obtaining information about the system. Basically, a random number
generator of good quality\(^3\) is needed for sampling randomly over the distri-
butions. For instance, some of the important distributions that need to be rep-
resented and sampled over in ADS MC calculations are the probabilities for
different reactions to occur (cross sections) and interaction lengths. What must
be done is thus to insert enough incoming particles (protons or neutrons) to
describe the system with reasonable statistics, follow each particle throughout
its course and reactions in the system until it is consumed or exits the system.
When new particles are produced in a reaction, these must naturally be fol-
lowed as well. At each step a random number is sampled and applied to the
appropriate probability distribution, which yields the fate of the particle until
the next sampling step, e.g. until the next interaction occurs. The description
of the MC procedures applied to ADS given here is only schematic, a more
refined description necessarily involves a thorough description of the statisti-
cal evaluation of the results. This, however, falls outside the scope of this
summary, but more in-depth descriptions can be found in [25] and [26].

A major advantage with MC is that point-wise cross sections can be
employed within the sampling process (no division of neutron energies into
groups is needed, as opposed to the case of deterministic methods), which

\(^3\)Advanced random (quasi-random) number generation is generally available as a standard fea-
ture in modern Unix-based computing environments.
enables full treatment of cross section resonances and a detailed description of the neutron spectra in the system. Furthermore, the spatial resolution is only limited by the arithmetic precision implemented in the programming code and/or computing platform, and may thus in principle be considered infinite. However, it is obvious that the following of a large number of inserted and generated particles throughout their life within the system requires a lot of calculational steps and thus processing power. This has traditionally been the most significant limitation of MC methods, but with recent and expected future advancements in processing power, the issue is becoming less and less pertinent. Another limitation, which MC share with the other methods, is the quality of nuclear data, cf. Chapter 5.

4.3.6 The Monte Carlo Code Package EA-MC

There are only a few codes that are suited particularly for ADS simulation. One of the most specialised computer code package for the application, EA-MC, was developed by C. Rubbia and his group at CERN. The EA-MC simulation code package integrates neutron transport and evolution of the material composition in the same code [27], i.e. it has fully integrated and parallelised burnup simulation capabilities in addition to the standard steady-state calculation option (cf. Paper VII). All simulation-based analyses presented within this thesis were performed with EA-MC. In the cases where spallation neutron sources were applied to the subcritical systems, EA-MC was used in combination with the high-energy physics code FLUKA [28, 29]. Due to the fact that current nuclear data (which is the data available to EA-MC) generally exists only up to 20 MeV, FLUKA and its advanced theoretical models are needed for carrying out particle transport at higher energies. The EA-MC/FLUKA codes are described more thoroughly in the included research papers and references. On a general note, some other codes applicable to ADS are the Monte Carlo codes MCNP [30] and MCNPX [31, 32], as well as the deterministic multi-group alternative ERANOS [33].
5. Nuclear Data

The role of nuclear data (ND) is to provide quantitative information on nuclear processes that can not be described with satisfactory accuracy by a physical model alone. ND are normally based on both experimental results and theoretical considerations, depending on available empirically obtained data and energy regime. Considering the particular mixing of theory and experiment, as well as the span of nuclei, reaction channels and energies involved, it is evident that the topic of ND is vast and complex. Therefore, the account here will be limited to some of the aspects of ND that are relevant to the studies presented in the included research papers.

5.1 Nuclear Data in ADS Simulations

Accurate ND is a fundamental premise for calculations that are made in order to determine the parameters of a nuclear reactor core. In the case of 3D Monte Carlo neutronic simulation techniques that are applied to ADS, these data, and in particular the neutron cross sections, are directly used for calculating every type of relevant reaction or nuclear process, as well as particle paths (and thus spatial distributions) present in the studied system. Inaccuracies in ND can thus cause serious systematic errors in the results of ADS simulations. Figure 5.1 shows the capture, i.e. \((n,\gamma)\), cross sections of \(^{240}\text{Pu}\) according to JAR-95 [34] and JENDL-3.2 ND [35] compilations.

The example of Figure 5.1 is representative of the discrepancies that can exist between ND libraries for nuclides that are of importance to transmutation. In this case, JENDL-3.2 seems to have only partly resolved resonances in a region from about 100 eV up to 6 keV. These particular differences between the JAR-95 and JENDL-3.2 ND libraries were observed during the analysis presented in Paper VII and are (among some other deviant data) responsible for a larger amount of higher actinides being produced over time with JENDL-3.2 than with JAR-95 in transmutation calculations with time evolution.

Several international ND compilations exist, notably the ENDF/B, JENDL and JEF/JEFF series [35] are widely used. The data libraries were principally developed with LWR deterministic calculations as a main priority, and their accuracy for this purpose is normally sufficient. However, when applied to fast, subcritical systems, they yield unacceptably large differences in their
prediction of important system parameters vital to the safety and economy of these systems (cf. Papers II, III, V and VII). The situation is normally better in the case of fission (n,f) and capture neutron cross sections, at least for nuclides and energies of importance to LWR technology, whereas the inelastic (n,n’) scattering and (n,xn) reactions of fast neutrons are less well described.

In order to address and internationally coordinate the needs of ND, the OECD/NEA has set up the High Priority Nuclear Data Request List (HPRL) [36], which provides general recommendations for the neutron cross sections that are considered of importance. Table 5.1 relates the isotopes of the HPRL that are considered to be of particular importance to ADS analysis. On a general note, the HPRL suggested requirements are lower (an accuracy around ±10%) for isotopes that are representative of structural materials and fission products (FP). This is reasonable since these do not occur in large quantities or provide a large portion of the multiplicative processes in an ADS core. However, if parasitic transmutation of FP is to be realised, a stricter criterion may later be needed on their account.

However, the HPRL is not stringently based on cross section sensitivity studies, and its recommendations are thus more indicative than conclusive. By application of different ND sets, more thorough information can be obtained. The thus resulting discrepancies can be examined by means of the neutron
Table 5.1: The OECD/NEA High Priority Nuclear Data Request List, important nuclides and recommended accuracies. FPY denotes fission product yield, DNY delayed neutron yield, FNS fission neutron spectra and SFH spontaneous fission half-lives.

<table>
<thead>
<tr>
<th>Isotopes Data Accuracy</th>
<th>Data</th>
<th>Accuracy</th>
</tr>
</thead>
<tbody>
<tr>
<td>232Th, 233U, 235U, Pu-isot. (1 eV-500 keV)</td>
<td>(n,γ)</td>
<td>±10%</td>
</tr>
<tr>
<td>232Th, 238U, 240Pu, 239Pu</td>
<td>(n,n’)</td>
<td>±5%, ±10%</td>
</tr>
<tr>
<td>232Th, 238U (&gt;500 keV)</td>
<td>(n,f)</td>
<td>±10%</td>
</tr>
<tr>
<td>233U, 235U, 239Pu, 241Pu</td>
<td>FPY (92&lt;A&lt;140)</td>
<td>±3%</td>
</tr>
<tr>
<td>232Th, 238U, Pu-isot.</td>
<td>DNY</td>
<td>±3% to ±7%</td>
</tr>
<tr>
<td>237Np, 241Am</td>
<td>(n,γ), (n,f), (n,n’), (n,2n)</td>
<td>±10%</td>
</tr>
<tr>
<td>237Np, 238U, 241Am, 243Am, 244Cm</td>
<td>FNS, DNY, SFH</td>
<td></td>
</tr>
<tr>
<td>Fe, Cr, Ni, Pb, Bi, W</td>
<td>(n,n’), (n,xn)</td>
<td>±5% to ±10%</td>
</tr>
<tr>
<td>99Tc, 129I, 131Xe, 133Cs, 135Cs, 149Sm, 151Sm</td>
<td>(n,γ)</td>
<td>±10%</td>
</tr>
</tbody>
</table>

balance in different materials (Papers II, III and V) and study of one-group cross sections1 (Papers II and III). These studies (as well as the one of Paper VII) indicate that higher precisions than what is suggested by the HPRL are needed for certain nuclides.

Strictly taken, comparisons of simulations produced with different ND only indicate calculational precision and *not* accuracy. However, such comparisons do suggest what accuracies to expect when comparing calculation and measurement, since the accuracy between measurement and prediction can be supposed to assume magnitudes similar to (or possibly larger than) the precision acquired with different ND. Correspondingly, an apparent agreement between results produced with different ND is no guarantee for the ND quality – it is possible that the data only agrees due to the evaluations being based on similar premises.

1One-group cross sections are formed by convoluting the neutron flux spectra of the materials of interest with pointwise cross sections.
5.2 The n_TOF Experiment

As much of the simulation work presented in this thesis points out, the benefit of more accurate nuclear data and in particular neutron cross sections for ADS applications is unquestionable. One of the European research efforts lately addressing this fact is the n_TOF-ND-ADS programme, the neutron time-of-flight experiment at CERN. The main goal of n_TOF is to produce, evaluate and disseminate high precision cross sections for the majority of the isotopes relevant to waste incineration and ADS design, i.e. capture and fission cross sections for the minor actinides, capture cross sections for the main fission products, as well as (n,xn) reactions for structural and coolant materials [37].

Following the n_TOF proposal of 1999 [38], the facility was built and commissioned during 2000 and 2001 by the Emerging Energy Technologies (EET) group at CERN (Paper IV). The n_TOF collaboration performed active data collection between 2002 and 2004, and a possible recommencement of measurements could take place in 2007. Figure 5.2 shows how the facility is situated in relation to the CERN PS ring and highlights central components.

![Figure 5.2: A top-down view of the n_TOF facility layout.](image)

As the figure indicates, 20 GeV protons from the CERN PS accelerator complex [39] are sent to impinge on an $80 \times 80 \times 60$ cm$^3$ solid, high-purity (99.999 %) lead spallation target, where neutrons of a wide energy range are produced. The neutrons travel down a $\sim$185 m flight-path to the detector station in the experimental area, which is situated between 182.5 m and 190 m downstream from the spallation target. In combination with the possibility to
receive sharp pulses at a relatively low repetition rate, the long flight-path enables excellent resolution over a large neutron energy span. The facility and the commissioning is further described in Paper IV.

During its operation, the n_TOF facility has allowed systematic and accurate study of neutron-induced reactions in an energy range between 1 eV and 250 MeV. By means of the particular features of the n_TOF facility and the high performance detectors used, quality measurements of neutron-induced cross sections of radioactive samples of modest quantities have been possible, cf. Table 5.2 [40]. The measured data supersedes that of the presently available ND and will significantly contribute to the accuracy of future ND compilations.

Table 5.2: Some of the isotopes measured in the n_TOF experiment [40]. The cross sections are measured relative to the reference nuclei indicated in the lowermost row.

<table>
<thead>
<tr>
<th>Capture</th>
<th>Fission</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{151}\text{Sm}$, $^{204,206,207,208}\text{Pb}$</td>
<td>$^{233,234,235,236}\text{U}$</td>
</tr>
<tr>
<td>$^{209}\text{Bi}$, $^{232}\text{Th}$, $^{139}\text{La}$</td>
<td>$^{232}\text{Th}$</td>
</tr>
<tr>
<td>$^{24,25,26}\text{Mg}$</td>
<td>$^{237}\text{Np}$</td>
</tr>
<tr>
<td>$^{90,91,92,93,94,96}\text{Zr}$</td>
<td>$^{241,242}\text{Am}$</td>
</tr>
<tr>
<td>$^{186,187,188}\text{Os}$, $^{240}\text{Pu}$</td>
<td>$^{245}\text{Cm}$</td>
</tr>
<tr>
<td>$^{233,234}\text{U}$, $^{237}\text{Np}$, $^{243}\text{Am}$</td>
<td></td>
</tr>
<tr>
<td>$^{197}\text{Au}$</td>
<td>$^{235,238}\text{U}$</td>
</tr>
</tbody>
</table>
6. Summary of Papers

Paper I
Comparative Assessment of the Transmutation Efficiency of Plutonium and Minor Actinides in Fusion/Fission Hybrids and ADS

The transmutation efficiency of plutonium and minor actinides are assessed for the case of ANSALDO’s Energy Amplifier Demonstration Facility based on molten lead-bismuth eutectic cooling, classical MOX-fuel technology and operating at 80 MW_{th}. Detailed Monte Carlo transport calculations were performed for different types of external neutron sources: D-D and D-T fusion neutron sources and proton induced spallation neutron sources. The fuel core is described on a pin-by-pin basis, thus allowing for detailed scans of the main neutronic properties, e.g. neutron flux spectrum and power density distributions.

Papers II & III
Sensitivity Analysis of Neutron Cross Sections Relevant for Accelerator-Driven Systems

The cross section sensitivity in 3-D Monte Carlo steady-state calculations of accelerator-driven systems is reviewed. The concept studied was ANSALDO’s 80 MW_{th} Energy Amplifier Demonstration Facility. The simulations were carried out with the state-of-the-art computer code package EA-MC, developed by C. Rubbia and his group at CERN. Calculations yield large discrepancies in vital system characteristics when applying different sets of nuclear data. Considerations based upon one-group cross-sections together with a neutron balance comparison indicate the particular isotopes and neutron XS that appear to need better precision and thus are recommended for further measurement. Nuclides that are listed fulfil certain criteria: they show very high XS discrepancies, appear in significant amounts enough to need higher XS accuracy, were observed to cause direct effects on the steady-state neutronics, or a combination of these three factors.
Paper IV
Results from the commissioning of the n_TOF spallation neutron source at CERN

The new neutron time-of-flight facility (n_TOF) has been built at CERN and is since 2001 operational. The facility is intended for the measurement of neutron induced cross-sections of relevance to Accelerator-Driven Systems (ADS) and to fundamental physics. Neutrons are produced by spallation of the 20 GeV proton beam, delivered by the Proton Synchrotron (PS), on a massive target of pure lead. A measuring station is placed at ≈185 m from the neutron producing target, allowing high-resolution measurements. The facility was successfully commissioned with two campaigns of measurements, in November 2000 and April 2001. The main interest was concentrated in the physical parameters of the installation (neutron fluence and resolution function), along with the target behavior and various safety-related aspects. These measurements confirmed the expectations from Monte Carlo simulations of the facility, thus allowing to initiate the foreseen physics program.

Paper V
Importance of Neutron Cross-Sections for Transmutation

Cross-section discrepancies justify the need to improve the present data for several isotopes and reaction channels, for a wide range of neutron energies from thermal to high-energy. This paper follows up the work performed in the context of the n_TOF-ND-ADS project of the EURATOM 5th Framework Program, i.e. Paper III, where a preliminary analysis of the effects of different cross-section data was carried out using the Monte Carlo code package FLUKA-EAMC. That study was based on the Pb-Bi cooled 80 MWth Energy-Amplifier prototype, and included comparison of parameters such as source multiplication coefficient $k_{syr}$, neutron spectra, neutron balance and one-group cross-sections for different isotopes using different nuclear-data evaluations. This work expands this analysis to other isotopes of interest such as $^{233}$U, $^{243}$Am, $^{244,245}$Cm and the long-lived fission fragments (LLFFs) $^{99}$Tc and $^{129}$I. A direct comparison of nuclear data libraries to indicate the spread between values was performed. The paper also extends the sensitivity analysis of the parameters mentioned above to moderated systems, such as TRADE (Triga Accelerator-Driven Experiment): a 1 MW Triga Reactor coupled with a 110-140 MeV–2 mA proton cyclotron. Study of the discrepancies in the thermal and epithermal regions is essential for the design of systems for the transmutation of LLFF (transmutation by adiabatic resonance crossing, TARC) and also...
important for minor actinides (MAs) for which sub-threshold fission should not be neglected. These studies highlight the relative importance of different isotopes and assess the required accuracy for the given reaction cross-sections and the precision needed of the measurements carried out at the CERN n_TOF facility.

Paper VI
EA-MC Neutronic Calculations on IAEA ADS Benchmark 3.2

The neutronics and the transmutation properties of the IAEA ADS benchmark 3.2 setup, the "Yalina" experiment or ISTC project B-70, were studied through an extensive amount of 3-D Monte Carlo calculations at CERN. The simulations were performed with the state-of-the-art computer code package EA-MC, developed by C. Rubbia and his group at CERN. The calculational approach is outlined and the results are presented in accordance with the guidelines given in the benchmark description. A variety of experimental conditions and parameters are examined; three different fuel rod configurations and three types of neutron sources are applied to the system. Reactivity change effects introduced by removal of fuel rods in both central and peripheral positions are also computed. Irradiation samples located in a total of 8 geometrical positions are examined. Calculations of capture reaction rates in $^{129}$I, $^{237}$Np and $^{243}$Am samples and of fission reaction rates in $^{235}$U, $^{237}$Np and $^{243}$Am samples are presented. Simulated neutron flux densities and energy spectra as well as spectral indices inside experimental channels are also given according to benchmark specifications. Two different nuclear data libraries, JAR-95 and JENDL-3.2, are applied for the calculations.

Paper VII
Neutron Cross Section Sensitivity for Minor Actinide Transmutation in Energy Amplifier Systems

The nuclear data sensitivity in 3-D Monte Carlo burnup calculations of minor actinide transmutation in Energy Amplifier Systems is assessed. Ansaldo Nuclide’s 80 MWth Energy Amplifier Demonstration Facility (EADF) design serves as a technical and geometrical platform for the analysis. The accelerator-driven EADF is a fast, subcritical system based on classical MOX-fuel technology and on molten Lead-Bismuth Eutectic
cooling. For Monte Carlo simulations, the state-of-the-art computer code package EA-MC, developed by C. Rubbia and his group at CERN, is utilised. In particular, the impact of nuclear data discrepancies on transmutation properties prediction with increasing exposure is examined.
7. Conclusions and Prospects

Based upon projections of future energy demand and available fuel resources, it is argued that nuclear power must provide an important contribution to forthcoming worldwide energy needs. One of the main public concerns applied to nuclear energy generation is the long-lived, high-level radioactive waste (HLW) that arises from the production process. Accelerator-driven systems (ADS), which essentially are subcritical, neutron source-driven reactors, are suggested as a means for reducing HLW volumes and storage times from geological timescales at hundreds of thousands of years down to merely 1,000 years or less.

The particular studies accounted for mainly concern the calculational premises for numerical prediction of ADS behaviour and characteristics. Nuclear data (ND) effects are examined both for the cases of the Energy Amplifier Demonstration Facility (EADF) and the Triga Accelerator-Driven Experiment (TRADE), which are representative of systems with a fast and a thermal neutron economy, respectively. The EADF is further reviewed for ND effects on transmutation processes within the context of full-scale time evolution, i.e. burnup calculations. A description of the n_TOF experiment at CERN, which has yielded ND vital to ADS design and development, and its commissioning phase is also included. The main calculational tool employed for the presented ADS calculations, EA-MC, is studied by means of an international benchmark exercise, the Yalina experiment or IAEA ADS Benchmark 3.2. The deuteron-deuteron (DD) and deuteron-triton (DT) fusion source neutron distributions, on which the Yalina benchmark is based, are further applied to the EADF configuration. This enables a comparison of the device as driven by DD- and DT-sources to the normal case where a spallation target supplies the source neutrons.

Based upon experimental capabilities and critical reactor experience, as well as economical and safety considerations for ADS, precision requirements for calculated reactivity parameters are suggested. In terms of reactivity change per cycle, $\Delta \rho / \text{cycle}$, it is recommended that an accuracy at the scale of ten pcm (1 pcm = 1 percentmille = $10^{-5}$) or at maximum, some tens of pcm should be aimed for. The source multiplication factor, $k_{\text{src}}$, is proposed a corresponding calculational precision at the scale of maximum a few hundred pcm.
For the case of ADS operating with a fast neutron spectrum, ND-induced discrepancies of up to 2 % in $k_{\text{eff}}$ were observed for cases employing different ND compilations. Even the substitution of single isotopes between ND libraries was found to yield differences of up to 1.5 % in $k_{\text{eff}}$. These studies also employed newer ND versions, but due to these results, no major improvement of overall ND agreement could be inferred. In the case of thermal systems, the polyethylene-moderated Yalina setup showed $k_{\text{eff}}$ discrepancies of around 1 % with DT and DD fusion sources, whereas the fission source showed no larger deviations than 0.1-0.5 %. An ND effect arising from the introduction of a harder source neutron spectrum in an essentially thermal device is thus demonstrated. Similar calculations on the TRADE experiment, which employs a spallation neutron source, yielded smaller differences of 0.2-0.3 %. Indications are that the difference between this and the Yalina results can be largely explained by the comparably large dimensions of the TRADE core and the abundance of water moderator therein. Neutron slowing-down is here far more effective than in the Yalina assembly, and hence TRADE is not as reliant as Yalina on nuclear data in the high-energy regime, which in turn produces smaller ND-related errors in main system parameters.

Among other results, the neutron cross section sensitivity studies performed indicate isotopes and ND that should be revisited in measurement and evaluation. Some of the neutron cross sections included in measurement recommendations are:

- For fuel materials:
  - Fast fission for $^{232}$Th, $^{238}$U, $^{240}$Pu and $^{242}$Pu,
  - Capture for $^{238}$Pu, $^{240}$Pu and $^{241}$Pu,
  - Inelastic scattering for $^{238}$Pu, $^{239}$Pu, $^{240}$Pu and $^{241}$Pu,
  - Elastic scattering for $^{238}$Pu and $^{241}$Pu,
  - $(n,xn)$ for most of the actinide isotopes examined.
- For cladding materials:
  - In particular capture for Fe isotopes,
  - Other cross sections for Fe isotopes and $^{52}$Cr are not essential, but discrepancies are large.
- For coolant materials:
  - $(n,xn)$ for Pb isotopes and $^{209}$Bi,
  - Inelastic scattering for Pb isotopes.

Studies with updated ND libraries and based on individual isotope exchange showed improved agreement between ND for the particular cases of fission and capture in $^{240}$Pu. Furthermore, these studies generally indicate the need for better data on $^{241}$Pu and $^{244}$Cm, as well as $^{90}$Zr, due to the isotope-level discrepancies registered.

The examination of ND effects on ADS burnup calculations show that $\Delta \rho$/cycle is in excess of 400 pcm, overshooting the precision goal by roughly
one order of magnitude. A difference in the prediction of Pu buildup causes
the effect, and indications are that the (n,γ) cross section of 238U is responsible
and would preferably need to be determined within 1% accuracy. Radiotoxic-
ity considerations imply that (n,γ) in 239Pu and 240Pu, as well as possibly 241Pu
and 241Am, would need improvement in order to ensure the quality of burnup transmutation calculations. In the case of LLFP, better fission yield data for
129I are requested, since reliable nuclide mass content prediction seems un-
feasible with current fission yield precision. Several of the measurement rec-
recommendations yielded by the neutron cross section sensitivity studies have
been addressed within the framework of the n_TOF experiment.

The IAEA ADS Benchmark 3.2, the "Yalina" experiment, was studied and
reviewed by means of the Monte Carlo neutron transport code package EA-
MC. Only limited, preliminary data were accessible and no other benchmark
reports were available at the time of writing, but comparisons with the pre-
liminary measurements of effective multiplication factors (k_{eff}) indicate that
sensible results were obtained. It appears k_{eff} is increasingly underestimated
at lower fuel loading, but before accurate experimental data are available def-
inite conclusions can not be drawn. One of the observations was that even an
essentially thermal, subcritical source-driven system can be highly dependent
on accurate fast neutron cross sections if the system employs source neutrons
of high energy and the geometrical conditions allow the penetration of fast
neutrons into vital segments of the core. Some minor issues affecting the per-
formance of EA-MC for the Yalina geometry were discovered and reviewed.
These will lead to future enhancement of the code. The work also resulted in
general recommendations for the design of subcritical experiments planned
for code benchmarking purposes.

The neutronic properties of three different neutron source configurations of
the EADF were examined by means of 3-D Monte Carlo techniques. Two of
these source configurations, the DD- and DT-sources are related to the fusion
neutron sources used in the Yalina experiment. The results indicate that the
accelerator-driven and the fusion-DD source systems would exhibit similar
neutronic properties. The fusion-DT source driven configuration differs from
the others, giving rise to a k_{sync} approximately 1.5% higher than the others. The
effect was seen to be mainly due to the DT-fusion characteristic neutron emis-
sion energy spectrum, which produces a significantly larger share of (n,xn)
produced neutrons. It was further established that the choice of source config-
uration has negligible impact on transmutation efficiencies and transmutation
rates. The results verify the relevance of fusion neutron source-driven subcrit-
critical experiments to ADS neutronics, but also point out some key differences
between such systems and spallation neutron source-driven ones.

The ND-related work presented suggests further study within the field of
neutron cross section sensitivity analysis. Due to a lack of data, a full anal-
ysis with a cross section sensitivity-uncertainty code utilising cross section covariance data files was not feasible at the time when the project should have been started. Such an investigation would be of great interest, since it would allow for better quantification of the impact of individual neutron cross sections. Concerning burnup calculations, a more accurate evaluation of ND effects for the particular case of MA transmutation would be attainable, if an assessment was made for a system free from uranium- and plutonium-based fuels. The benchmarking effort for EA-MC could also be enriched by inclusion of calculations on the First Energy Amplifier Test (FEAT) [41, 42], an experiment performed at CERN in 1995 for verification of the Energy Amplifier principles. From the European Union Framework Programme 7, funding for the recommencement of the n_TOF measurements will hopefully be reserved, thus allowing the experiment to continue its operation and production of high-quality ND in 2007.
8. Summary in Swedish

Översatt till svenska, bär denna avhandling titeln Studier av accelerator-drivna system för transmutation av kärnavfall. De specifika studierna omfattar områden såsom kärndata för simulering av acceleratordrivna system (ADS), olika neutronkällors inverkan på simuleringsresultat, samt verifiering av beräkningsverktyg, s.k. benchmarking.

8.1 Energitillgångar och -produktion


Kärnkraften är den storskaliga energikälla som visar de bästa egenskaperna för att klara av mer omfattande expansionskrav inom energisektorn. Dagens kärnkraftsindustri har utvecklat och implementerat säkerhetsstandarder som långt överstiger den generella nivån hos övrig industri, samtidigt som en kultur av öppenhet mot samhället aktivt eftersträvas. Kärnkraftsindustrin kan därför anses mogen att framöver ta ett större ansvar för världens energiproduktion. Världens urantillgångar är förhållandevis rikliga och har potential att tillgodose stora delar av världens energibehov i ett hundratal år med lättvattenreaktorer och i tusental år om snabba reaktorer (och eventuellt även ADS) utnyttjas för produktion av nytt fissilt material. För att kärnkraften skall nå en hög grad av acceptans hos allmänheten, behövs goda alternativ för omhändertagandet av det radioaktiva avfallet. ADS har därför föreslagits att ingå som en del i sluthanteringen av det mest radioaktiva avfallet, eftersom tekniken möjliggör en betydande reduktion av avfallsvolymer och lagringstider. Lagringstiderna kan minskas från hundratusentals år till mindre än 1.000 år.
8.2 Radioaktivt avfall

I samband med kraftproduktion med hjälp av kärnkraft uppstår radioaktivt avfall. Detta klassificeras primärt som högaktivt (high-level waste, HLW) eller lågaktivt avfall¹ (low-level waste, LLW). HLW består av mycket radioaktiva fissions- eller infångningsprodukter som uppstår i kärnbränslet under drift. LLW innebär avfall som uppkommer i anläggningen under drift, t.ex. aktiverade maskindelar, strukturmaterial och skyddskläder, samt rester från medicinsk och industriell användning av radionuklider. Acceleratordriven transmutation hör till HLW.

HLW indelas vanligt i två grupper: transuraner (TRU) och fissionsprodukter. Transuraner uppstår från uran genom transmutationsprocesser som förekommer i alla verksamma nukleärt multiplikativa system, jfr. Figur 3.1, avsnitt 3.1.1. Det mest betydande bidraget till den långlivade radiotoxiciteten (anger hur skadliga radionuklider är för levande organismer) i HLW kommer från TRU, jfr. Tabell 3.1 i avsnitt 3.1.1. Fissionsprodukter bildas efter sönderfall från de två neutronrika kärnfragment som uppstår efter en kärnklyvning. Många av de långlivade av dessa (LLFP) är vattenlösda och således mycket mobila. Det största bidraget, kring 95 % av volymen, till långtidsrisken hos LLFP hörer från 99Tc och 129I. Därför vore det av intresse att transmutera även dessa i tillägg till TRU.

Den lösning som de flesta länder gått in för, består i att det mest långlivade avfallet djupförvaras i stabila geologiska formationer, jfr. Figur 3.4, avsnitt 3.3.1. Trots att metoden understöds av omfattande forskning och placeringen av slutförvar föregås av noggranna geologiska undersökningar, förhåller sig allmänheten ofta skeptisk till detta alternativ.

Snarare som ett komplement än som ett alternativ till det geologiska djupförvaret, kunde ADS föras in som en komponent i hanteringen av kärnavfallet. ADS kan användas för att transmutera HLW, m.a.o. behandla avfallet på teknisk väg så att långlivat radioaktiva kärnor omvandlas till stabila eller kortlivade kärnor. Efter transmutation av HLW i ADS minskas avfallets volym och de långlivade komponenterna avlägsnas. Restprodukterna som kvarstår efter behandlingen måste dock slutförvaras, om än kraven på förvarets långtida säkerhet kan ställas lägre. Det bör påpekas att ADS förutsätter att långlivade isotoper kan återcirkuleras, vilket innebär att kärnavfallet måste upparbetas.

¹En tredje kategori, medelaktivt avfall tas ibland med i klassificeringsschemat. Aktiverade maskindelar och strukturmaterial räknas då oftast in i denna kategori.
8.3 Acceleratordrivna system

Till grundprinciperna för ADS hör att systemet är underkritiskt och drivs med en yttre neutronkälla som i sin tur drives med hjälp av en accelerator, jfr. Figur 4.1, avsnitt 4.2. Denna levererar högenergipartiklar, vanligen protoner, som får att kollidera med ett strålmål utformat för att producera ett högt antal neutroner per inkommande partikel.

I strålmålet produceras neutroner med hjälp av spallationsprocesser. Nukleär spallation är inte en klart definierad fysikalisk process, utan innebär i korthet en mängd av kärnreaktioner, där de inkommande partiklarnas energi är så hög att fler än två eller tre partiklar slås ut från målkärnorna samtidigt som deras massor och atomnummer ändras, jfr. Figur 4.2 i avsnitt 4.3.1. Beroende på den kinetiska energin hos de inkommande partiklarna och på vilket material spallationsmålet består av, kan antalet avgivna partiklar – speciellt neutroner – vara stort. Även andra typer av neutronkällor kan användas, speciellt för småskaliga underkritiska experimentuppställningar. Bl.a. används ofta fusionsneutronkällor baserade på reaktionerna deuterium-deuterium (DD) och deuterium-tritium (DT). Sådana neutronkällor tillämpas även i studier ingående i denna avhandling (Publikation I och VI).

Härden hos ADS är underkritisk, vilket innebär att en nukleär kedjereaktion som startas inte är självunderhållande och därför kommer att avstanna. I ett underkritiskt system kvantifieras neutronmultiplikationen med hjälp av den s.k. källmultiplikationskonstanten, $k_{src}$. Jämfört med den effektiva multiplikationskonstanten, $k_{eff}$, som används för att karaktärisera kritiska reaktorer, beaktar $k_{src}$ även källneutronernas rymddistribution och energispektra.

En följd av de underkritiska egenskaperna hos ADS är att bränslen som inte kan utnyttjas i konventionella kritiska reaktorer p.g.a. säkerhetsbegränsande egenskaper, kan användas säkert i ADS med dess till följd av underkriticiteten utökade reaktivitetsmarginaler. Utvecklingen av ADS har lett till att de system som idag föreslås använder sig av snabba neutroner, eftersom dessa möjliggör fission även i kärnor som inte låter sig klyvas av termiska neutroner. Prototypen baserad på C. Rubbias energiförstärkarkoncept [5], EADF, är representativ för modern ADS-design och används som en teknisk bas för de flesta av analyserna i avhandlingen [19].

Beräkningar av egenskaperna hos ADS är mer krävande än hos konventionella reaktorer. Dels är neutronflödet mer ojämnt fördelat och dels måste neutroner och andra partiklar med hög kinetisk energi behandlas. Detta gör att de deterministiska metoder som traditionellt används för reaktorberäkningar är olämpliga för ADS-beräkningar. Istället används simuleringsskoder baserade på Monte Carlo-metoder. Dessa är statistiska till sin natur och går enkelt uttryckt ut på att följa ett stort antal partiklar som skickas in i ett system från
deras inträde i systemet, samt alla reaktioner och reaktionsprodukter de orsakar, ända till dess att de på sätt eller annat försvinner ur systemet.


8.4 Kärndata

Syftet med kärndata är att ge kvantitativ information om kärnprocesser som inte kan beskrivas tillräckligt väl enbart genom fysikaliska modeller. Framtagningen av kärndata baseras i allmänhet både på teoretiska och experimentella överväganden. Detta innebär att betydande skillnader mellan olika utvärderingar, d.v.s. olika kärndatabibliotek, kan uppkomma beroende på vilka data och modeller som ligger till grund för framtagningen.


Ett europeiskt initiativ till förbättring av kärndata är n_TOF-ND-ADS-projektet. Målsättningen för n_TOF-experimentet vid CERN är att framställa, utvärdera och sprida tvärsnittsdata av hög precision för en majoritet av isotoper som är relevanta för transmutation och ADS-design, d.v.s. infångnings- och fissionstvärsnitt för mindre aktinider, infångningstvärsnitt för de huvudsakliga fissionsprodukterna, samt (n,xn)-tvärsnitt för strukturoch kylmedelsmaterial [37]. I avhandlingen redogörs för resultat från n_TOF:s uppstarts- och verifieringsperiod (Publikation IV).

8.5 Slutledningar och utsikter

Baserat på experimentella möjligheter och erfarenheter från kritiska reaktorer ges rekommendationer för att beräkningsprecisionen för reaktivitetsändring
per cykel, $\Delta \rho$/cykel, inte bör vara sämre än något tiotal pcm (1 pcm = 1 percentmille = $10^{-5}$). Motsvarande precision för $k_{src}$ bör ligga på en skala av något hundratal pcm.

Diskrepanser på upp till 2 % i $k_{src}$ kan dock observeras mellan simuleringar baserade på olika kärndatabibliotek. Även för $\Delta \rho$/cykel hittas diskrepanser på över 400 pcm, vilket överstiger målet med ungefär en storleksordning. För termiska system är skillnaderna mindre, men ju snabbare neutroner källan avger, desto större blir diskrepanserna om systemet inte är tillräckligt väl modererat. Rekommendationer för nuklider och reaktioner som bör prioriteras vid mätningar ges på basen av utförda analyser av tvärsnittskänslighet.

Simuleringskoden EA-MC har undersöpts inom ett internationellt kodverifieringsprojekt, Yalina-experimentet eller mer formellt IAEA ADS Benchmark 3.2, och befunnits ge tillfredsställande resultat enligt de data som varit tillgängliga. Verifieringen har resulterat i ett antal förslag på förbättringar i EA-MC, samt i generella rekommendationer för utformningen av underkritiska experiment i kodverifieringssyfte.

DD-, DT- och spallationsneutronkällors inverkan på den underkritiska neutroniken har undersömts inom ramen för EADF. DT-källor ger den största avvikelsen i $k_{src}$ från det spallationskälladrivna referenssystemet med en skillnad på 1,5 %. Valet av neutronkälla har dock konstaterats ha ringa inverkan på transmutationsrater och -effektivitet. Resultaten verifierar relevansen av fusionskälladrivna underkritiska experiment (såsom Yalina-experimentet) för ADS-neutroniken, men påvisar också några va sentliga skillnader mellan sådana system och spallationsneutro ndrivna.

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