External Source Effects and Neutronics in Accelerator-driven Systems

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Abstract

Transmutation of plutonium and minor actinides in accelerator-driven systems (ADS) is being envisaged for the purpose of reducing the long-term radiotoxic inventory of spent nuclear reactor fuel. Consequently, the physics of sub-critical systems are being studied in several different experimental programs across the world, one of them being the MUSE (MUltiplication of External Source) program. In these experiments, an intense external neutron source is coupled to a sub-critical core. In order to investigate the neutronics and source effects in a sub-critical system, Monte Carlo simulations have in this thesis been performed for a model representative of the MUSE-4 experiments. The investigations have focused on three different neutronic parameters; the neutron energy spectrum, the external neutron source efficiency ($\phi^*$) and the dynamic neutron source response.

In order to study the beam power amplification of an ADS, we have introduced a new parameter, the proton source efficiency ($\psi^*$). $\psi^*$ represents the average importance of the external proton source, relative to the average importance of the eigenmode neutron production. It is defined in analogy with the neutron source efficiency $\phi^*$, but relates the core power to the source protons instead of to the source neutrons. $\phi^*$ is commonly used in the physics of sub-critical systems, driven by any external neutron source (spallation source, (d,d), (d,t), $^{252}$Cf spontaneous fission etc.). On the contrary, $\psi^*$ has been defined only for ADS studies, where the system is driven by a proton-induced spallation source. The main advantages with using $\psi^*$ instead of $\phi^*$ are that the way of defining the external source is unique and that $\psi^*$ is proportional to the core power divided by the proton beam power, independently of the neutron source distribution. The second part of this thesis has focused on studying $\psi^*$ as a function of different system parameters, thereby providing a basis for an ADS design with optimal proton beam amplification.
List of Publications

The following papers are included in this thesis.


<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>ACER</td>
<td>Calculational routine in the NJOY code, also nuclear data format for Monte Carlo codes</td>
</tr>
<tr>
<td>ADS</td>
<td>Accelerator-Driven Systems</td>
</tr>
<tr>
<td>ALINA</td>
<td>Reprocessing technique for separating actinides from lanthanides</td>
</tr>
<tr>
<td>ATALANTE</td>
<td>ATelier Alpha et Laboratoires pour ANalyses, Transuraniens et Etudes de retraitement</td>
</tr>
<tr>
<td>ATW</td>
<td>Accelerator-driven Transmutation of Waste</td>
</tr>
<tr>
<td>BISTRO</td>
<td>Two-dimensional transport code, used by ERANOS</td>
</tr>
<tr>
<td>BTP</td>
<td>Bis-Triazinyl-Pyridine, an organic extractant</td>
</tr>
<tr>
<td>CEA</td>
<td>Commissariat à l’Energie Atomique (France)</td>
</tr>
<tr>
<td>CEM</td>
<td>Cascade Exciton Model</td>
</tr>
<tr>
<td>CERCER</td>
<td>CERamic-CERamic composites</td>
</tr>
<tr>
<td>CERMET</td>
<td>CERamic-METal composites</td>
</tr>
<tr>
<td>CNRS</td>
<td>Centre National de la Recherche Scientifique (France)</td>
</tr>
<tr>
<td>CYANEX 301</td>
<td>Bis-(2,4,4-trimethylpentyl) dithiophosphinic acid, an organic extractant</td>
</tr>
<tr>
<td>DIAMEX</td>
<td>DIAMide EXtraction process</td>
</tr>
<tr>
<td>DIDPA</td>
<td>Di-IsoDecylPhosphoric Acid</td>
</tr>
<tr>
<td>DTL</td>
<td>Drift Tube Linac</td>
</tr>
<tr>
<td>ECCO</td>
<td>European Cell COde</td>
</tr>
<tr>
<td>ECR</td>
<td>Electron Cyclotron Resonance</td>
</tr>
<tr>
<td>EDC</td>
<td>Effective Dose Coefficient</td>
</tr>
<tr>
<td>EdF</td>
<td>Electricité de France</td>
</tr>
<tr>
<td>EFTTRA</td>
<td>Experimental Feasibility for Targets and TRAnsmutation</td>
</tr>
<tr>
<td>ENDF</td>
<td>Evaluated Neutron Data File</td>
</tr>
<tr>
<td>ERALIB1</td>
<td>ERAnos LIBrary</td>
</tr>
<tr>
<td>ERANOS</td>
<td>European Reactor ANalysis Optimized System</td>
</tr>
<tr>
<td>ESS</td>
<td>European Spallation Source</td>
</tr>
<tr>
<td>FLUKA</td>
<td>FLUctuating KAscade simulation program</td>
</tr>
<tr>
<td>FR</td>
<td>Fast Reactor</td>
</tr>
<tr>
<td>GENEPI</td>
<td>GEnerator de NEutrons Pulsés Intenses</td>
</tr>
<tr>
<td>GSI</td>
<td>Gesellschaft für SchwerIonenforschung, the German research center for heavy ion physics</td>
</tr>
<tr>
<td>Acronym</td>
<td>Definition</td>
</tr>
<tr>
<td>---------</td>
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</tr>
<tr>
<td>SAD</td>
<td>Sub-critical assembly in combination with the proton accelerator in Dubna</td>
</tr>
<tr>
<td>SANEX</td>
<td>Selective ActiNides(III) EXtraction</td>
</tr>
<tr>
<td>SCRF</td>
<td>Super-Condacting Radio-Frequency</td>
</tr>
<tr>
<td>SESAME</td>
<td>Selective Extracting Separation of Americium by Means of Electrolysis</td>
</tr>
<tr>
<td>SINQ</td>
<td>Swiss spallation neutron source</td>
</tr>
<tr>
<td>SNR300</td>
<td>Schneller Natriumgekühlter Reaktor, German fast sodium-cooled reactor</td>
</tr>
<tr>
<td>SNS</td>
<td>Spallation Neutron Source</td>
</tr>
<tr>
<td>SODERN/GENIE</td>
<td>14 MeV Neutron Generator, using the (d,t)-reaction</td>
</tr>
<tr>
<td>SPX</td>
<td>SuperPhenix</td>
</tr>
<tr>
<td>TALSPEAK</td>
<td>Trivalent Actinide-Lanthanide Separation by Phosphorus reagent Extraction from Aqueous Complexes</td>
</tr>
<tr>
<td>TBP</td>
<td>Tri-Butyl-Phosphate</td>
</tr>
<tr>
<td>TIERCE</td>
<td>A high-energy particle transport code system</td>
</tr>
<tr>
<td>TRASCO</td>
<td>TRAsmutazione SCOrie, a research project for the design of an ADS</td>
</tr>
<tr>
<td>TRPO</td>
<td>TRialkyl Phosphine Oxide</td>
</tr>
<tr>
<td>TRU</td>
<td>TRansUranic elements</td>
</tr>
<tr>
<td>TRUEX</td>
<td>TRansUranium EXtraction</td>
</tr>
<tr>
<td>UOX</td>
<td>Uranium OXide fuel</td>
</tr>
<tr>
<td>XADS</td>
<td>Experimental Accelerator-Driven System</td>
</tr>
<tr>
<td>XADT</td>
<td>Experimental Accelerator-Driven Transmuter</td>
</tr>
</tbody>
</table>
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I am indebted to several different people for their contribution to this thesis. Therefore, I use this place to express my gratitude to some of them. I thank

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Chapter 1

Introduction

The growing stockpiles of radiotoxic nuclear waste accumulated over the years by the operation of existing nuclear reactors are a matter of public concern. Different options of how to deal with this growing problem are investigated, transmutation in accelerator-driven systems (ADS) being one of them [1,2,3]. In principle, the sub-criticality of ADS allows for much higher concentrations of minor actinides in the core than is acceptable in critical reactors. In combination with appropriate reprocessing of the spent fuel, the use of ADS could become an efficient means for reducing the long-term radiotoxicity of the nuclear waste. Calculations show that by recycling of plutonium, americium and curium, a reduction factor of about 100 is theoretically possible [4].

In an ADS, a sub-critical core is coupled to a high-power proton accelerator. The high-energy protons (~1000 MeV) impinge on a target of heavy metal, generating a large number of neutrons via spallation reactions. The produced spallation neutrons leak out from the target, thus providing the surrounding sub-critical core with a strong external neutron source. The new and advanced technologies of accelerator-driven systems are being thoroughly investigated worldwide and the optimal choice among many design options is still an open question. Several independent areas, such as core coolant studies, the spallation module, the accelerator technology and the reprocessing and fuel fabrication technologies will need many years of development before they could be implemented on a large-scale industrial ADS. A general introduction to transmutation of nuclear waste and a description of the status of some of the related research areas under development are presented in Chapter 2.

Several different experimental programs around the world are focusing on the development of ADS, one of them being the MUSE experiments. The current phase of this project is MUSE-4 [5], which started in 2000 via the 5th Framework Programme of the European Community and where detailed analyses of the neutronics of a source-driven sub-critical system are performed. The possibility for the accurate measurements being carried out has been achieved by the coupling of the neutron generator GENEPI [6] and the MASURCA facility, in which different sub-critical configurations can be loaded. GENEPI is used to accelerate a 250 keV...
deuteron beam towards either a deuterium or a tritium target, producing well-characterized neutron sources via nuclear fusion reactions ((d,d)- and (d,t)-reactions, respectively). One of the objectives of the present work has been to investigate, by Monte Carlo simulation, different neutronical aspects of the sub-critical MUSE-4 core, coupled to an external neutron source. Apart from the presence of the (d,d)- and the (d,t)-sources, a spallation source has also been simulated, in order to compare some of the source characteristics of the MUSE experiments with a future spallation-driven ADS. In Chapter 3, the MUSE experiments are described and the investigations of the neutronic properties of a MUSE-4-like core, in terms of energy spectrum, neutron source efficiency ($\phi^*$) and dynamic neutron source response are presented.

One of the objectives when designing an ADS is to strive for as high power as possible to be produced in the core (yielding high transmutation rates), using as low proton beam power as possible. Since the construction of a reliable high-power proton accelerator is an extremely difficult technical task and its operation is very expensive, the optimization of the proton beam power amplification could have an important impact on the overall design of a future ADS and on the economy of its operation. Naturally, this optimization must be achieved in conjunction with the various safety constraints as well as other technical and physical conditions. The neutron source efficiency parameter ($\phi^*$) is related to the total energy released by fission in the core and, for constant neutron source intensity and a fixed reactivity level, it is proportional to the core power. Therefore, $\phi^*$ is commonly used to study the ratio of the core power over the external source power. The source efficiency is dependent on several different system parameters, such as target radius, coolant material, fuel composition and proton energy.

In an ADS, the source particles are protons, which produce neutrons in the target via spallation reactions. In order to calculate $\phi^*$, the external neutron source first has to be defined and then the efficiency of this neutron source can be determined. A problem associated with $\phi^*$ is that the external neutron source can be defined in different ways, leading to different results. Indeed, different groups in the ADS field studying the neutron source efficiency do use different source definitions, which in many cases makes results from different studies difficult to compare. Another drawback with the neutron source efficiency may appear when $\phi^*$ is studied as a function of different system parameters. If a certain system parameter, e.g. the target radius, is changed, the neutron source distribution and the number of source neutrons per incident proton ($Z$) may also change, which consequently would alter the initial conditions for the study. In order to represent the beam power amplification in this case, $\phi^*$ needs to be weighted with $Z$.

In contrast to the source neutrons, which have no uniquely defined beginning of life, the source protons can only be approached in one way. For the purpose of studying the source efficiency and the beam power amplification in ADS, we have therefore introduced a new parameter, $\psi^*$, which refers to the source protons instead.
of to the source neutrons. $\psi^*$ thus relates the core power directly to the proton beam. The advantages in using the proton source efficiency instead of the neutron source efficiency is that there is no ambiguity in how to define the external source and that it is proportional to the beam power amplification, without the need of a weighting factor ($Z$). The proton source efficiency is treated in Chapter 4, where investigations of $\psi^*$ as a function of several different system parameters, such as target radius, coolant material, axial proton beam impact, proton energy, fuel composition and choice of inert matrix are also presented.
Chapter 2

Transmutation of Nuclear Waste

2.1 Nuclear Waste

2.1.1 Composition of the Nuclear Waste

According to the Code of Practice issued by the International Atomic Energy Agency (IAEA) [7], “radioactive waste is any material that contains or is contaminated with radionuclides at concentrations or radioactivity levels greater than the exempt quantities established by the competent authorities and for which no use is foreseen”. In Europe, about 145 operating nuclear reactors produce a total power of about 125 GW electric power, which annually generates 2500 tons of spent fuel [8]. Most of this (~94.5%) still consists of the original uranium, while about 4.2% has been converted to fission products and 1.2% to transuranic elements (TRU, the elements with atomic number higher than 92). The relative composition, together with the half-lives and the effective ingestion dose coefficients (EDC) [9], of the transuranic elements in spent light-water reactor (LWR) uranium oxide (UOX) fuel after four years of cooling are listed in Table I. The exact amount and composition of the spent fuel varies somewhat between different discharged spent fuel assemblies, depending on the initial uranium enrichment and on the discharge burn-up. However, the magnitudes are similar for all LWR fuels, except for those containing recycled plutonium (MOX fuels).

There are mainly two families of nuclear waste that have to be disposed of; fission products and the transuranic elements. The transuranic elements are produced in nuclear reactors by neutron absorption in heavy nuclei, e.g. in $^{238}$U. All transuranic elements are unstable and will disintegrate via alpha, beta or gamma decay. Further neutron captures and successive disintegrations during reactor operation generate a wide spectrum of actinides. As an example, $^{239}$Pu is produced by the absorption of a neutron in $^{238}$U, followed by two successive $\beta^-$-decays. An example of a chain of reactions starting with $^{238}$U and producing Pu and Am is
\[ n + {}^{238}U \rightarrow {}^{239}U + \gamma \rightarrow {}^{239}Np + e^- + \bar{\nu} \rightarrow {}^{239}Pu + e^- + \bar{\nu} \]
\[ n + {}^{239}Pu \rightarrow {}^{240}Pu + \gamma \]
\[ n + {}^{240}U \rightarrow {}^{241}Pu + \gamma \rightarrow {}^{241}Am + e^- + \bar{\nu} \]

The transuranic elements constituting the most hazardous part of the spent fuel in the long term are generally long-lived with half-lives of up to several hundred thousand years.

### TABLE I

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life (Years)</th>
<th>Effective Ingestion Dose Coefficient (Sv/Bq)</th>
<th>Relative Mass (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{237}$Np</td>
<td>$2.14 \times 10^6$</td>
<td>$1.1 \times 10^{-7}$</td>
<td>4.6</td>
</tr>
<tr>
<td>$^{238}$Pu</td>
<td>87.7</td>
<td>$2.3 \times 10^{-7}$</td>
<td>2.3</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>$2.41 \times 10^4$</td>
<td>$2.5 \times 10^{-7}$</td>
<td>49.0</td>
</tr>
<tr>
<td>$^{240}$Pu</td>
<td>$6.56 \times 10^3$</td>
<td>$2.5 \times 10^{-7}$</td>
<td>21.5</td>
</tr>
<tr>
<td>$^{241}$Pu</td>
<td>14.4</td>
<td>$4.7 \times 10^{-9}$</td>
<td>11.4</td>
</tr>
<tr>
<td>$^{242}$Pu</td>
<td>$3.75 \times 10^5$</td>
<td>$2.4 \times 10^{-7}$</td>
<td>6.2</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>432</td>
<td>$2.0 \times 10^{-7}$</td>
<td>2.8</td>
</tr>
<tr>
<td>$^{243}$Am</td>
<td>$7.37 \times 10^3$</td>
<td>$1.9 \times 10^{-7}$</td>
<td>1.6</td>
</tr>
<tr>
<td>$^{244}$Cm</td>
<td>18.1</td>
<td>$1.2 \times 10^{-7}$</td>
<td>0.5</td>
</tr>
</tbody>
</table>

The fission products, on the other hand, are created when a heavy nucleus undergoes fission and is disintegrated into two fission fragments, also releasing two or three prompt neutrons. These fission products are generally short-lived and most of them disappear rapidly after the reactor has been shut off. However, a number of them has relatively long half-lives and remain toxic for an extended period of time. $^{90}$Sr and $^{137}$Cs have half-lives of about 30 years and are the most radiotoxic isotopes of the spent fuel during the first decades after discharge. $^{99}$Tc, $^{129}$I and $^{135}$Cs have half-lives longer than 100 000 years and, though much less toxic, might possibly turn out to be a potential problem in the very long run. The half-lives and effective ingestion dose coefficients of the most hazardous fission products are listed in Table II.
### TABLE II
Half-lives and Effective Ingestion Dose Coefficients for some of the Radiotoxic Fission Products

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life (Years)</th>
<th>Effective Ingestion Dose Coefficient (Sv/Bq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{90}$Sr</td>
<td>28.64</td>
<td>$2.8 \cdot 10^{-8}$</td>
</tr>
<tr>
<td>$^{99}$Tc</td>
<td>$2.1 \cdot 10^5$</td>
<td>$7.8 \cdot 10^{-10}$</td>
</tr>
<tr>
<td>$^{129}$I</td>
<td>$1.57 \cdot 10^7$</td>
<td>$1.1 \cdot 10^{-7}$</td>
</tr>
<tr>
<td>$^{135}$Cs</td>
<td>$2.0 \cdot 10^6$</td>
<td>$2.0 \cdot 10^{-9}$</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>30.17</td>
<td>$1.3 \cdot 10^{-8}$</td>
</tr>
</tbody>
</table>

#### 2.1.2 Radiotoxic Inventory

The activity of an unstable isotope is defined as the number of disintegrations per second and is measured in units of Becquerels (Bq). However, in order to better express the harmful potential of the uptake of certain isotopes by humans, the concept of radiotoxicity is used. To convert the activity of the inhaled or ingested radionuclide, it has to be multiplied by a so-called effective dose coefficient (EDC), expressed in sieverts per becquerel (Sv/Bq), where a sievert is the unit describing the biological effect of radiation deposited in an organism. The EDC is thus a measure of the damage a radionuclide would do to the body, taking into account factors such as the metabolism of the nuclides in the organism, the type and energy of the emitted radiation, tissue-weighting factors etc. In general, the EDC are several orders of magnitude higher for the actinides than for the fission products. The EDC are published and regularly updated by the International Commission on Radiological Protection (ICRP) \[9\].

The overall radiotoxicity of the spent fuel obviously decreases with time, as the radioactive nuclides decay. The evolution over time of the radiotoxic inventory is depicted in Fig. 1, showing the different contributions from the transuranic elements and the fission products. It is seen that the contribution of the fission products dominates the radiotoxicity during the first 30 or 40 years after discharge. Thereafter, it decreases relatively rapidly, along with the decay of $^{90}$Sr and $^{137}$Cs, and becomes negligible after a few hundred years. For time periods longer than a few hundred years, the transuranic elements govern completely the radiotoxicity, with the largest contribution coming from the plutonium isotopes and $^{241}$Am. $^{241}$Am is the decay product of $^{241}$Pu and represents the major part of the radiotoxic inventory in the time span between about 50 and 2000 years. After that, $^{239}$Pu and $^{240}$Pu are the main contributors to the radiotoxicity.

As a reference, the radiotoxicity of the spent fuel can be compared with the equilibrium radiotoxicity of natural uranium and its radioactive daughters in a uranium ore, which is approximately 20 mSv/g \[11\]. It is seen in Fig. 1 that it would
take about 300 000 years for the spent fuel to decay to this level. It is also indicated that the radiotoxicity of the very long-lived fission products ($^{99}$Tc, $^{129}$I and $^{135}$Cs) is well below the reference level at this time. However, while the solubility of the actinides in water is generally very low (except for $^{237}$Np), these fission products show relatively high mobilities in the geosphere and therefore need to be taken into account in the study of the nuclear waste disposal.

2.2 Partitioning and Transmutation

In order to reduce the burden on a geological repository, partitioning and transmutation (P&T) is being investigated as a means to reduce the long-lived radiotoxic inventory of the nuclear waste. Partitioning of the spent fuel means handling and separation of the actinides that are subject to transmutation, and fabrication of new fuels that will be dedicated to transmutation of waste. Transmutation refers to the transformation of a long-lived radiotoxic isotope into a short-lived or a stable product. As the transuranic elements (Np, Pu, Am and Cm) represent the largest part of the radiotoxic inventory in the long term, these isotopes are the main subjects of most transmutation studies. The transmutation of waste is relevant, both in the case of the continuous use of nuclear power as an important contributor to a country’s electricity production, as well as in the scenario of a nuclear phase-out.
2.2.1 Requirements for Efficient Actinide Burning

The best way to transmute the actinides is to fission them. This can be done efficiently and on a large-scale only in nuclear reactors. In principle, all types of nuclear reactors (thermal and fast, critical and sub-critical) can be used to burn the actinides, although fast reactors (FR) have some fundamental advantages over thermal reactors. The main goal with a transmutation reactor is to reduce as much as possible the net radiotoxic inventory of the spent fuel. The basic requirements for such a reactor to be efficient are therefore maximal fissioning of actinides combined with minimal production of new actinides (via neutron capture reactions). This can be achieved by adapting the following two objectives;

- The amount of fertile material (e.g. $^{238}\text{U}$) should be minimized in order to limit further TRU breeding.
- The neutron energy spectrum should be as hard as possible in order to favor fission over capture for the actinides.

Many of the actinides to be transmuted (the even-neutron numbered) have very low fission cross-sections for slow neutrons. This is due to the absence of the pairing binding energy effect in odd-neutron numbered nuclei, which is the intermediate step when an even-neutron numbered nucleus is fissioned by a neutron. This means that the fission over absorption ratio is essentially zero for these isotopes in a soft neutron spectrum and capture is strongly favored over fission. However, in the energy range between about 0.1 and 1.0 MeV, the fission cross-sections for these isotopes increase dramatically, which is indicated in Fig. 2, where the fission over

![Fig. 2. Fission cross-section over absorption cross-section for $^{239}\text{Pu}$ and $^{241}\text{Am}$ (ENDF/B-VI).](image)


absorption cross-section is displayed for $^{239}$Pu (even number of neutrons) and $^{241}$Am (odd number of neutrons). The obvious conclusion from this is that fast neutron reactors enhance the burning of the transuranic elements, while thermal reactors favor breeding. Another important advantage of fast reactor systems over thermal reactors is that the neutron economy is improved as the fission over absorption cross-section increases.

### 2.2.2 Adverse Safety Effects for Minor Actinide Fuel

The removal of uranium from the fuel and the adding of minor actinides (MA) have serious negative impacts on several safety parameters. This is true for all critical reactors, both thermal and fast. Firstly, the negative Doppler feedback, which is essential for the power control of a reactor, is significantly reduced. The main contribution to the Doppler effect comes from the broadening of the $^{238}$U capture resonances, and therefore it is brought to a minimum in uranium-free fuels. To solve this problem it has been proposed to add materials with absorbing resonances in the fast neutron energy spectrum, in order to restore, to some extent, the Doppler effect. However, if the fraction of americium is high, this will have very little effect (even with the addition of $^{238}$U). Due to the high absorption cross-sections of both $^{241}$Am and $^{243}$Am for fast neutrons, most of the neutrons would be absorbed by the americium before they reach the absorption resonances. Secondly, the fraction of delayed neutrons ($\beta$) is much lower for MA-containing fuels. This parameter is crucial for the safe operation of a reactor, since it establishes the margin to prompt criticality. $\beta$ is a specific property for each individual isotope and it is particularly large for $^{238}$U, while very small for the minor actinides. A third adverse effect, a positive void worth, arises when the coolant material (LBE, sodium or gas) in an FR is combined with its fast neutron spectrum. The situation is further deteriorated for high fractions of americium in the fuel.

### 2.2.3 Different Transmutation Strategies

The dedicated reactors envisaged to burn the minor actinides could be either fast reactor systems or so-called accelerator-driven systems (ADS). In a critical fast reactor (even for innovative systems) the maximum fraction of MA would be limited, determined by the safety margins, to a few percent. This implies that a large part of the nuclear reactor park would have to be contaminated by minor actinides. In order to overcome the safety deterioration for minor actinide-burning cores and to minimize the number of required dedicated reactors, the ADS concept has been proposed. In an ADS, a sub-critical reactor core is coupled to an intense external neutron source. The fact that the reactor is sub-critical, without the self-sustained neutron multiplication chain reaction, increases drastically the margin to prompt criticality and, hence, the safety of the reactor. Thanks to the sub-criticality, the safe operation of the reactor is less dependent on the reactivity feedbacks and the delayed neutron fraction, and this allows for a much higher flexibility in choosing the desired
proportions of isotopes in the fuel. As the fraction of minor actinides in the fuel is more or less unlimited, the number of dedicated MA-burning reactors in the reactor park can be kept relatively small.

Several different concepts concerning the handling of the nuclear waste have been proposed \cite{8,12}. Six of them, depicted in Fig. 3, are briefly treated here. Some countries, including Sweden, have adopted the “once-through” cycle (Strategy 1), in which all of the discharged spent fuel is considered as waste and must be disposed of in geological repositories (or possibly transmuted if the national policies are

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**Fig. 3.** Different fuel cycle strategies. Percentage in parentheses indicates a fraction of a nuclear power park corresponding to ADS.
changed). Other countries, such as France and Japan, reprocess their spent fuel and recycle the plutonium and the uranium in LWR-reactors. Only the minor actinides and the fission products are considered as waste, which is planned to be deposited in geological repositories, or, alternatively, to be transmuted. An extension of this strategy is the Pu-burning concept (Strategy 2), where the plutonium reprocessed from the MOX-LWR cycle is recycled indefinitely in a closed loop in fast reactors.

Concerning the transmutation of also the minor actinides, two major types of strategies have emerged. In the “mono-stratum” fuel cycle (Strategy 3), all of the waste is recycled and transmuted in conventional (albeit modified and possibly innovative) critical reactors. The other alternative is to burn the minor actinides in dedicated transmutation reactors (e.g. ADS), as in the “double strata” concept (Strategy 4). The main advantage of the double-strata fuel cycle is that only a limited number of dedicated transmutation reactors would be needed, as low as 5-10% of an entire nuclear power plant population, whereas a significantly higher fraction of conventional critical reactors would be needed to burn the waste in the mono-stratum concept.

In the original double-strata concept, initially proposed by JAERI in 1984 [13], the plutonium (and uranium) alone is recycled in critical reactors (LWR and FR) in the first stratum, while the minor actinides are directed to the second stratum to be burned in dedicated reactors. Stated simply, the first stratum would be devoted mainly to electricity production and waste mass reduction, while the second stratum would mainly concern radiotoxicity reduction. Moreover, the first stratum is based both on conventional types of reactors and well-known reprocessing technologies. The second stratum, on the other hand, has to deal with far more complicated challenges, such as the development of the dedicated MA-burners (ADS or advanced FR), as well as the techniques of reprocessing americium and curium (involving preferably pyrochemical methods). As depicted in Fig. 3, the plutonium from the standard LWR-UOX reactors is first reprocessed and recycled (once or twice) in conventional LWR-MOX reactors. In the final stage of the first stratum, the plutonium is recycled indefinitely in fast reactor systems in a closed cycle. All the minor actinides reprocessed in the first stratum are transferred to the second stratum, where they are recycled indefinitely. All losses from the reprocessing are considered as high-level waste and are sent to geological repositories. Calculations show that, if accelerator-driven systems are used in the second stratum, only about 5-7% of the reactor park would have to consist of dedicated MA-burning systems [1]. In the first stratum, approximately 70% of the electricity produced would be generated by conventional UOX-LWR, 11% by MOX-LWR and 19% by FR.

Alternatively, the possibility of recycling neptunium together with plutonium in FR in the first stratum has also been considered [1]. Neptunium is a less problematic fuel material than americium and curium and could probably be burned in critical reactors. It is also more easily reprocessed and can be separated in the same process (PUREX) with plutonium. However, as the radiotoxicity of neptunium is very low (Fig. 1), it could also be omitted and directly disposed of.
Several variations of the double-strata concept have been considered, diverging somewhat from the original proposal, depending on factors such as national policies and interests. The following two-component strategies, employing ADS in the second stratum, are possible scenarios [8];

- The plutonium produced in the LWR could be recycled once in MOX-LWR and then sent to the ADS together with the MA (Strategy 5). In this case, a fraction of 15% ADS of the reactor park would be required. As approximately 6 UOX-LWR produce the necessary actinide inventory to fabricate 1 MOX-LWR, about 12% of the reactor park would be of MOX type and the rest (~73%) of UOX type.

- All actinides (Pu and MA) produced in the conventional LWR are transferred directly to the ADS (Strategy 6). This scenario would require a slightly higher fraction of ADS (~21%) than the previous scenario.

Apart from choosing the type of reactor in which to transmute the radiotoxic inventory of the spent fuel, the options of how the elements to be transmuted should be added to the rest of the fuel, have to be considered. Generally two ways to incinerate the actinides into the reactor core are investigated. Either they could be dispersed homogeneously with the rest of the fuel (homogeneous mode) or they could be isolated in specialized fuel assemblies (targets) positioned in strategic places throughout the core. In the former case, the actinides are mixed with standard fuels. In a dedicated ADS burner, the fuel would consist of mainly (or only) TRU, which would probably be distributed homogeneously. If a strategy of the monostratum type is chosen, using FR to burn all TRU, the heterogeneous mode offers the advantage of reprocessing the standard fuel and the TRU-fuel separately. As a result, the target fuels could reach very high burn-up, since they do not have to be recycled with the rest of the core.

2.2.4 Radiotoxicity Reduction

The transmutation efficiency of an isotope is determined mainly by two basic factors, namely the burn-up in each fuel cycle and the losses from the reprocessing of the spent fuel. Stated simply, assuming a maximum burn-up of 10-20% implies that on average, each waste atom has to pass through 5-10 transmutation cycles. If, for example, a separation loss of 0.1% is assumed (realistic for plutonium), this means that the total accumulative losses are 0.5-1.0%. Consequently, the mass is reduced by a factor of 100-200. For americium and curium, the reprocessing methods are more complicated and larger separation losses can be expected.

Many different studies on transmutation scenarios have been performed, assuming different types of reactors and different fuel cycle strategies. When plutonium is recycled in LWR, as in the first step of the double-strata fuel cycle, a large amount of minor actinides are built up. Therefore, the radiotoxicity is actually increased. Considering also the indefinite Pu recycling in FR (but without the
recycling of MA), a total radiotoxicity reduction factor of 3-6 can be achieved [4]. Hence, the Pu recycling in the first stratum, apart from the economical utilization of the initial uranium resources, is mainly an efficient way to reduce the mass of the spent fuel, and thus to keep the number of required MA-burning ADS low.

Recycling of also americium reduces the total radiotoxicity by a factor of about 10, but produces significant amounts of plutonium and curium. The curium production is the reason why the radiotoxicity reduction is limited by a factor of 10. In order to reach reduction factors of up to 100, curium also has to be transmuted. However, the main problem with recycling of curium is that it is strongly radioactive and very hot, which complicates the reprocessing and the fuel fabrication methods. When obtaining the reduction factor of 100, a reprocessing loss of 0.1% for plutonium and 1.0% for the MA were assumed. The radiotoxicity reduction is strongly dependent on the reprocessing losses and if it could be decreased for the MA, a reduction factor of a few hundred could possibly be achieved.

2.2.5 Accelerator-driven Systems

The basic idea of accelerator-driven systems (ADS) is to operate a sub-critical reactor driven by an intense high-power proton accelerator. The fact that the reactor core is sub-critical compensates for the several adverse effects on the safety that the uranium-free MA-fuel introduces. The larger the sub-criticality is, the larger is the margin to prompt criticality. For most cases, a sub-criticality level in the order of 0.95 is assumed to be sufficient to compensate for the deteriorated safety, caused mainly by the loss in Doppler reactivity feedback, the decrease in fraction of delayed neutrons and the positive void worth. Since the fission multiplication chain reaction in a sub-critical core is not self-sustained, an external neutron source must be supplied to the core. These external source neutrons are multiplied in the sub-critical core and sustain a constant power production in the reactor.

The external neutron source in an ADS is sought to be produced by an intense proton beam impinging on a heavy-metal (probably lead or lead-bismuth) target and generating a large number of neutrons via spallation. A high-power proton accelerator is used to accelerate the protons to energies in the order of 1000 MeV and to guide them towards the spallation target, as depicted in Fig. 4. The spallation neutrons leak out from the target, after different kinds of interactions with the target nuclei, and are subsequently multiplied in the surrounding sub-critical core.

As was mentioned earlier, a fast neutron spectrum is needed in order to achieve high transmutation rates. For this purpose, metals like sodium, lead or lead-bismuth, or gas are eligible candidates as coolant materials. The most promising materials for the spallation target are lead or lead-bismuth eutectic, due to their favorable spallation-neutron production characteristics and their suitable thermal properties. Several different fuel forms are also being studied, of which some of the most promising are oxides, nitrides and metal fuels. These different design options will be further treated below.
2.3 Reprocessing Technologies

Reprocessing of nuclear spent fuel is being performed on an industrial basis in several countries (e.g. France, United Kingdom, India and Russia). The purpose of the reprocessing is to extract the uranium and plutonium from the discharged UOX-fuel and to recycle it as MOX-fuel. The remaining elements (minor actinides, fission products and reprocessing losses) are incorporated into the vitrified high-level waste, destined for interim storage or final disposal. Discharged MOX fuel can also be reprocessed if it is diluted with standard UOX fuel. The technique used in all industrial reprocessing plants existing today is the PUREX process (hydrochemical method). However, in order to recover also the minor actinides (and possibly some of the fission products), new reprocessing technologies are required. Generally, there are two types of processes that can be applied to the reprocessing of the nuclear waste; hydrochemical (aqueous) and pyrochemical (molten salt) processes [10,14,15].

2.3.1 Hydrochemical Methods

The PUREX Process

The most important hydrochemical reprocessing technique to separate U and Pu, and the only one used today on a commercial basis, is the PUREX process. It applies the organic extractant molecule, tri-butyl-phosphate (TBP) to extract U and Pu from the spent fuel, which has been dissolved in nitric acid (HNO₃). The TBP molecule has the property of extracting actinides with even oxidation states IV and VI from an acidic medium. The stable oxidation states in nitric acid of U and Pu are VI and IV,
respectively, and they are thus separated into the organic phase, while the rest of the dissolved nuclear waste remains in the aqueous phase. By changing the oxidation state of Pu from IV to III, Pu can then be separated from U in a second purification cycle. This process is referred to as the “standard PUREX process”. The recovery yield of U and Pu is high, close to 99.9%.

Np has similar chemical properties to those of U and Pu, but it exists in the nitric acid as a mixture of oxidation states V and VI, of which only the latter can be extracted by TBP. Hence, Np is only partly separated (~60%) together with the U and Pu stream. This fraction is then separated from U and Pu in the second purification cycle and added to the rest of the high level liquid waste (HLLW). The “improved PUREX process”, on the other hand, is based on the standard process, but with improved separation of Np. One method of recovering all Np consists in using an oxidizing reagent to change the oxidation state of the whole Np inventory to VI, allowing it to be extracted by TBP. Np recovery yields of 95% have been demonstrated and improvements towards 99% are under development. Research has also shown that it is possible to extract the long-lived fission products Iodine and Technetium in the PUREX process.

**Am and Cm Separation**

Americium and curium are both trivalent ions (their stable oxidation state is III) and since they cannot be extracted by TBP in the standard PUREX process, they remain in the aqueous phase. In order to separate these elements using hydrochemical methods, new extractant molecules are needed and several innovative processes are under development. The current approach to recover Am and Cm is usually divided into a three-stage process (also called the “extended PUREX process”). A flow sheet including the different processes separating the individual actinides are shown in Fig. 5. The first stage is based on the co-extraction of actinides (An) and lanthanides (Ln) from the rest of the fission products in the PUREX waste stream. One way of doing this is by using molecules from the diamide family, which has been applied in the DIAMEX process, developed by CEA (France). It has been demonstrated that recovery yields of 99.9% can be achieved in this process. Other processes developed (or under development) for this purpose are TALSPEAK (USA), DIDPA (Japan), TRUEX (USA) and TRPO (China).

The second stage aims at separating the actinides from the lanthanides. This separation is difficult for two main reasons. Firstly, the chemical properties of the two series of elements are very similar; importantly, they exist in nitric acid in the same oxidation state (III). Secondly, the mass ratio of Ln/An is high (in the order of 20 or higher), which indicates the need for a strongly selective mechanism in order to keep the Ln contamination low. One of the methods that has been developed to separate An from Ln is the SANEX process. It utilizes the fact that An(III) ions present a slightly larger extension of their electron clouds than do Ln(III) ions. It has been found that bis-triazinyl-pyridine (BTP) molecules are among the most suitable extractants for the An-Ln separation. BTP has an affinity for An(III) ions of more
than a hundred times that for Ln(III) ions, and the SANEX process has demonstrated a recovery yield of the actinides of 99.9%. Other extractants with high selectivity for actinides are used in the CYANEX 301 and the ALINA processes.

**Fig. 5.** Flow sheet of advanced hydrochemical reprocessing [14].

The separation of Am from Cm can be accomplished in the third stage, if it is intended to manage these elements separately. The separation is based on the fact that Am can exist in nitric solutions in several different oxidation states, whereas this property is not shared by Cm, which exists only as trivalent ion. In the SESAME process, Am is oxidized to IV or VI by an electrochemical method, after which it can be selectively extracted by the TBP molecule (same extractant as in the standard PUREX process). Alternatively, the SESAME process could be used to extract Am directly from the PUREX waste stream or after the co-extraction of An and Ln. This is technically more complicated, due to the larger quantities of waste and the larger amount of different elements that have to be managed.

**Advantages and Disadvantages with Hydrochemical Reprocessing**

Hydrochemical reprocessing is particularly suited for large quantities of standard LWR-UOX fuels, where long cooling times are allowed. Indeed, it is being successfully used worldwide on an industrial basis, utilizing the PUREX process to separate U and Pu from the nuclear waste. Moreover, the recovery yield of the other transuranic elements, using the different processes described above, have been demonstrated to be as high as up to 99.9% for both Am and Cm.
However, there are some important drawbacks in the hydrochemical reprocessing of MA-containing fuels. The first is the limited solubility in nitric acid for many fuel forms. This is especially the case for metal fuels, but also for oxide fuels with high fractions of Pu, possibly dispersed with an inert matrix. It has been shown that the solubility for Pu-containing fuels starts to deteriorate for Pu concentrations higher than about 30%. On the other hand, nitride fuels pose no problems for the PUREX reprocessing technique, which also enables the recovery of $^{15}$N. The other shortcoming of the aqueous methods is the low radiation stability of the organic extractant molecules used in the processes. This is particularly unfavorable when dealing with highly radioactive TRU-fuels and when multi-recycling is required, implying the need for short cooling-times.

### 2.3.2 Pyrochemical Methods

A promising alternative to hydrochemical reprocessing is the pyrochemical method, in which molten salts are used for the waste refining. Pyrochemistry has been used within the metallurgical industry for several decades and has also been investigated for the purpose of nuclear waste reprocessing (mainly in the USA, Japan and Russia). The high radiation resistance of the molten salts and the absence of neutron-moderating water make the solvent suitable for spent fuel separation. Pyrochemical methods for fast reactor fuel reprocessing are applied on a pilot scale in Argonne West (U.S.) and in Dimitrograd (Russia).

In the pyrochemical processes, the spent fuel is dissolved at high temperatures (500-800°C) in molten salt (mainly fluorides and chlorides). Separation of the actinides by electro-refining is the process that is primarily considered for nuclear waste reprocessing, although several different pyrotechnical methods are under development. One of the major advantages of pyrochemical reprocessing, compared to hydrochemical methods is the much higher radiation resistance of the inorganic reactants used in the process. This allows for the possibility to reprocess high-burn-up ADS fuels (including large fractions of minor actinides) with minimum cooling time, possibly as short as one or two years to allow for the decay of $^{242}$Cm. In addition to this, pyrochemical methods offer good solubility for most types of fuels. Another advantage, thanks to the radiation stability, is the higher compactness of equipment and the possibility of forming an integrated system between the irradiation and the reprocessing facilities. This would considerably reduce the expenses and the difficulties associated with the transport of the highly radioactive fuel. Finally, the absence of water in the solvent reduces the criticality risks during separation and pyrochemical processes are also more proliferation-resistant than aqueous methods.

Due to the advantages of the pyrochemical processes stated above, these methods are considered to be well suited for the second stratum fuel cycles, dedicated to the recycling of minor actinides. The hydrochemical methods, on the other hand, seem to offer the best solutions for U and Pu recycling in the first stratum. However,
pyrochemical methods involve far more complex challenges, in terms of theory and technical implementation, than hydrochemical methods. Several decades of research will probably be required before it can be implemented on an industrial scale as a mature technology. Among the drawbacks of pyrochemical methods are the difficulty of separating the individual actinides and the somewhat higher degree of fission product contamination. If the reprocessed fuel is planned to be recycled in a fast reactor spectrum, however, the contamination is of less importance. Another challenge is the management of the very hot and highly corrosive processing medium.

2.4 Advanced Fuel Options for ADS

The choice of fuel form envisaged for the transmutation of spent fuel in accelerator-driven systems (so-called advanced fuels) is still an open question. These fuels will contain high fractions of plutonium and minor actinides (preferably uranium-free) and will be irradiated to high burn-up, in order to achieve a high degree of transmutation. These new circumstances, compared to conventional reactors, imply the need for evaluating and developing alternative fuel materials. There is much experience and know-how about uranium-based fuel forms, but much less about the characteristics and fabrication technology of the corresponding Pu and MA-based ones. Several different options of advanced fuels are being investigated, oxides, nitrides and possibly metal fuels being the most promising [15]. Other fuel forms under investigation (not cited in this thesis) are carbide fuels, composite fuels and coated particle fuels.

There are currently two facilities in Europe that can be used for the handling and fabrication of americium and curium fuels; ATALANTE (CEA, Marcoule) and the MA-laboratory at the Institute for Transuranium Elements (JRC, Karlsruhe). The capacity is currently limited to a few grams for curium and about 150 g of americium (MA-lab). Facilities also exist in the U.S., Japan and in Russia.

2.4.1 Oxide Fuels

Compared to other advanced fuel options, oxide fuels have the advantage that their characteristics are known from the current industrial MOX fuel operation and fabrication technology. This advantage is limited, however, due to the significant differences that can be expected for MA-containing oxide fuels. The physico-chemical properties of uranium-free fuels are generally inferior to the corresponding uranium-based fuel forms. Among the negative consequences of going from a uranium-based fuel to fuels containing high fractions of Pu and MA are lower melting point (decreases continuously from $\text{UO}_2$ (3113 K) to $\text{AmO}_2$ (2448 K)), lower thermal conductivity and poorer chemical stability. Moreover, a general problem for all fuel forms with high MA content is the helium gas production, leading to intolerable swelling of the fuel. This has been demonstrated in the EFTTRA experiment, where uranium-free MA-containing fuels have been
irradiated. The helium is produced when $^{242}\text{Cm}$ (created from $^{241}\text{Am}$) disintegrates via alpha-decay.

Despite the degradation of the transuranium oxide fuel properties, compared to standard UOX or MOX fuels, they still exhibit the important advantage, over other advanced fuel options, of having comparatively high chemical stability. This implies relatively simple handling and fuel fabrication, which is very important when dealing with MA-containing materials. Oxide fuels are also stable in air, which makes the safety requirements for the fabrication process less demanding. The major drawback with oxide fuels is the low thermal conductivity, leading to high operating temperatures. Therefore, a shift of emphasis towards CERMET (ceramic-metal) oxides has recently been made, where the actinide oxide is dispersed in a metallic matrix, e.g. molybdenum. Two different types of oxide fuels are currently being considered in Europe as candidates for the ADS fuel cycle; CERCER (ceramic-ceramic) oxides (e.g. (Pu,MA)O$_2$-MgO) and CERMET oxides (e.g. (Pu,MA)O$_2$-Mo).

2.4.2 Nitride Fuels

The major advantage of nitride fuels is its favorable thermal properties (up to five times higher thermal conductivity for UN than for UO$_2$), whereas the melting temperatures are similar. This implies lower operating temperature, and/or higher linear power rating. The nitride actinides also show good mutual miscibility and it is therefore expected that the solid solution (Np, Pu, Am, Cm)N exists in a wide range of compositions. Other advantages associated with nitride fuels are the compatibility with the PUREX reprocessing method (which has some limitations for oxide fuels) and the chemical compatibility with water, air and stainless steel cladding materials. However, nitride fuels exhibit poorer chemical and thermal stability, which makes them more difficult to fabricate and which may lead to safety problems during power-temperature excursions, such as pressurization of the reactor vessel (caused by dissociation of the nitride and production of N$_2$ gas). Further, the addition of an inert matrix, e.g. ZrN, is expected to improve the thermal stability of nitride fuels. Another disadvantage with nitride fuels is the production of $^{14}\text{C}$. This requires the enrichment of $^{15}\text{N}$ [16], which has been proven feasible, but which will make the fuel fabrication process more difficult and expensive.

2.4.3 Metal Fuels

The main advantage of metallic fuels is that the thermal conductivity is high. For uranium-based alloys, the melting temperature is also reasonably high, around 1623 K. However, moving to uranium-free fuels with high contents of plutonium and minor actinides, the thermal properties are considerably deteriorated, both with respect to the melting point and the thermal conductivity. Consequently, in order to increase the margin to melting, the addition of an inert matrix is required, of which the most promising candidate is zirconium (melting point 2128 K). Another
disadvantage with metallic fuels is their incompatibility with liquid lead and lead-bismuth.

2.5 Coolant Options for ADS

Since the neutron spectrum in ADS must be hard in order to achieve high transmutation rates, the choice of core coolant is restricted to liquid metals (Pb, Pb-Bi, Na) or gas (He or CO₂) [8,17]. The main advantages of liquid metals are their attractive thermal properties and the possibility of operating close to atmospheric pressure, while the major drawbacks are the opacity (makes inspections and repair more difficult), the positive void worth [18] and the adverse chemical properties. Gas, on the other hand, is not chemically reactive and allows easy in-service, but needs very high pressure for efficient cooling. The molten salt concept, which exhibits many attractive properties, is also being investigated. However, this technique lies well beyond the state of the art and is not a realistic option for a demonstrator ADS within the near future.

2.5.1 Lead or Lead-bismuth

Lead or lead-bismuth as coolant medium has the advantage of being the same material as the spallation target, which will most probably consist of one of these materials. This would avoid the physical separation of the spallation target and the sub-critical system. Moreover, the very high boiling point (~1700 °C) is favorable in order to delay or prevent the core cooling problems that may arise in the unlikely event of a loss of heat removal accident (loss of coolant due to boiling is thus very unlikely). Pb and Bi also posses comparatively good neutronic properties, such as very low capture cross-sections, which is favorable for the neutron economy of the multiplying system. The physical properties of Pb, Pb-Bi and Na are listed in Table III. Their relatively high melting point, on the other hand, places constraints on the operating temperatures during shutdown and refueling, in order to avoid freezing of the coolant. Using pure Pb with a melting point of 327 °C, would require operating

<table>
<thead>
<tr>
<th></th>
<th>ρ [g/cm³]</th>
<th>T_melt [°C]</th>
<th>T_boil [°C]</th>
<th>k [W/m·K]</th>
<th>C_p [J/kg·K]</th>
<th>V[m/s]A</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>11.07</td>
<td>327.5</td>
<td>1749</td>
<td>16</td>
<td>150</td>
<td>2.5</td>
</tr>
<tr>
<td>Pb-Bi</td>
<td>10.24</td>
<td>123.5</td>
<td>1670</td>
<td>12.9</td>
<td>147</td>
<td>2.5</td>
</tr>
<tr>
<td>Na</td>
<td>0.857</td>
<td>97.7</td>
<td>883</td>
<td>71.6</td>
<td>1300</td>
<td>9.0</td>
</tr>
</tbody>
</table>

A: Maximum coolant velocity
temperatures of 400-600 °C, with no drop below 400 °C at any time. These high temperatures also further aggravate the structural material corrosion problems. However, using lead-bismuth eutectic (LBE) as coolant, with a melting point of only 123.5 °C, would enable the operating temperature to be reduced by about 200 °C, facilitating the shutdown procedure and relaxing the corrosion strains. The presence of Bi, however, leads to the production of the alpha emitter $^{210}$Po (half-life of 138 days) and the related radioactivity confinement concerns need to be managed. Moreover, the availability of Bi is limited and would probably allow only a smaller number of ADS based on LBE. Experience of Pb-Bi-cooled reactors is available in Russia (submarines), although the application in ADS needs more thorough research.

2.5.2 Sodium

Sodium is the best-known liquid metal coolant for fast reactor systems. There is extensive experience and knowledge from system design and operation of fast critical sodium-cooled reactors (e.g. Phenix, Bor-60, BN600, Superphenix) and most of the required compatible reactor components are available. Sodium has very good thermal properties (high thermal inertia and thermal conductivity, and relatively high boiling point), leading to efficient cooling and reducing the core cooling problems in the case of a loss of heat removal accident. Drawbacks connected to this choice of coolant are the violent chemical reactivity with air and water, as well as the positive void worth, which is particularly high when combined with a minor actinide-containing fuel [18].

2.5.3 Gas

Using gas (He or CO$_2$) as coolant would facilitate in-service inspection and repair of the reactor core, due to its transparency and the void worth is close to zero, as the gases are more or less transparent to neutrons. Moreover, He is inert and does not exhibit adverse effects concerning chemical compatibility with structural materials. The major drawback associated with gas as a coolant is that the system needs high pressure (50-70 bars), with the related consequences on the vessel loading and the increased risk of failures, possibly leading to loss of coolant. In the event of loss of coolant, the decay heat removal is expected to be more troublesome than with liquid metal cooling. Finally, as the core is pressurized, the physical separation of the spallation target and the sub-critical system is a disadvantage.

2.6 The Spallation Target

The design of the spallation target module should be based on the optimization of neutronic efficiency (maximum neutron yield), while ensuring the removal of the heat released in the spallation process as well as the physical and chemical integrity of the structural and target materials. The spallation target constitutes the physical
interface between the accelerator and the sub-critical core and it is one of the most innovative components of the ADS, for which only limited experience exists. It will be subject to severe radiation and mechanical stresses, induced both by the high power gradients generated by the intense proton beam (possibly up to 40 MW for a future commercial ADS) and by the material damage caused by high-energy particles and spallation products. The neutron production in the spallation process is most efficient for large nuclei and several heavy metals (e.g. Ta, W, Hg, Pb, Pb-Bi and U) have been investigated as possible target material candidates.

2.6.1 Liquid Heavy-metal Target

Two main options for the spallation target material are being considered, namely solid or liquid metal. While all current spallation sources operate with a solid metal target, the heavy liquid metal (HLM) choice appears to be the only realistic one for a large-scale ADS, due to the extremely high power densities. The main advantages of liquid metals are the superior heat removal capabilities and the significant reduction of the radiation damage to the target. However, the corrosion and erosion of the structural materials, the risk for release of volatile spallation products and the possible need for a beam window separating the beam tube from the target region are associated problems that have to be dealt with.

Among the studied heavy liquid metals, Pb and Pb-Bi eutectic (LBE) have emerged as the primary candidates. LBE has the clear advantage of having a low melting temperature (123.5°C compared to 327.5°C for pure Pb), which would facilitate the heating of the system before operation, as well as reducing the risk of target solidification in case of a beam interruption or a reactor shutdown. If LBE is chosen as core coolant material, a full compatibility between the target loop and the core coolant primary loop could also be achieved. The main drawback with LBE, as mentioned in the previous section, is the production of the alpha emitter, $^{210}$Po.

A third target material that is being considered is Hg, with the main advantages being the absence of $^{210}$Po-activity and the possibility of not having to heat up the system before operation. Hg is studied in the frame of the Spallation Neutron Source (SNS) [19], which is being constructed in Oak Ridge, U.S.. The spallation experiments are based on a pulsed proton beam of 1.4 MW (1.4 mA and 1000 MeV proton beam) impinging on a target of Hg. However, the high volatility of Hg imposes very strict requirements on the integrity of the system and the boiling point of 356 °C is probably not high enough for the operation temperatures required for a large-scale ADS. Another disadvantage of Hg is the high neutron absorption cross-section.

2.6.2 Window or Windowless Option

Another major choice that has to be made concerning the spallation target module is whether to connect the accelerator channel to the spallation target via a solid window, or to let the beam impinge directly on the HLM (windowless option). If a
window is used, it has to consist of a material of high strength and high radiation resistance, as it will be exposed to extremely high proton irradiation doses, high temperatures and large HLM pressures. Only limited experience from window irradiation is currently available, but it is estimated that the window would have a life-time not longer than 6 months, due to the heavy radiation damage. In the MEGAPIE project, carried out at the PSI, a Pb-Bi target will be irradiated by the SINQ proton beam (590 MeV and 1.8 mA). The beam and the target will be separated by a beam window, which is expected to reach an irradiation dose of about 10 dpa (displacement per atom) during a period of six months.

A windowless design avoids the problems associated with high-temperature window stresses (although there is a cold window higher up in the accelerator channel), but involves other technical difficulties. The main challenge is to manage the metal and the spallation product evaporation contaminating the beam line vacuum. This option will be experimentally investigated in the MYRRHA project [20], where a windowless LBE target will be irradiated by a 350 MeV and 5 mA proton beam. The proton beam will drive a sub-critical core of about 30 MW thermal power.

2.6.3 The Spallation Process

The nuclear process of spallation can be divided into two stages, as shown in Fig. 6. In the first stage, called the “intranuclear cascade”, a high-energy particle impinging on a target nucleus interacts directly with the individual nucleons inside the nucleus,

![Fig. 6. The different stages of the spallation process.](image-url)
the interactions being described by particle-particle cross-sections. High-energy secondary neutrons, protons or pions may be emitted as a result of this interaction. The nucleus is left in a highly excited state, the energy being distributed throughout all the nucleons of the nucleus. In the second stage, which is much slower, the highly excited nucleus undergoes “evaporation”, releasing nucleons or light ions with an energy of typically about 1 MeV. The nucleus might also undergo fission in this stage. When the nucleus reaches an energy level that is less than about 8 MeV above its ground state, further de-excitation occurs by the emission of gamma rays.

In a single spallation reaction, about 15 neutrons are created from a 1000 MeV proton impinging on a Pb nucleus. However, some of these emitted neutrons are energetic enough to induce further spallation reactions (or n,xn-reactions) in other target nuclei. This multiplication thus gradually leads to a larger number of neutrons leaving the target. In a large Pb-Bi target, about 30 neutrons are typically created for each 1000 MeV proton.

2.6.4 High-energy Codes and Models

High-energy particle transport, e.g. in spallation interactions, is usually performed by a high-energy Monte Carlo code, based on several different physical models. For reactor problems, these codes are generally coupled, below a certain cut-off energy (usually 20 MeV), to a neutron transport code, such as MCNP [21]. There are presently a number of different models and computation codes for simulating high-energy particle transport, of which HETC [22,23] and LAHET [24] are two of the most widely used. The physical models used for high-energy interactions are in most codes divided into the following different categories; Intranuclear Cascade Models (INC), Multistage Pre-equilibrium Models (MPM) [25], Evaporation and Break-up Models and High-Energy Fission Models.

Intranuclear Cascade and Pre-equilibrium Models

The concept of the INC model was developed several decades ago in the early stages of high-energy nuclear interaction modeling. It is a relatively fast model, in terms of computation speed, and within some energy ranges it is the only available tool for simulating hadron-nucleus interactions. As a result, several different INC models have emerged and have become more and more refined and widespread.

In the INC models, the incoming particle interacts directly with individual nucleons inside the nucleus. The nucleus is assumed to be a cold, free gas of nucleons, confined within a potential that describes the nucleon binding energies and the nuclear density as a function of the radius. Standard Wood-Saxon potentials are used and the quantum effects of Pauli blocking are generally taken into account. Some of the INC models available today are Bertini [26], ISABEL [27], the Cugnon [28,29] INCL models, CEM [30] and FLUKA [31]. Bertini is the oldest and most widely used INC model and is the default e.g. in the MCNPX code [32], which is based on the LAHET code system. Nevertheless, the Bertini model has shown some
disagreement with experimental results, obtained at the Saturne accelerator at Saclay (France) [33]. For example, it largely over-predicts the low-energy neutron production resulting from high-energy protons impinging on different heavy-metal targets. The Cugnon INCL model used by the TIERCE code [34], on the other hand, yields neutron production rates in good agreement with the experiments, although this model has some other deficiencies that have yet to be overcome.

After the termination of the INC model, a multistage pre-equilibrium model (MPM) may optionally be applied for subsequent de-excitation of the residual nucleus. During this intermediate stage, the excited nucleus may emit nucleons or light nuclei. When the Bertini INC model is used in the LAHET code system, the use of this pre-equilibrium model is recommended for most cases. It was also shown in the SATURNE experiments that the Bertini model followed by the MPM clearly improves the performances.

Evaporation and Breakup Models

At the completion of the INC model (and the MPM, if used), an evaporation model or, for lighter nuclei, the Fermi-Breakup model may be applied for the de-excitation of the nucleus. The energy of the highly excited residual nucleus is dissipated by evaporation of neutrons, protons, deuterons, tritons, $^3$He and alpha-particles. The most widely used evaporation model is the Dresner Model [35], usually associated with the Atchison [36] fission model, but the GSI model [37], for example, also appears to be a promising alternative.

For the disintegration of light nuclei (A ≤ 22), the Fermi-Breakup model [38] has replaced the evaporation model. The de-excitation process is treated as a sequence of simultaneous breakups of the nucleus into two or more products. The products may be a stable or unstable nucleus, or a nucleon. Any unstable nucleus is subject to further breakups until all products are stable.

High-energy Fission Models

Instead of undergoing evaporation after the intranuclear cascade, the highly excited nucleus may also undergo fission. There are two main models for fission induced by high-energy interactions; the Rutherford Appleton Laboratory (RAL) Model [39] and the ORNL Model [40]. The RAL model, which is the default e.g. in MCNPX covers fission for nuclei larger than Z=71, while the ORNL model only treats actinides.

2.7 Accelerator Technology

The operation of an ADS will require a high power proton accelerator (HPPA) and the two types of accelerators that are being considered are linacs and cyclotrons [8,12,41]. The most powerful linac existing today is the LANSCE accelerator at Los Alamos, operating with a maximum proton energy of 800 MeV and a mean beam current of 1 mA, whereas the biggest cyclotron is the separated sector unit at the
Paul Scherrer Institute (PSI) with maximum 590 MeV and an average current of 1.8 mA. There are also several large accelerators that are currently under construction, some of the most important projects being the Spallation Source Neutron (SNS) (1.4 mA and 1.0 GeV pulsed linac) [19], the KEK/JAERI project (600 MeV linac coupled to a 3 GeV and a 50 GeV synchrotron) and the European Spallation Source (ESS) (1.33 GeV pulsed linac) [42,43]. Furthermore, the LEDA (Low Energy Demonstration Accelerator) accelerator, which has operated at a current of about 100 mA for many hours [44], is an important proof that an accelerator of such strength can be built.

However, all of these accelerators are far from meeting the requirements imposed by the operation of a future large-scale ADS. The most important areas for accelerators envisaged for ADS application, where significant development and technological improvement are needed, concern performance, reliability and availability, and operation and safety.

### 2.7.1 Accelerator Performance

**Required Proton Beam Intensity**

A proton energy in the order of about 1 GeV combined with an accelerator current of 20-30 mA (20-30 MW power) will probably be required for a large-scale industrial ADS (~1000 MWth), while about 5 mA using 600 MeV protons (~3 MW power) is estimated to be enough for an experimental facility with a power in the order of 100 MWth, such as the XADS demonstrator [8]. The required proton energy lies well within the present capabilities of accelerator technology, but the high currents require significant improvement. It has been shown that accelerator powers of up to 100 MW (100 mA, 1 GeV) using super-conducting linacs are feasible, constructional costs being the main limiting factor. On the other hand, 1 GeV and a few mA (possibly up to 10 mA) are considered as the upper limits of what is achievable for cyclotrons. Consequently, both linacs and cyclotrons could be used for a 100 MW experimental facility, while only linacs can be eventually upgraded to a future commercial ADS.

Assuming that the total core power ($P_{\text{tot}}$), the proton energy and the reactivity range ($k_{\text{min}}<k_{\text{eff}}<k_{\text{max}}$) have been fixed, the proton beam current needed to drive the sub-critical core ($i_p$) can be determined. It can be shown that

$$i_p = \frac{1 - k_{\text{eff}}}{k_{\text{eff}}} \cdot \frac{P_{\text{tot}} \cdot \bar{\nu}}{\bar{E}_f \cdot Z \cdot \phi^*}, \quad (1)$$

where $P_{\text{tot}}$ is the total core power, $\bar{\nu}$ the average neutron yield per fission, $\bar{E}_f$ the average energy released in a fission and $Z$ the number of neutrons produced in the target per incident proton [45]. If $P_{\text{tot}}$ is expressed in units of MW and $\bar{E}_f$ in MeV
in the equation, \( i_p \) is given in mA. \( \varphi^* \) is the neutron source efficiency, which is defined as the ratio of the average importance of the external source neutrons to the average importance of fission neutrons [46,47]. The source efficiency is treated in more detail below and Eq. (1) is derived in Appendix A. Eq. (1) shows that, apart from the strong dependence on the reactivity level, the proton current is inversely proportional to the number of neutrons per proton (\( Z \)) and to the efficiency of these neutrons (\( \varphi^* \)). Assuming a total core power of 100 MWth, a \( k_{\text{eff}} = 0.95 \) and using typical values of the neutron yield (\( Z \sim 30 \), assumed for 1000 MeV protons impinging on a large Pb-Bi target) and the neutron source efficiency (\( \varphi^* \sim 1.0 \)), a required current (\( i_p \)) of 2.6 mA is obtained. A fuel type with \( \bar{\nu} \approx 3.0 \) and \( \bar{E}_f \approx 200 \) MeV has also been assumed. Note, however, that \( Z \) and \( \varphi^* \) are dependent on several system parameters, such as the proton beam energy, the target geometry and, to some extent, the core design. For example, for smaller target radii the number of neutrons per proton decreases, but their efficiency is strongly enhanced. As a result, the proton current needed to sustain constant core power decreases with decreasing target radius [Paper III], in particular for a Pb-Bi-cooled system. The required proton current and beam power for some different reactivity levels of a 100 MWth core is given in Table IV. With a proton energy of 1000 MeV and the lower limit of the sub-criticality being \( k_{\text{eff}} \sim 0.95 \), it can be concluded that for a large-scale ADS with a thermal power of 1000 MW, a proton current of 25-30 mA will be required, which corresponds to a proton beam power of 25-30 MW. Such large beam powers will be possible to achieve only with a superconducting linac.

### TABLE IV

<table>
<thead>
<tr>
<th>( k_{\text{eff}} )</th>
<th>( E_p = 600 ) MeV (( Z \sim 15 ))</th>
<th>( E_p = 1000 ) MeV (( Z \sim 30 ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>( i_p )</td>
<td>( P_{\text{acc}} )</td>
<td>( i_p )</td>
</tr>
<tr>
<td>0.90</td>
<td>11.5 mA</td>
<td>6.9 MW</td>
</tr>
<tr>
<td>0.95</td>
<td>5.3 mA</td>
<td>3.2 MW</td>
</tr>
<tr>
<td>0.98</td>
<td>2.1 mA</td>
<td>1.2 MW</td>
</tr>
</tbody>
</table>

A: \( E_p \) is the proton energy

---

**The Proton Beam Characteristics**

The main characteristics of the proton beam, that is its energy and intensity, will be determined mainly by two physical quantities, namely the number of neutrons produced per proton and per GeV (\( Z/E_p \)) and the amount of energy deposited in the target window. The first quantity is an approximate measure of the efficiency of the proton beam, in terms of number of produced neutrons for a given amount of
accelerator power. This is an increasing function of proton energy and reaches its optimum level at about 1000-1500 MeV (Fig. 7). Expressing the accelerator power required to feed the sub-critical core as (using Eq. (1))

\[
P_{\text{acc}}(MW) = i_p(mA) \cdot E_p(GeV) = \frac{1 - k_{\text{eff}}}{k_{\text{eff}}} \cdot \frac{P_{\text{tot}} \cdot \phi \cdot E_p}{E_f} \cdot \frac{Z}{\phi^*},
\]

it can be seen that the power is minimal when \(Z/E_p\) reaches its maximum. It is shown in Table IV that the required current using a 600 MeV proton beam is about twice that for a 1000 MeV beam and the required accelerator power is thus larger by about 20%. This is in good agreement with Fig. 7, which shows that the neutron yield per proton and per GeV is also 20% larger for 1000 MeV protons than for 600 MeV protons. Looking at Eq. (2), it can be seen that in order to obtain the exact relationship between the neutron yield and the accelerator power, the number of neutrons should be multiplied by their efficiency (\(\phi^*\)), which can vary considerably for different target and core designs. This product is called “proton source efficiency” (\(\psi^*\)) and is treated in [Paper III]. However, when varying the proton energy, as in Fig. 7, the change in \(\phi^*\) is rather small, as well as the change in curve shape.

The second quantity, the energy deposited in the target window (due to ionization losses) is a decreasing function of proton beam energy. Since the required current is also lower for higher proton energies, the window damage is strongly reduced by increasing the proton energy. Hence, considering the physical properties of the beam-window-target interactions, a proton energy in the order of 1 GeV or higher is clearly favored.

![Fig. 7. Neutron yield for proton beams of 400 MeV to 2.5 GeV energy, impinging on a Pb-Bi target with radius = 40 cm and height = 75 cm.](image)
Reliability and Availability

The reliability and continuity of the accelerator beam is a new and challenging constraint on the accelerator technology and will require considerable improvement of a large number of accelerator components. A reduction of beam interruptions of a few orders of magnitude is needed in order to meet the requirements for the coupling of an accelerator to an industrial sub-critical reactor.

The beam trips are generally divided into two categories; those shorter and those longer than one second. Current research shows that the beam interruptions shorter than about one second do not significantly affect the thermodynamics of the reactor or other plant parameters (e.g. thermal power, temperature, primary flow, pressure). Therefore, no specific limit on the frequency of trips is required for this category. On the contrary, beam trips longer than one second will lead to a variation of these parameters or even to a plant shutdown and a frequent repetition of such trips can significantly damage the reactor structures. The number of allowable long-duration beam trips depends on the properties of the structural components, but it is generally considered that some hundreds per year would be acceptable. However, considering the availability and the cost efficiency of the reactor operation, the limit of beam trips long enough to cause a shutdown will be much lower, maybe in the order of 10 per year. This value, which is still significantly higher than the average number of shutdowns for present industrial nuclear power plants, is acceptable, since the main purpose of ADS is transmutation rather than electricity production. Hence, the major challenge within accelerator technology, in terms of improving reliability and availability, is the reduction of long-term beam interruptions.

Operation and Safety

The control of the power and the monitoring of the reactivity level of the core pose other challenges for the operation of a source-driven sub-critical reactor. Concerning the time structure of the proton beam, a pulsed mode operation appears to be promising for this purpose, since it allows for a repeated beam interruption (~1 ms). For example, using the Pulsed Neutron Source Method, the sub-criticality can be determined by monitoring the neutron flux decay process following a source pulse. As long as the frequency is high enough, no penalties in terms of thermal stresses are expected. Other safety functions that have to be taken into account are the containment of radioactive materials and the radiological protection of the operators under normal operating conditions.

2.7.2 Linear Accelerators

Basic Technology

The basic principle of a linear accelerator (commonly called linac) is that the charged particles are accelerated, either by electrostatic fields or oscillating radio-frequency (rf) fields, along a straight line. The particles travel through a series of
hollow “drift tubes”, alternately connected to the opposite poles of an ac voltage source (Fig. 8). The energy transfer to the particles occurs in the electric field between the tubes, whereas the inside of the tubes are field-free (hence the name, “drift tube”). The polarity of the voltage is reversed while the particles are traveling inside the tubes and the lengths of the tubes are chosen so that the particles reach the gap between the tubes at the moment when the electric field is accelerating. As the velocity of the particles increase, so must the length of the tubes also increase, approaching a constant value as the particles become relativistic. In order to reach high energies, since the final energy of the particles is equal to the sum of the voltages to which they have been exposed, either the number of tube segments or the voltage of the rf-source may be increased. As the velocity of the particles quickly becomes large, it is desirable that the rf-frequency is high, in order to keep the tube lengths reasonably short.

![Fig. 8. Principle design of the Widerö accelerator.](image)

**The Linac Envisaged for ADS**

The HPPA linac envisaged for ADS can schematically be divided into three different parts; the injector, the intermediate section and the superconducting high-energy section. In a typical design proposal, depicted in Fig. 9 [48], the injector consists of three parts with the first stage being the ion source, producing a high-intensity proton beam. The ECR (electron cyclotron resonance) ion sources have proved very suitable for producing intense proton beams and to be highly reliable. The source is followed by an RFQ (radio-frequency quadrupole), accelerating (to 5 MeV), focusing and bunching the beam, and a DTL (drift tube linac) accelerating the protons to about 10 MeV.

The intermediate section consists of a number of classical normal-conducting (copper structure) DTL, in which the particles reach an energy of about 85 MeV. At higher energies, in the last section of the linac, superconducting cavities and focusing quadrupoles are needed. This is the longest section and brings the protons
up to their final energy (possibly 450-600 MeV for a demonstrator or about 1000 MeV for a prototype ADS). In addition to the first 100 m of the two first parts, approximately 180 m are needed to reach 450 MeV, while about 300 m are needed to reach 1000 MeV. Alternatively, a transition to super-conducting technology at lower energies (~20 MeV) has been considered, e.g. in the TRASCO project and in the US roadmap for ATW [49].

![Typical layout for a super-conducting HPPA linac](image)

Fig. 9. Typical layout for a super-conducting HPPA linac [48].

The main advantage of super-conducting radio-frequency-cavities (SCRF) compared to normal-conductors is the very high efficiency of the power transferred from the rf-transmitters to the proton beam. Using classical copper structure cavities, about half of the power is dissipated in the walls, while for SCRF cavities almost 100% efficiency can be reached. This leads to considerable economical advantages as well as relaxed constraints on the construction of the rf-source. Besides this obvious economical impetus for using SCRF cavities, they are also favored for several technical reasons. One is that large beam openings, not achievable in classical linacs, drastically reduce the problems associated with the activation in the structural components induced by beam halo. Another reason is that stronger accelerating fields are possible (10 MeV/m is foreseen, compared to 1.6 MeV/m for classical copper cavities), which considerably reduces the length (and costs) of the high-energy section.

### 2.7.3 Cyclotron Accelerators

A cyclotron is a circular accelerator consisting of two large dipole magnets and two semicircular metal chambers (called Dee:s because of their shape), in which the particles orbit (Fig. 10). The Dee:s are connected to an oscillating voltage, generating an alternating electric field in the gap between the two Dee:s, in which
the particles are accelerated. When they are inside the Dees, however, they sense no electric field and follow a circular path until they reach the gap and are accelerated again. In this way, the particles that are emitted at the center of the device follow a spiral path, gaining a certain amount of energy each cycle, until they become energetic enough to leave the accelerator.

A difficulty with this type of classical cyclotron arises when the particles become relativistic, causing them to become out of phase with the alternating voltage. For protons, the maximum achievable energy due to this effect is about 40 MeV ($\gamma = 1.04$). In order to overcome this problem, either the frequency must be modulated or the magnetic field must increase with the orbital radius. This has been accomplished, for example, by the development of the frequency-modulated synchro-cyclotron. On the other hand, increasing the magnetic field by increasing the radius has the undesirable effect of defocusing the beam. However, focusing can be restored by dividing the dipole magnets into sectors with alternating high and low magnetic fields (isochronous cyclotron). The strongest focusing effect is obtained in a separated sector cyclotron. This is the principle for the 590 MeV PSI cyclotron, which is constructed using 8 separate magnet sectors.

Cyclotrons may be a realistic alternative to linacs as the accelerator to drive an ADS if a relatively small reactor is envisaged. The maximum achievable beam power is estimated to be 5-10 MW, so a thermal output power of about 100 MW appears to be the upper limit for a cyclotron-driven ADS. An alternative that is being considered is to operate several independent cyclotrons. In this scenario, a 500 MW core could possibly be managed. This would also increase the reliability of the proton beam and given the lower unit cost of a cyclotron compared to a linac, it could equally be an economically favorable option. Most of the conceptual design proposals of ADS cyclotrons are based on a version of the PSI ring cyclotron upgraded to about 1 GeV and 10 mA, yielding a maximum power of 10 MW.
2.8 The European Roadmap for the Development of ADS Technology

2.8.1 XADS

The main goals of the European Roadmap are to propose a technological route for the transmutation of nuclear waste in accelerator-driven systems and to prepare a detailed plan for its first phase; the realization of a 100 MW experimental ADS (XADS) [8,50]. The XADS program, which is envisaged to cover the time-period of the 6th and the 7th Framework Programs, is the first step in the European Roadmap for development towards a large-scale and industrial application of ADS. The scheduled milestones of the Roadmap are listed in Table V. During the operation of the XADS, which will use conventional MOX fuel, the basic physical principles of an accelerator-driven system (mainly the coupling between the proton accelerator and the sub-critical core) will be studied and demonstrated. The MOX fuel could be either existing fuels from the SNR300 or Superphenix (SPX) reactors, or new MOX fuels specifically constructed for the XADS. The latter choice has the advantage that the fuel design can be adapted to the design of the reactor system.

<table>
<thead>
<tr>
<th>Year</th>
<th>Event</th>
</tr>
</thead>
<tbody>
<tr>
<td>2005</td>
<td>Start of detailed design of XADS</td>
</tr>
<tr>
<td>2008</td>
<td>Start of construction of XADS</td>
</tr>
<tr>
<td>2013-2015</td>
<td>Start of operation of XADS</td>
</tr>
<tr>
<td>2017</td>
<td>Decision on how to go to XADT</td>
</tr>
<tr>
<td>2025</td>
<td>Start of operation of XADT</td>
</tr>
<tr>
<td>2030</td>
<td>Start of operation of PROTO-ADT</td>
</tr>
<tr>
<td>2040-2050</td>
<td>Start of industrial application</td>
</tr>
</tbody>
</table>

It has been agreed that a thermal power of about 30-100 MW is suitable for the XADS. A power level of 100 MW would require a maximum accelerator current of about 5 mA and an accelerator power in the order of 3 MW. The proton beam energies that are being considered lie between 600 and 1000 MeV. It has also been agreed that the spallation module should be separated from the primary coolant, whereas the choices of many other system parameters, such as coolant and spallation target materials, are still unresolved. For the spallation module, the choice between the window and windowless concept will be based on the results from the MEGAPIE and the MYRRHA experiments, respectively. The XADS will also have the role of an irradiation tool and a limited number of dedicated MA-based fuel assemblies will be inserted in the reactor for irradiation studies. These advanced fuels, characterized by a high content of Pu and minor actinides and preferably
uranium-free, will be irradiated to relatively high burn-up in order to achieve high transmutation rates. The start of the construction of the XADS is scheduled for 2008 with the start of operation beginning 2013-2015.

2.8.2 XADT and the ADS Prototype

In the second phase of the ADS development, the MOX fuel will be replaced by dedicated fertile-free fuel, thereby allowing full demonstration of the transmutation process. This will be realized either by converting the XADS to an experimental accelerator-driven transmuter (XADT) or, should this not be feasible, by constructing a new facility. A high priority with the XADS design phase will be to provide the maximum practical flexibility in order to make it possible to convert the plant to XADT without major modifications. According to the present time-schedule, the decision whether this can be accomplished or whether a new facility for the XADT has to be constructed must be taken around 2017-2018. After this, the operation of the XADT could possibly begin around 2025. It is assumed that the separation processes of minor actinides and the fabrication technologies of dedicated MA-based fuels that are currently under development will be ready by the time the XADT program starts.

Around 2030, the construction of a prototype ADS could be started. This prototype must have all the features of an industrial ADS deployed at a later stage (power, coolant, fuel, spallation module etc.). Essentially, it should also be based on the same technology as the XADT, except that it would be larger. Until this time, a spallation module, including an accelerator of about 20 to 40 MW power and with availability far beyond its current status, must be developed and constructed. Apart from the accelerator technology, the realization of a large-scale ADS will also rely on several other independent factors, such as the development and construction of advanced processing and fabrication plants, various safety issues and many different technological challenges. Eventually, if the operation of the prototype is successful, accelerator-driven transmutation systems could be applied on a large and industrial scale from around 2040.
Chapter 3

Studies of Sub-critical Neutronics in MUSE-4

As a step towards a future ADS, the MUSE experiments provide a valuable mean for investigating the physics of sub-critical systems in the presence of an external source, and for validating experimentally the theoretical methods and calculation tools developed to characterize such a system. A major part of this thesis has been devoted to numerical Monte Carlo simulations investigating the neutronics and the neutron source effects for a model representative of the sub-critical MUSE-4 experiments. Most of the results from these studies have been presented in [Paper I and II].

3.1 The MUSE Experiments

3.1.1 The MASURCA Facility

MASURCA is the experimental reactor at CEA/Cadarache dedicated to neutronic studies of accelerator-driven systems in the framework of the MUSE program. The MASURCA core is rather small, with a height of about 60 cm and a radius of about 50 cm. The reactor power is low (maximum 5 kW) and the core cooling is provided by air.

The MASURCA reactor has a very flexible design and several different core configurations are possible. The core can be loaded with different fuels (e.g. thorium, uranium and plutonium), different coolants can be used (sodium, lead, gas) and different levels of criticality are possible, both critical and sub-critical. A schematic view of MASURCA, coupled to the GENEPI neutron generator, is shown in Fig. 11. The core of MASURCA is composed of a number of quadratic sub-assemblies (10.6×10.6 cm), each of them being composed of 32 (U,Pu)O₂ rodlets and 32 Na rodlets (simulating the coolant medium) with a diameter of 1.27 cm.
3.1.2 The MUSE Experimental Programs

In order to validate experimentally the main physical principles of an accelerator-driven reactor, the basic idea in the MUSE experiments is to separate the sub-critical multiplication process from the external source characteristics. This can be done by using a well-defined neutron source (in terms of energy and position) to drive the sub-critical core. The methodology is a step-wise approach, starting from a known and tested critical reference configuration. In the first step, the characterization of the multiplying medium alone is performed, after which the insertion of an external source and the investigation of the response in the medium is made.

The MUSE program started at CEA/Cadarache in 1995 with the short MUSE-1 experiment [51]. In MUSE-2 (1996) [47], diffusing materials (sodium and stainless steel) were placed around the external source in order to modify its neutron importance and to study the effects from it. In both MUSE-1 and MUSE-2, an intense $^{252}$Cf neutron source was used as the external source. In MUSE-3 (1998) [52], the californium source was replaced by the neutron generator, SODERN/GENIE26, producing 14 MeV neutrons by (d,t)-reactions. Several levels of reactivity were investigated and experiments with different “buffer zones” (sodium, stainless steel and lead) were performed.

In the on-going MUSE-4 experiments [5] (started in 2000 in international collaboration via the 5th Framework Program of the European Community), the neutron generator GENEPI [6], especially developed for the MUSE-4 experiments, was introduced. With its improved performances (in terms of source intensity and quality of the neutron pulse) and the use of both (d,d)- and (d,t)-reactions it has extended the range of experimental possibilities and has considerably improved the accuracy of the measurements. For example, the accurate dynamic measurements,
based on the pulsed mode operation of GENEPI, enable experimental reactivity determination of the sub-critical multiplying media. The techniques that are investigated for this purpose are the pulsed neutron source method, the inverse kinetics method, the Rossi-α method and the Feynman-α method [53]. At the end of the MUSE experiments, a validated experimental technique (including experimental uncertainties) will be proposed for the development of a reactivity meter that can be envisaged for a future ADS.

The geometry of the second sub-critical configuration of MUSE-4 (SC2) is shown in Fig. 12. The GENEPI deuteron accelerator tube is introduced horizontally at the core mid-plane and the deuterium or tritium target is located at the core center. The neutron source is surrounded by a lead “buffer” medium in order to simulate the diffuse properties of a spallation source. The fuel is MOX fuel with 72% ^{238}U, 21% ^{239}Pu and 5% ^{240}Pu with the addition of small amounts of some other actinides. The simulated coolant medium is sodium. A more detailed description of MUSE-4 is given in [54].

![Fig. 12. XY-cross-sectional view of the second sub-critical configuration (SC2, $k_{\text{eff}} \sim 0.97$) of the MUSE-4 experiments. The core is composed of a number of sub-assemblies with the dimension 10.6\times10.6 \text{ cm}.

The following experimental configurations, all with sodium as coolant medium, are being studied in the MUSE-4 experiments;

- One critical reference configuration (GENEPI shut off). The reactivity will be experimentally determined by classical pilot rod shutdown measurements.

- Three sub-critical configurations (SC0 with $k_{\text{eff}} \sim 0.994$, SC2 with $k_{\text{eff}} \sim 0.97$ and SC3 with $k_{\text{eff}} \sim 0.95$)
- Two complementary asymmetrical configurations with a $k_{eff}$ of about 0.95 and 0.93, obtained from the reference configuration and SC1, by complete insertion of one of the safety rods.

It was also decided to include, in an extended phase of MUSE-4, a configuration partially cooled by lead. In MUSE-5, experiments with a fully lead-cooled core and with a gas-cooled core will be carried out.

Most neutronics computational tools and data libraries existing today have not yet been sufficiently validated for accelerator-driven systems coupled to an intense external neutron source. In the MUSE-4 collaboration, relatively large discrepancies between various codes used to compute the reactivity of a MUSE-type core have been observed. However, the MASURCA reactor used in the MUSE experiments, together with the introduction of GENEPI, offers a good opportunity for test and validation of available standard and new reactor codes. For this purpose, a benchmark on computer simulation of MASURCA critical and sub-critical experiments, particularly concentrated on the MUSE-4 experiments, has been proposed by the OECD Nuclear Energy Agency [55]. One of the objectives of the benchmark is to define a reference calculation route (including nuclear data, methods and calculation codes) for the prediction of various neutronic parameters of an ADS.

As complementary experiments to MUSE-4, the SAD experiments (Sub-critical assembly in combination with the proton accelerator in Dubna) will be performed within an expanded collaboration of the MUSE-4 program. Different spallation neutron sources (Pb, W, Pb-Bi targets) produced by a 660 MeV proton synchrotron will be investigated. The experiments will permit the characterization of the propagation of spallation neutrons into the different materials (e.g. target, fuel, coolant and structural materials) encountered in a possible future ADS.

3.1.3 The GENEPI Neutron Generator

GENEPI (GEnerateur de NEutrons Pulsés Intenses), depicted in Fig. 13, is a high-intensity pulsed neutron generator constructed by CNRS/ISN/Grenoble and has been especially developed for the MUSE experiments in the MASURCA facility. A duoplasmatron providing short pulses of deuterons and a high-intensity electrostatic accelerator are combined to create the 240 keV deuteron beam, which is guided towards either a deuterium or a tritium target in the center of the core. The (d,d)- and the (d,t)-reactions produce neutrons of, on average, 2.67 MeV or 14.1 MeV, respectively. The GENEPI deuteron guide is introduced horizontally at the core mid-plane and the deuterium or tritium target is located at the core center. The main characteristics of GENEPI in the (d,t)-mode are given in Table VI. The special features of GENEPI are the rather high intensity (∼50 mA peak) combined with a pure and very short (1 µs) and sharp-edged pulse, which is desirable in order to be able to study the pure neutron propagation out into the fuel of the reactor, independently of the influence of the neutron source. The frequency can be varied.
from 10 Hz to 5 kHz and each pulse comprises 25 million neutrons, which corresponds to about $10^{11}$ neutrons per second for the maximum frequency. GENEPI can be operated in either continuous or pulsed mode. The beam has a gaussian profile with a diameter of 20 mm at half maximum. In order to keep the deuterons together while they are propagating the 2-3 m into the center of the core, electrostatic planar electrodes are placed in the beam tube, creating effective

**Fig. 13.** View of GENEPI. The HV terminal (1), the duoplasmatron (2), the accelerating tube (3), three electrostatic quadropoles (4,6,7), the dipole (5), the beam line (8) inside the reactor (9) towards the tritium target (10).

<table>
<thead>
<tr>
<th>TABLE VI</th>
</tr>
</thead>
<tbody>
<tr>
<td>GENEPI Characteristics of the (d,t)-mode</td>
</tr>
<tr>
<td>Peak current</td>
</tr>
<tr>
<td>Mean current</td>
</tr>
<tr>
<td>Pulse length</td>
</tr>
<tr>
<td>Deuteron energy</td>
</tr>
<tr>
<td>Frequency</td>
</tr>
<tr>
<td>Beam diameter</td>
</tr>
<tr>
<td>Target</td>
</tr>
<tr>
<td>Activity of target</td>
</tr>
<tr>
<td>Neutron energy (mean)</td>
</tr>
<tr>
<td>Neutron production (peak)</td>
</tr>
<tr>
<td>Neutron production (mean)</td>
</tr>
</tbody>
</table>
focusing. The target consists of a combination of titanium and tritium/deuterium. A thin layer of the combination is placed on a 1.5 mm thick cylindrical (\(\phi=30\) mm) plate of natural copper. One of the major advantages of the introduction of GENEPI is the possibility of making accurate dynamic measurements, which enables experimental determination of the reactivity of the sub-critical multiplying medium.

### 3.1.4 The GENEPI-generated External Neutron Sources

The reaction induced by the GENEPI generator (in the (d,t)-mode) is a deuterium-tritium nuclear reaction, creating a neutron and an alpha particle. The energy released is 17.6 MeV, of which the neutron obtains on average 14.1 MeV. The Coulomb barrier for this reaction is about 400 keV and the deuterons thus have to tunnel through this barrier. The probability for this kind of reaction to occur is a combination of the penetration probability and the cross-section of the (d,t)-interaction, and in this case it is maximal at about 100 keV. The energy of the deuterons in GENEPI is 240 keV, which allows for some slowing down inside the target. If the incoming deuteron energy is low, the angular distribution of neutrons is essentially isotropic with neutron energy of 14.1 MeV. However, with increasing deuteron energy, the neutron density and the neutron energy are higher in the forward direction than in the backward direction.

In the MUSE-4 studies presented in this thesis, the (d,d)- and the (d,t)-reactions themselves have not been simulated with MCNP. Instead, the double-differential cross-section and the energy dependence of the neutron emission angle are taken from [56]. These distributions are plotted in Fig. 14 and Fig. 15, and are listed in the figure:

![Figure 14](attachment:fig14.png)

**Fig. 14.** Double-differential cross-section and energy dependence as a function of neutron emission angle for the (d,d)-reaction.
Appendix B. The energy of the emitted neutrons in the laboratory system ranges from about 2.0 to 3.1 MeV for the (d,d)-neutrons and from about 13.1 to 15.2 MeV for the (d,t)-neutrons, with a maximum emission probability density peaking in the forward direction (0 degrees). For the (d,t)-reaction, the double-differential cross-section has similar shape as the energy dependence, with the maximum at 0 degrees and minimum at 180 degrees. This is expected according to basic kinematics theory. For the (d,d)-reaction however, the double-differential cross-section has a minimum at 90 degrees. The reason for this behavior is a quantum effect from the interference of the incoming and outgoing wave functions of the particles involved in the reaction.

3.2 The Simulation Tools Used in the Numerical Studies

The major part of the numerical simulations included in this thesis was performed with the Monte Carlo codes MCNP (version 4C3) and MCNPX (version 2.2.3), both using the data file ENDF/B-VI. The Intranuclear Cascade Model applied by MCNPX is the Bertini package. For comparison, the deterministic fast reactor code ERANOS [57] was used as modeling tool in some of the studies [Paper II].

3.2.1 The Monte Carlo Method

The Monte Carlo method is a stochastic process, which allows for simulating particle transport in any arbitrary geometry. While deterministic codes solve the Boltzmann transport equation for an average particle, Monte Carlo obtains a
solution by simulating individual particles and then inferring their average behavior. It is particularly useful for complex problems that cannot be modeled by deterministic codes. The problem description is often relatively short and easily constructed. The method consists of following each particle, created somewhere in the geometry, throughout its life (from birth to death, e.g. via absorption or escape) and its interactions with other particles (fission, capture, scattering etc.). The process is run for a large number of source particles to obtain a statistically reliable result and the program records the average behavior of the simulated particles.

To simulate the particle interaction with matter using a Monte Carlo method such as MCNP, nuclear data libraries (e.g. JEF, JENDL, ENDF/B) containing cross-section information for all relevant isotopes and processes are used. This data is then processed into a format appropriate for MCNP (so-called ACER format) by the code, NJOY [58], which produces new data files and a “directory” file, XSDIR, which is directly used by MCNP. A flow sheet describing this nuclear data treatment procedure is shown in Fig. 16.

![Fig. 16. Flow sheet of nuclear data treatment, finally applied by a simulation tool such as MCNP.](image)

The basic advantage of Monte Carlo codes over deterministic codes is that they require no averaging approximations in space, energy and time. One disadvantage of Monte Carlo, however, is that the solution contains statistical errors. All results in Monte Carlo represent estimates with associated uncertainties and the calculations can be rather time consuming as the required precision of the results increases.

### 3.2.2 MCNP and MCNPX

In MCNP, the allowed particles are neutrons, electrons and photons, with the neutron energy range being limited to 20 MeV, which is sufficient for classical reactor simulations and most neutron transport problems. However, this limitation is a problem when simulating an accelerator-driven system involving high-energy interactions such as proton-induced spallation.

MCNPX on the other hand, is an extended version of MCNP in which the major capabilities of the high-energy transport code, LAHET and MCNP have been merged together. In MCNP, particle transport relies entirely on nuclear data contained in externally supplied cross-section tables ($E_n < 20$ MeV), which are derived from evaluated nuclear data files. In LAHET, on the other hand, particle
transport is accomplished by using various theoretical physical models embedded in the code, covering the energy range up to several GeV. In MCNPX, the table-based data are used whenever they exist, as such data are known to yield the best results. When they do not exist, the code built-in physical models are used. LAHET and MCNPX also have the possibility to transport many particles other than neutrons, the most important in the context of ADS simulations being protons. The nuclear data libraries (e.g. JEF, JENDL, ENDF/B) have traditionally been limited to 20 MeV. However, progress is underway with the development of cross-section tables (e.g. LA150 [59]) covering the important energy range from 20 up to 150 or 200 MeV.

3.2.3 ERANOS

ERANOS is a deterministic fast reactor code system [57] using cross-section libraries based on the JEF2.2 evaluated file. ERANOS is well validated for classical sodium-cooled fast reactors. This validation has been recently extended to Pu-burning cores with steel-sodium reflectors and high Pu-content. However, the code is not yet fully validated for systems characterized by large sub-criticalities and the presence of high-energy neutrons from spallation.

In ERANOS, 1-D cell or 2-D sub-assembly calculations are performed with the ECCO code, while core calculations can be performed with different 2-D or 3-D, diffusion or transport theory modules. In the study presented in [Paper II], the two-dimensional $S_n$ transport code BISTRO [60] was used. Two sets of nuclear data libraries are available with ERANOS, both of them based on the JEF-2.2 evaluated file. The first one is directly derived from JEF-2.2, whereas the other one, called ERALIB1 [61], has been generated in a nuclear data statistical adjustment procedure. The energy range in ERANOS is limited to 20 MeV and it thus needs to be combined with a high-energy code in order to be able to simulate high-energy spallation-driven systems.

3.3 Neutron Energy Spectra

One of the objectives of the studies presented in this section was to compare the effects from the GENEPI-generated (d,d)- and (d,t)-neutron sources with the effects from a spallation source, representative of a future ADS. All of the sources were coupled to the same sub-critical MUSE-4 core. MCNP was used for all calculations with the (d,d)- and the (d,t)-sources, while MCNPX was used to simulate the configurations involving the 1000 MeV proton-induced spallation source.

3.3.1 Neutron Leakage Spectra from the Lead Buffer

One way to investigate the neutron source effects is to first study the sources without the multiplicative medium present and to compare the different neutron leakage spectra. For this purpose, the surrounding fuel and shielding were temporarily
removed, as shown in Fig. 17. For the simulation of the spallation source, the lead buffer/target in the model was extended by one extra sub-assembly towards the proton beam, replacing part of the accelerator tube. Using MCNPX, 1000 MeV protons were directed towards the lead target, generating the spallation source.

The energy spectra of the neutrons exiting the lead buffer are plotted in Fig. 18 and it is seen that the spectrum from the (d,d)-source has a large peak between 2 and 3 MeV, which is the energy range with which the neutrons are emitted by the

**Fig. 17.** Configuration of only the lead buffer region. To the left: (d,d)- or (d,t)-source neutrons emitted at the center of the core. To the right: 1000 MeV protons accelerated towards the extended lead buffer creating a large number of spallation source neutrons.

**Fig. 18.** Neutron leakage spectrum from the lead buffer/target for a (d,d)-source, a (d,t)-source and a spallation source (1000 MeV protons).
GENEPI generator. Hence, only a small fraction of the source neutrons have lost their initial energy. This is an expected result, since the energy loss by elastic scattering of neutrons in lead is quite small. However, for the (d,t)-source, the energy of the neutron spectrum has decreased significantly, which is explained by the (n,2n)-reactions in the lead buffer, induced by the 14 MeV neutrons. The (n,2n)-reaction in lead has a threshold at about 7 MeV (Fig. 19), explaining why there is no such effect for the (d,d)-source. However, about 35 % of the neutrons exiting the lead buffer have not interacted with the lead and are still in the 14-MeV peak. For the spallation source, most of the source neutrons have rather low energy compared to the initial proton energy, with the maximum density at a little less than 2 MeV. This is a typical neutron leakage spectrum for 1000 MeV protons impinging on a lead target of this size. About 7 % of the spallation neutrons, however, still have energy higher than 20 MeV.

![Fig. 19. Neutron microscopic cross-sections for $^{239}$Pu fission, $^{238}$U fission and (n,2n)-reactions in $^{206}$Pb, $^{207}$Pb and $^{208}$Pb (ENDF/B-VI).](image)

The GENEPI-generated neutron sources in the MUSE experiments were surrounded by a lead buffer with the purpose to simulate the neutron diffusion of an actual lead (or lead-bismuth) target in an ADS. The comparison of the neutron leakage spectra shows that the neutrons from the (d,t)-source in MUSE-4 have a rather similar energy spectrum as the neutrons from the spallation source have, and can from this point of view be considered more representative for a spallation-driven system than the (d,d)-source.

### 3.3.2 Neutron Spectra in the Core

The neutron energy spectra for the three different sources have been computed with the entire core present (including fuel, reflector and shields), according to Fig. 20. As in the case of the neutron leakage calculations in the previous section, the
target/buffer was extended by one extra sub-assembly for the calculations with the spallation source (Fig. 17). The spectra were calculated in two different positions; one in the lead buffer and one in the fuel (marked with black dots in Fig. 20). The calculations were performed for the second sub-critical state of MUSE-4 ($k_{eff} \sim 0.97$). The spectra for the other sub-critical states ($SC0, k_{eff} \sim 0.994$ and $SC3, k_{eff} \sim 0.95$) are not shown here, since they are rather similar to the spectra of SC2. However, as there is less fission multiplication for larger sub-criticalities, the origin of the sources becomes a little more pronounced in SC3 and vice versa in SC0. Some of the results presented in this section have also been published in [Paper I].

![Fig. 20](image.png)

**Fig. 20.** The two positions (marked with the black dots) where the neutron energy spectra have been calculated.

In Fig. 21, the neutron energy spectra for the three different sources calculated in the lead buffer are plotted. The spectra are very similar to each other, the energy density being maximal at about 500 keV. Several spectrum characteristics of the multiplying fuel can be recognized, for example the two dips in the neutron fluxes caused by the scattering resonances in sodium ($\sim 3$ keV) and oxygen ($\sim 0.4$ MeV). This indicates that the neutron spectrum in this position is rather dominated by the fission multiplication in the fuel and that many of the neutrons from the fuel enter into the lead buffer. However, a smaller fraction of the neutrons have energies different from the average behavior and the two peaks representing the origins of the GENEPI-generated neutron sources and the high-energy tail of the spallation source are very clear in this position. It should be noted that the position in the lead buffer where the energy spectra have been calculated is only about 5 cm from the position where the GENEPI source neutrons are emitted.
In Fig. 22, the neutron spectra in the fuel at a point located 21 cm from the center of the core and about 10 cm into the fuel are depicted. Naturally, the fuel spectrum characteristics in this case are even more pronounced than in the lead buffer. The three different curves are very similar and almost no traces from the origins of the external neutron sources can be observed. Only about 0.15 % of the (d,t)-source neutrons are still in the 14 MeV peak and about 0.04 % of the neutrons in the spallation-driven system have energies higher than 20 MeV.
The computed neutron spectra show that the energy spectra in the core, originating from the three different external sources are very similar to each other. These results indicate the validity of one of the basic hypotheses of the experiments, namely that the choice of using a spallation source or the source neutrons produced by the (d,d)- or the (d,t)-reactions, will affect very little the neutron spectrum in the fuel. Only inside the lead buffer and at the buffer/core interface some differences are observed. We therefore conclude that, for the purpose of computing neutron spectrum-weighted quantities, the presence of the external source can be neglected beyond a few centimeters into the fuel, whatever the neutron source energy distribution.

3.3.3 Distribution of the Spallation Neutrons

The distribution of the neutrons produced in the spallation target was calculated for 1000 MeV protons impinging on the extended MUSE-4 target. The properties, in terms of energy and space, of the neutrons emerging from the proton-induced spallation reactions were recorded. These emitted neutrons have been called “primary neutrons”, as they include only the first neutron in each neutron chain reaction. No neutrons generated in reactions induced by other neutrons (secondary neutrons) are included. The definition of primary neutrons is further treated in [Paper II] and in Section 3.4.

The spatial distribution of the positions where the primary neutrons were created was found to be rather limited. Axially, most of the neutrons were created in the upper part of the lead target (77% within the first 20 cm), as shown in Fig. 23. It is also seen that the neutron density, which in addition to the primary neutrons include

![Fig. 23. To the left: 1000 MeV protons impinging on the extended MUSE-4 lead buffer/target. In the middle: Axial distribution of the primary spallation neutrons. To the right: Radial distribution of the primary spallation neutrons.](image)
also secondary neutrons, has a different shape with its maximum about 10 cm below the top of the target. The radial distribution was found to be very peaked around the axis of the incident proton beam, about 98% of the neutrons were created within a 3 cm radius. In this case, the radius of the uniformly distributed proton beam was 2 cm. The energy distribution of the primary neutrons produced by the 1000 MeV protons is displayed in Fig. 24. We note that 16.8% of the neutrons have energies higher than 20 MeV and 3.3% of them higher than 150 MeV. The neutrons with very high energy were mainly emitted in the forward direction of the proton beam.

![Fig. 24. Energy spectrum of the primary spallation neutrons created by the 1000 MeV protons.](image)

On average, there were about 14.5 primary neutrons produced in each proton-induced spallation reaction. This value may be compared with the total number of neutrons produced in the lead target, i.e. about 21 neutrons per incident proton. Thus, almost a third of the neutrons that exit the lead target and enter into the fuel are secondary neutrons, most of them created in neutron-induced spallation reactions and (n,xn)-reactions. In a large cylindrical target \( r \geq 30 \text{ cm} \), this fraction is about 50%, the total number of neutrons created per proton being as large as 30, while the number of primary spallation neutrons are the same, independently of the target size.

### 3.4 Neutron Source Efficiency

The neutron source efficiency \( \phi^* \) was determined for the GENEPI-generated neutron sources and for the spallation source, all of them coupled to the second sub-critical state (SC2) of the MUSE-4 experiments (Fig. 12).

#### 3.4.1 Definition of \( \phi^* \)

The neutron flux distribution \( \phi \), in a sub-critical core is the solution to the inhomogeneous steady-state neutron transport equation.
\[ A\phi_s = F\phi_s + S, \] (3)

where

\[ A\phi_s = \Omega \cdot \nabla \phi_s + \sigma \phi_s - \sum_{x \neq f} \int\! \int \sigma'_{x'} f_{x'} \phi'_{x'} d\Omega' dE' \] (4)

\[ F\phi_s = \int\! \int \frac{1}{4\pi} \sigma_f \nu(r; E' \rightarrow E) \cdot \phi'_s d\Omega' dE' \] (5)

\( F \) is the fission production operator, \( A \) is the net neutron loss operator and \( S \) is the external source. The neutron source efficiency [46,47], usually denoted \( \phi^* \), represents the relative efficiency of the external source neutrons and can be expressed according to the following equation [62];

\[ \phi^* = \left( \frac{1}{k_{\text{eff}}} - 1 \right) \cdot \frac{< F\phi_s >}{< S_n >}, \] (6)

which is valid in the range \( 0 < k_{\text{eff}} < 1.0 \). \(< F\phi_s > \) is the total production of neutrons by fission and \(< S_n > \) is the total production of neutrons by the external source. In the above formula, the brackets imply integration over space, angle and energy. Eq. (6) relates the total fission neutron production \(< F\phi_s >\) to the external neutron source, \( \phi^* \) and the reactivity \((1 - 1/k_{\text{eff}})\). It shows that, for given values of \( k_{\text{eff}} \) and \(< S_n >\), the larger \( \phi^* \) the larger the fission power produced in the system. The quantities on the right hand side of Eq. (6) are standard outputs from MCNP and MCNPX.

### 3.4.2 Estimation of the Statistical Error in \( \phi^* \)

In order to estimate the statistical uncertainty in \( \phi^* \), the formula for “propagation of error” was applied (Eq. (7)), assuming that the errors of \(< F\phi_s >\) and \(< S_n >\) (labeled \( F \) and \( S \) in this sub-section) are \( \Delta F \) and \( \Delta S \).

\[ \Delta f(x_1, x_2, ...) = \sqrt{\left( \frac{\partial f}{\partial x_1} \Delta x_1 \right)^2 + \left( \frac{\partial f}{\partial x_2} \Delta x_2 \right)^2 + \ldots + \rho_{12} \cdot \frac{\partial f}{\partial x_1} \Delta x_1 \cdot \frac{\partial f}{\partial x_2} \Delta x_2 + \ldots} \] (7)

The correlation constant \( \rho \) could be either positive or negative; negative if \( k_{\text{eff}} \) and \( F \) are correlated and positive if they are anti-correlated. However, as a first
approximation, $\rho$ was assumed to be zero. With the derivatives $\frac{\partial \phi^*}{\partial k_{\text{eff}}}$, $\frac{\partial \phi^*}{\partial F}$ and $\frac{\partial \phi^*}{\partial S}$ inserted in Eq. (7), an expression for the relative error in $\phi^*$ can be obtained,

$$
\left( \frac{\Delta \phi^*}{\phi^*} \right)^2 \approx \left( \frac{1}{1-k_{\text{eff}}} \cdot \frac{\Delta k_{\text{eff}}}{k_{\text{eff}}} \right)^2 + \left( \frac{\Delta F}{F} \right)^2 + \left( \frac{\Delta S}{S} \right)^2.
$$

Eq. (8) was used in all simulations to estimate the statistical uncertainty in $\phi^*$.

### 3.4.3 Definition of the External Neutron Source in Spallation-driven Systems

Since the actual source particles in a system coupled to a proton-induced spallation source are protons and not neutrons, it is not obvious which is the best way to define the external neutron source. The procedure to calculate $\phi^*$ is usually divided into two steps, the first generating the source neutrons produced from the proton beam interacting with the target and the second step determining the efficiency of these source neutrons. Different source definitions are possible and they will result in different values and meanings of $\phi^*$. A brief summary of four different definitions of the external neutron source that have been used in the ADS field is given in [63]. Among these, the two most frequently used definitions, the target neutron leakage source and the energy cut-off source, as well as the primary neutron source, are described in the following. The fourth definition is the fission source, consisting of the first generation of fission neutrons.

#### The Target Neutron Leakage Source

The approach of the target neutron leakage source uses the neutrons that leak out radially from the target as source neutrons [64,65]. The method consists of, in the first step, transporting the high-energy protons and the secondary particles that they produce in the target. The neutrons that leak out from the target are defined as the source and their properties, in terms of position, direction and energy are written to a source file. Only the target is present in the first simulation, so no once-leaked neutrons re-entering the target are included in the source definition. In the second step, the leakage neutrons are reemitted as fixed source neutrons in a separate run and the efficiency ($\phi^*$) of them is determined. Since the target neutron leakage spectrum includes a high-energy tail, both step 1 and 2 need to be simulated with a high-energy transport code (which can simulate neutrons with energy up to the incident proton energy).
The Energy Cut-off Neutron Source

The other way to define the neutron source is to collect the neutrons that fall below a certain cut-off energy (usually 20 or 150 MeV) [66,67,68]. In the first step, a high-energy code is used to transport the accelerated protons and the secondary high-energy particles. The neutrons that are produced are either killed if they are born below the cut-off energy or transported until they fall below this energy. The properties of the killed neutrons are written to a source file, which is followed by their reemission in the second-step run and the calculation of $\phi^*$. An advantage of this approach is that the second step can be simulated with a low-energy transport code. The cut-off energy is set to the upper energy limit of the cross-section library that will be used in the second-step calculation. This is desirable since many reactor code systems are limited to the energy range covered by the cross-section data library (generally 20 or 150 MeV).

The Primary Neutron Source

The source neutrons produced from spallation are here defined as the neutrons produced directly in a spallation reaction induced by a proton (or by any other particle, except neutrons) [Paper II]. They are called “primary neutrons”, in contrast to “secondary” neutrons, which are the neutrons created by the primary neutrons (e.g. via neutron-induced spallation or (n,xn)-reactions). In this approach, the starting point for the chain of neutron multiplication is the moment when a neutron is born in a non-neutron-induced spallation reaction. The primary neutron might be multiplied by secondary spallation (if its energy is very high) and/or (n,xn)-reactions in the lead target, followed by fission reactions in the fuel, the end point of the neutron multiplication chain being the absorption or escape of the last neutron originating from the initial primary source neutron.

3.4.4 Calculations of $\phi^*$ for the MUSE-4 Model

The multiplication factor $k_{\text{eff}}$ and the total number of neutrons produced by fission ($<F\phi_s>$) were calculated for the three different sources. Knowing these two parameters $\phi^*$ can be determined according to Eq. (6). $<F\phi_s>$ is automatically normalized per source neutron in MCNP (and in MCNPX if the source particles are neutrons), so $<S_n>$ was always equal to 1 in the calculations. The corresponding statistical errors ($\pm$1 standard deviation) were calculated using Eq. (8). The results, including error estimates, are listed in Table VII.

For the calculations of the spallation source, the primary neutron source definition was used and the MCNPX simulations calculating $\phi^*$ were divided into two steps. In the first simulation, a large number of protons were accelerated (to 1000 MeV) towards the lead target. The protons were uniformly distributed across the beam of radius 2 cm. The angular, energy and spatial distributions of all neutrons that were created directly in the spallation interactions (primary spallation neutrons)
were recorded. After that, the neutron trajectories were immediately terminated. This procedure produces a spectrum of primary spallation neutrons, i.e. no secondary neutrons are included. In the second step, these primary spallation neutrons were supplied to the MCNPX code as fixed source neutrons for separate simulations and $\varphi^*$ was determined.

### TABLE VII

Source Efficiency for the MUSE-4 SC2 Configuration

<table>
<thead>
<tr>
<th>Source</th>
<th>$k_{\text{eff}}$</th>
<th>$&lt; F_{\phi_i} &gt;$ (D,D)-Source</th>
<th>$\varphi^*$ (D,D)-Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>(D,D)-Source</td>
<td>0.97285 (± 0.4 %)</td>
<td>48.8 (± 0.4 %)</td>
<td>1.36 (± 0.010)</td>
</tr>
<tr>
<td>(D,T)-Source</td>
<td>(± 18 pcm)</td>
<td>77.6 (± 0.5 %)</td>
<td>2.17 (± 0.020)</td>
</tr>
<tr>
<td>Spallation Source</td>
<td>80.6 (± 1.0 %)</td>
<td>2.25 (± 0.030)</td>
<td></td>
</tr>
</tbody>
</table>

The energy of the emitted (d,d)-source neutrons (2-3 MeV) is only slightly larger than the average energy of a neutron produced by fission. Since $\varphi^*$ is 1.0 for an average fission neutron, the value for the (d,d)-source is therefore expected to be equal or slightly larger than 1, which is indeed the case. For the (d,t)-source, the reason for the higher values of $\varphi^*$ is the larger fission rate, part of which coming from fissions induced by the neutrons multiplied by (n,2n)-reactions in the lead buffer. It is seen in Table VII that the number of fission neutrons per source neutron is large, approximately 59 % larger than for the (d,d)-source. The (n,2n)-multiplication of the 14 MeV neutrons increases the number of neutrons leaking out into the fuel and inducing fission chain reactions, thus enhancing the neutron source efficiency. In average, about 1.5 neutrons leave the lead buffer per initial 14 MeV neutron, compared to 1 neutron for each 2.7 MeV (d,d)-source neutron.

Concerning the spallation source neutrons, the values of $\varphi^*$ obtained in the simulations are somewhat higher than for the (d,t)-source. This is due to the fraction of primary neutrons having very high energy (Fig. 24). Most of the neutrons from the spallation process are born with an energy lower than the (n,2n)-cross-section threshold in lead (i.e. no (n,2n)-multiplication occurs), but the neutrons with very high energy contribute significantly to $\varphi^*$. Additional calculations investigating the contribution to $\varphi^*$ by the high-energy fraction of the spallation source show that the primary source neutrons with energies higher than 20 MeV (16.8 % of all source neutrons) contribute for about 50% to the total $\varphi^*$ [Paper II]. The explanation for this is that most of the high-energy neutrons from the spallation source have already been multiplied in the lead (most of them via secondary neutron spallation and (n,xn)-reactions) before they enter into the fuel. Each of them gives birth to a number of lower-energy neutrons, which then leak out of the lead and induce fission chain reactions in the fuel.
3.5 Dynamic Neutron Source Response

Techniques to monitor the sub-criticality level during operation in ADS are under development within the MUSE-4 program. With the introduction of GENEPI into MASURCA, it is now possible to perform accurate dynamic measurements, allowing for experimental reactivity determination of the sub-critical multiplying medium. The dynamic measurements are based on the pulsed mode operation of GENEPI.

In the experiments, after the reactivity calibration by the rod drop technique, the sub-critical level of the different configurations will first be precisely determined by the well-known static Modified Source Method (MSM). In the next step, based on reactor kinetics and neutron noise theory, different dynamic techniques are applied, in order to determine the reactivity level. The methods being studied are the pulsed neutron source method, the inverse kinetics method, the Rossi-$\alpha$ method and the Feynman-$\alpha$ method. These methods, as well as results and interpretations from the dynamic experiments have been presented in several documents [5,53,69].

3.5.1 The Pulsed Neutron Source Method

When a multiplying medium is sub-critical, the neutron density will decay exponentially and the pulse will quickly disappear, according to basic point-kinetic theory. The decay process after a neutron pulse in a sub-critical medium is governed by the following relation [70];

$$N(t) = N_0 e^{-\alpha t}, \quad (9)$$

where $\alpha = \frac{1 - k_{\text{prompt}}}{\Lambda}$ and $\Lambda$ = neutron generation time. Hence, $\alpha$ is the slope of the decaying curve and it is directly related to the reactivity. Eq. (9) is derived from the reactor kinetics equation, not taking the delayed neutrons into account. Within this time scale there is no contribution from delayed neutrons, which is the reason why $\alpha$ is directly proportional to $k_{\text{prompt}}$ and not to $k_{\text{eff}}$.

The “Pulsed Neutron Source Method” [70] is one of the most promising techniques for the experimental reactivity determination of the sub-critical core in a future ADS. The procedure consists of, after having registered the neutron intensity decay after a neutron pulse, determining the slope of the curve ($\alpha$) and then determine $k_{\text{prompt}}$, according to

$$k_{\text{prompt}} = 1 - \alpha \cdot \Lambda. \quad (10)$$

The neutron generation time, $\Lambda$ first has to be calculated and does not change much with reactivity. In order to obtain the effective reactivity, you also need to determine the effective delayed neutron fraction ($k_{\text{eff}} = k_{\text{prompt}} + \beta_{\text{eff}}$).
The time response in one of the detectors in MASURCA has been simulated for a GENEPI-generated (d,t)-neutron source pulse for the three sub-critical states, SC0, SC2 and SC3 of the MUSE-4 experiments. The generation time $\Lambda$ was approximately 0.60 microseconds. The results computed by MCNP are plotted in Fig. 25. It is clear from the figure that the more sub-critical the core is, the faster is the prompt decay rate. The $\alpha$-values corresponding to the sub-critical states (determined graphically from the plotted curves) are for SC0; $\alpha_0 \approx 1 \cdot 10^4$ ($k_{\text{prompt}} \approx 0.995$), for SC2; $\alpha_2 \approx 5 \cdot 10^4$ ($k_{\text{prompt}} \approx 0.97$) and for SC3; $\alpha_3 \approx 8 \cdot 10^4$ ($k_{\text{prompt}} \approx 0.95$). The results are in good agreement with Eq. (9).

![Fig. 25. Neutron source response after a (d,t)-source pulse for the three different sub-critical configurations in MUSE-4, SC0, SC2 and SC3.](image)

### 3.6 Replacement of Sodium Coolant by Lead Coolant

It was decided within the MUSE community to include, in an extended phase of the MUSE-4 experiments, a new configuration, in which 22 of the central sodium-cooled fuel sub-assemblies were replaced by lead-cooled sub-assemblies (the limit of the lead-cooled region being marked by the black line in Fig. 26). Three parameters; $k_{\text{eff}}$, the source efficiency $\varphi^*$ and the dynamic neutron source response were studied for this centrally lead-cooled configuration. The neutron source used in the study comparing this configuration with the original sodium-cooled configuration was the GENEPI-generated (d,t)-source.

When calculating $k_{\text{eff}}$ for the new lead-cooled configuration, it was found that it increases with about 1600 pcm. The reason for this is that there is less absorption in lead and that the energy spectrum is different. In order to maintain the same
reactivity for the two configurations, which is particularly important for the comparison of the neutron source response, some of the peripheral fuel sub-assemblies were removed. $k_{\text{eff}}$ was approximately 0.974 in both cases.

![Image](fig26.png)

**Fig. 26.** Configuration where 22 of the sodium-cooled sub-assemblies were replaced by lead-cooled sub-assemblies (the limit of the lead-cooled sub-assemblies is marked by the black line).

### 3.6.1 Source Efficiency

The results from the comparison of $\phi^*$, for the two configurations, are listed in Table VIII. As is shown, the value for the lead-cooled configuration is significantly higher than for the sodium-cooled configuration, 2.39 compared to 2.13. The reason for this difference is again the (n,2n)-multiplicative effect in lead, the same effect that caused the large difference in $\phi^*$ between the (d,d)- and the (d,t)-source. Since there is more lead in the central part of the core in the lead-cooled configuration, where there are still many neutrons with energy higher than about 7 MeV, there is more (n,2n)-reactions. These circumstances enhance the neutron multiplication as

<table>
<thead>
<tr>
<th>Configuration</th>
<th>$k_{\text{eff}}$</th>
<th>$\phi^*$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sodium-cooled</td>
<td>0.97428 (± 20 pcm)</td>
<td>2.13 (± 0.02)</td>
</tr>
<tr>
<td>Centrally lead-cooled</td>
<td>0.97382 (± 27 pcm)</td>
<td>2.39 (± 0.03)</td>
</tr>
</tbody>
</table>

TABLE VIII

Source Efficiency $\phi^*$ for the Sodium-cooled and the Centrally Lead-cooled MUSE-4 Configurations
well as $\phi^*$. Since the source efficiency relates the source intensity to the power produced in the system, we conclude that the replacement of sodium coolant by lead coolant in the 22 central fuel sub-assemblies increases the power by approximately 12%.

### 3.6.2 Dynamic Neutron Source Response

Finally, the neutron source response from a pulse insertion from the (d,t)-source, registered by a detector in the core, was calculated and it is seen in Fig. 27 that the intensity is somewhat higher (on average ~15%) for the lead-cooled configuration. The explanation for this is the same as for the difference in source efficiency, namely the multiplicative effect of the (n,2n)-reactions in the lead. Since the neutron multiplication is higher in the second case, the intensity registered by the detector will also be higher. We also observe that the prompt decay rate is more or less the same for the two systems. The neutron generation time $\Lambda$, calculated by MCNP, changes very little and since $k_{\text{eff}}$ is the same, the $\alpha$-value and the decay rate must also be the same.

![Fig. 27. Neutron source response in one of the detectors from a neutron source pulse.](image)
Chapter 4

Proton Source Efficiency in ADS

An important factor when designing an ADS is to optimize the beam power amplification, i.e. the core power over the accelerator power, given that the reactor is operating at a certain sub-critical reactivity level. Optimizing the source efficiency and thereby minimizing the proton beam requirements can have an important impact on the overall design of an ADS and on the economy of its operation. The neutron source efficiency parameter $\phi^*$ is commonly used to study this quantity, since it is related to the number of fissions produced in the core (which is proportional to the total core power) by an average external source neutron.

However, calculating $\phi^*$ for an ADS introduces some complications. Since the neutron source is generated by a proton beam/target simulation, the distribution of the source neutrons is dependent on the target properties and the proton beam properties. In order to determine $\phi^*$, the external neutron source first has to be defined and then the efficiency of this neutron source can be determined. The major drawback with using $\phi^*$ is that the neutron source can be defined in several different ways (Section 3.4), and the results are directly dependent on the choice of definition. Therefore, completely different values of $\phi^*$ are often observed [64,66, Paper II], due to different choices of external neutron source definition. Another complication associated with the neutron source efficiency is that, studying $\phi^*$ as a function of a certain system parameter might change the neutron source distribution and the number of neutrons produced per source proton ($Z$). If a change in the studied system parameter changes the distribution of the neutron source, $\phi^*$ has to be weighted by $Z$. With the target neutron leakage definition, examples of parameters affecting the source distribution are the target dimension, the proton beam energy and the axial proton beam impact position. Other system parameters, such as the core coolant material, the fuel composition and the core dimensions are independent of the target region and do not affect the neutron source. In contrast to the target neutron leakage source, it has been shown that using the energy cut-off definition, the neutron source distribution is rather insensitive to changes in the target radius.
[66]. However, substituting the coolant material or changing the fuel composition in this case might affect the distribution of the neutron source.

### 4.1 Introduction of the Proton Source Efficiency

With the purpose of providing a simple means for studying the core power over the beam power, we have introduced a new parameter, called “proton source efficiency” and denoted $\psi^*$. $\psi^*$ refers to the number of fission neutrons produced in the system by each source proton [Paper III]. The advantages with using the proton source efficiency instead of the neutron source efficiency is that there is no ambiguity in how to define the external source and that it is proportional to the beam power amplification, without the need of a weighting factor ($Z$).

#### 4.1.1 Definition of $\psi^*$

$\psi^*$ represents the product of $\varphi^*$ and the number of source neutrons generated per source proton ($Z$). We thus have the following relation between the proton source efficiency $\psi^*$ and the neutron source efficiency $\varphi^*$;

$$\psi^* = \varphi^* \cdot Z,$$

where $Z = \langle S_n \rangle / \langle S_p \rangle$, $\langle S_n \rangle$ and $\langle S_p \rangle$ being the total number of emitted source neutrons and source protons, respectively. $\psi^*$ could also, in analogy with $\varphi^*$, be expressed in terms of $k_{eff}$ and the total number of neutrons produced by fission in the core by each source proton. Inserting Eq. (6) in Eq. (11) it is expressed in the same way as $\varphi^*$, only with the replacement of $\langle S_n \rangle$ by $\langle S_p \rangle$,

$$\psi^* = \frac{1}{k_{eff} - 1} \cdot \frac{\langle F \phi_x \rangle}{\langle S_p \rangle}.$$

$\langle F \phi_x \rangle / \langle S_p \rangle$ is the total production of neutrons by fission over the total number of source protons.

#### 4.1.2 Comparison of $\psi^*$ and $\varphi^*$

Investigating $\psi^*$ and $\varphi^*$ as functions of the target radius and of coolant material illustrates the comparison of system parameters that affect and do not affect the neutron source distribution. Using the target neutron leakage definition as an example and changing the coolant material, $Z$ does not change, so $\psi^*$ and $\varphi^*$ vary in exactly the same way. For a fixed target radius we have, following from Eq. (11),

$$\frac{\psi^*_{Pb-Bi}}{\psi^*_{Na}} = \frac{\varphi^*_{Pb-Bi}}{\varphi^*_{Na}}.$$

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When varying the target radius, on the other hand, $Z$ changes, so $\psi^*$ and $\varphi^*$ varies in different ways. For a fixed coolant material we have

$$\frac{\psi^*_{R1}}{\psi^*_{R2}} \neq \frac{\varphi^*_{R1}}{\varphi^*_{R2}}.$$  \hspace{1cm} (14)

Consequently, if one wants to use the neutron source efficiency parameter $\varphi^*$, it needs to be weighted with $Z$ when studying the target radius, whereas this is not necessary when comparing different coolant materials. However, applying the proton source efficiency and always referring to $\psi^*$, none of this has to be considered.

### 4.1.3 Relationship Between $\psi^*$ and the Core Power

The total power produced by fission in the core ($P_f$) can be expressed as the product of the total number of fission events and the average available energy released in a fission, according to the following relation;

$$P_f = \frac{\langle F \phi_s \rangle}{\nu} \cdot \bar{E}_f .$$ \hspace{1cm} (15)

Inserting Eq. (12) in Eq. (15), we obtain

$$\frac{P_f}{\langle S_p \rangle} = \frac{\bar{E}_f}{\nu} \cdot \frac{k_{eff}}{1-k_{eff}} \cdot \psi^* .$$ \hspace{1cm} (16)

For a given fuel composition, $\bar{E}_f$ and $\bar{\nu}$ can, for the purpose of this study, be considered to be constant. Even though there are high-energy neutrons entering into the fuel (the neutron yield $\nu$ is not constant with respect to neutron energy), the fraction of fissions in the core that are induced by high-energy neutrons is very small. Therefore, a change in the neutron yield for these fission events will have very little impact on $\bar{\nu}$. The variations of $\bar{E}_f$ and $\bar{\nu}$ have been calculated for the series of different studies presented in this report and were found to be much smaller than the statistical errors in the simulations (except in the case where the fuel composition was modified). When the fuel composition changes, $\bar{\nu}$ might also change, which should be kept in mind if different fuel types are compared. If we further make the approximation that the energy produced by fission is proportional to the total core power produced in the core, we find that $\psi^*$ is proportional to the total power divided by the source intensity (output/input power).
4.2 $\psi^*$ as a Function of Different System Parameters

In order to find an optimal system design that minimizes the proton current needs, the proton source efficiency $\psi^*$ was studied as a function of a number of different system parameters, such as the target radius, coolant material, axial proton beam impact position, proton beam energy and fuel composition [Paper III].

4.2.1 System Modeling

A homogenized model representing a nitride-fuelled and lead-bismuth-cooled ADS (maximum 800 MWth) has been studied. The height of the active core in the reference model (Fig. 28) is 100 cm and the outer radius is 70 cm. The inner radius is 20 cm, which is also the boundary of the lead-bismuth target. The accelerator tube has a radius of 15 cm and the axial position of the proton beam impact is 25 cm below the top of the core. The radius of the radially uniform 1000 MeV proton beam is 7.5 cm. Above and below the active zone of the core, plena for accommodation of gas release are included, having lengths of 100 and 50 cm, respectively. The radial reflector is assumed to consist of 90% steel and 10% lead-bismuth. The relative fraction of fuel, cladding and coolant material used in the homogenized model correspond to a pin radius of 2.5 mm and P/D = 1.72. The nitride actinide fuel is in

![Fig. 28. RZ-view of the homogenized reference model. The 1000 MeV protons are guided through the accelerator tube and impinge on the Pb-Bi target. The different regions in the model are pure Pb-Bi (1), the plena (2), the active core (3) and the reflector (4).](image)
solid solution with ZrN. The volume fraction of ZrN was adjusted to 83%, in order to obtain a $k_{eff}$ of about 0.95. The fuel consists of 80% plutonium and 20% americium. The spallation target and the core coolant consist of lead-bismuth eutectic and the fuel pin cladding of 10% chromium and 90% iron.

The Monte Carlo code MCNPX (Version 2.3.0) in coupled neutron and proton mode was used for all simulations, relying on the evaluated nuclear data library ENDF/B-VI.8. The Intranuclear Cascade model used by MCNPX was the Bertini package.

### 4.2.2 $\psi^*$ as a Function of Target Radius

$\psi^*$ was computed for the lead-bismuth model for different target radii, both as the product of $\varphi^*$ and $Z$ (Eq. 11) and directly according to Eq. (12), in order to verify the consistency between the two expressions. $\varphi^*$ and $Z$ were calculated using the target neutron leakage definition (section 3.4.3). The results are listed in Table IX and it is seen that they are in good agreement with each other, the differences being within the statistical uncertainty. Hence, using a two-step simulation procedure, $\psi^*$ can be obtained according to Eq. (11), independently of the choice of neutron source definition. If MCNPX is used, $\psi^*$ can be obtained directly according to Eq. (12). $\psi^*$, $\varphi^*$ and $Z$ are also plotted as functions of target radius later on in this section.

#### TABLE IX

Computation of $\psi^*$ according to Eq. (11)$^A$ and Eq. (12)$^B$

<table>
<thead>
<tr>
<th>Target Radius</th>
<th>$\varphi^*$</th>
<th>$Z$</th>
<th>$\varphi^* \cdot Z^A$</th>
<th>$\psi^*$ $^B$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10$^C$</td>
<td>1.81</td>
<td>21.9</td>
<td>39.8</td>
<td>39.6</td>
</tr>
<tr>
<td>20</td>
<td>1.35</td>
<td>26.8</td>
<td>36.3</td>
<td>35.9</td>
</tr>
<tr>
<td>30</td>
<td>1.13</td>
<td>29.0</td>
<td>32.9</td>
<td>32.4</td>
</tr>
<tr>
<td>40</td>
<td>0.99</td>
<td>30.0</td>
<td>29.6</td>
<td>29.5</td>
</tr>
<tr>
<td>50</td>
<td>0.89</td>
<td>30.2</td>
<td>26.7</td>
<td>27.0</td>
</tr>
</tbody>
</table>

$^C$: For the 10 cm target radius, the radius of the accelerator tube was decreased to 10 cm. The results are also plotted in Fig. 29 and Fig. 30.

Concerning the dependence on the target radius, it is seen that $\psi^*$ decreases considerably when the radius increases. There are mainly two reasons for this behavior, one of them being the softening of the radial neutron leakage spectrum from the target, as the target is enlarged. The probability to induce fission for the neutrons entering the active part of the core strongly decreases with decreasing energy, in particular when the core is loaded with even-neutron number actinides. The other reason for the decrease in $\psi^*$ is that the axial target neutron leakage increases with increasing target radius. The major part of the axial leakage is in the backward direction, through the accelerator tube.
On the other hand, increasing the target radius increases the neutron multiplication inside the lead target, which leads to a higher number of neutrons created per source proton ($Z$ increases from 26.8 for $r=20$ cm to 30.2 for $r=50$ cm). The multiplicative effect of ($n,xn$)-reactions and secondary spallation in the lead target enhances the proton source efficiency. Consequently, with increasing target radius, there are more neutrons for each source proton entering into the fuel, although the efficiency of these neutrons is strongly reduced. Accordingly, it is shown in Table IX that $\psi^*$ decreases less rapidly with increasing target radius than $\varphi^*$ does, due to the increase in $Z$. These two competing factors in $\psi^*$ are thus well represented by Eq. (11).

Replacing the lead-bismuth coolant with sodium, it is shown in Fig. 29 that $\psi^*$ decreases for small target radii, while it remains about the same for large radii. The main reason for this is that there is no neutron multiplication in the sodium coolant, in contrast to lead-bismuth. The differences between the two curves indicate that the contribution from ($n,xn$)-multiplication in the Pb-Bi coolant is significant for target radii smaller than about 30 cm. As long as there is a fraction of leakage neutrons with energy higher than about 7 MeV (the ($n,2n$)-threshold in lead) there will be ($n,xn$)-neutron multiplication in lead. When the target radius is small, this high-energy fraction is rather high (6.1% have energies higher than 7 MeV for $r=20$ cm, compared to 0.8% for $r=50$ cm). When the target radius increases, the fraction of high-energy leakage neutrons decreases and at radii above 40 cm the $\psi^*$ values are essentially the same. The difference between the Pb-Bi and the Na coolant are further illustrated in Fig. 30, where the neutron source efficiencies are depicted together with the number of neutrons per source proton.

![Fig. 29. Proton source efficiency $\psi^*$ versus target radius for a Pb-Bi-cooled and a Na-cooled model.](image)
We conclude that in order to optimize the proton source efficiency and the ratio of the core power over the beam power, a target radius as small as possible should be chosen (increasing the radius from 20 cm to 50 cm decreases $\psi^*$ by about 25%). These results are in good agreement with other similar studies [66]. However, it has also been shown that, reducing the target radius has some undesirable effects, for example higher fluence/burn-up ratio (lower maximum burn-up) and more severe high-energy damages. Moreover, for a large-scale ADS, the target must be sufficiently large to be able to remove the heat from the high-power accelerator beam.

4.2.3 $\psi^*$ as a Function of Some Other System Parameters

The investigations of $\psi^*$ as function of coolant material, axial proton beam impact position, proton beam energy and fuel composition are treated in detail in [Paper III]. It was found that the axial position of the proton beam impact that maximizes $\psi^*$ was located approximately 20 cm above the core center for the reference model (accelerator tube radius = 15 cm), the variations between 0 and about 35 cm being relatively small. However, the dependence of $\psi^*$ on the impact position is sensitive to the accelerator tube radius and for a tube radius of 10 cm the maximum was found at about 13 cm above the core center. Reducing the tube radius from 15 to 10 cm also increases the maximum of $\psi^*$ by 3 %.

Investigating the proton source efficiency divided by the proton energy ($\psi^*/E_p$) as a function of the proton energy showed that a maximal accelerator power amplification is obtained for proton energies of about 1200 to 1400 MeV, but with rather small changes between 1000 and 2000 MeV. Finally, increasing the
americium content in the fuel from 20% to 60% decreases $\psi^*$ considerably, in particular for larger target radii. Due to the sharp decrease in fission cross-section below 1 MeV, americium is more sensitive than plutonium to the softening of the energy spectrum of the neutrons that enter into the fuel.

In order to guarantee the stability of uranium-free fuels at high temperatures, the use of inert matrices is foreseen. Different safety parameters of several possible inert matrix fuels have been studied [18]. In [Paper IV], three inert matrices (ZrN, YN and HfN), dispersed with a plutonium- and americium-mixed nitride fuel have been investigated in terms of $\psi^*$. $\psi^*$ has been studied as a function of target radius and has been compared for a single-zone core and a power flattened double-zone core. It was found that the HfN matrix fuel yields a lower $\psi^*$ than the ZrN and the YN matrix fuels. However, for the americium-based fuel and in particular for small target radii, the difference is relatively small. Due to other favorable properties of HfN it is therefore still an interesting option of inert matrix material, despite the loss in proton source efficiency. It was also shown that $\psi^*$ is lower for a power-flattened double-zone core, compared to a single-zone core. The differences are about 5% for the ZrN matrix fuels, while about 10% for the HfN matrix fuels. Comparing the ZrN matrix with the HfN matrix, assuming a double-zone core, the difference in $\psi^*$ is larger for the plutonium-based fuel (~12%) than for the americium-based fuel (~8%).

From the studies of $\psi^*$ we conclude that, in order to optimally design a cost-efficient ADS with high fuel performances, there is a trade-off arising between several different aspects. Various system parameters (e.g. target radius, axial beam impact position, proton beam energy and choice of coolant medium, fuel composition and inert matrix material) together with other target-core characteristics and different safety limitations have to be weighted against the advantage of optimizing $\psi^*$ and minimizing the proton beam requirements.
Chapter 5

Conclusions

The main work presented in this thesis has focused on Monte Carlo simulations investigating the neutron physics and different source effects in a sub-critical system, driven by an external neutron source. Detailed calculations with MCNP and MCNPX have been performed for a model representative of the on-going MUSE-4 experiments. Neutronic parameters, such as neutron energy spectrum, neutron source efficiency ($\phi^*$) and dynamic neutron source response have been investigated for different sub-critical configurations of MUSE-4. The effects from the (d,d)- and the (d,t)-sources produced by the GENEPI neutron generator have been studied and compared with the effects from a 1000 MeV proton-induced spallation source, coupled to the same sub-critical core.

The computed neutron spectra show that fission multiplication dominates at distances past a few centimeters into the fuel. This implies that, for the purpose of ADS core studies, the presence of the source may be ignored in the calculation of spectrum-weighted quantities, except possibly in the immediate vicinity of the external source. The calculations of $\phi^*$ for the different sources yield a much higher value for the (d,t)-source ($E \sim 14$ MeV) than for the (d,d)-source ($E \sim 2.7$ MeV), which is explained by the (n,2n)-multiplications of the 14 MeV neutrons in lead. The (n,2n)-reaction in lead has a threshold at about 7 MeV, which is the reason why the same effect does not appear for the (d,d)-source. In the case of the spallation source, the possibility of choosing different neutron source definitions and the consequences of this choice, have been emphasized. Using the primary neutron source definition, a value of $\phi^*$ similar to that of the (d,t)-source is obtained. Moreover, in the MUSE experiments accurate dynamic measurements enable the development of a technique for reactivity determination in sub-critical systems. One of the methods used is the Pulsed Neutron Source Method, which in this thesis has been demonstrated by MCNP simulations. The relation between the prompt decay rate after a neutron pulse and the sub-critical reactivity level is verified and the expected behavior that the more sub-critical the core is the faster is the decay is clearly shown.
In order to estimate the ratio between the total power produced in the core and the power supplied by the external source in a sub-critical system, the neutron source efficiency parameter \((\varphi^*)\) is commonly used. For the purpose of studying the proton beam power amplification (core power over beam power) in an ADS, however, we have introduced a new parameter, the proton source efficiency \((\psi^*)\). For different reasons, \(\psi^*\) is better suited for this purpose when the external source consists of protons. \(\psi^*\) is defined in analogy with \(\varphi^*\), but relates the core power to the source protons instead of to the source neutrons. Using \(\psi^*\) instead of \(\varphi^*\) avoids ambiguities connected to the definition of the external neutron source. Another advantage of \(\psi^*\) is that it is proportional to the core power divided by the proton beam power, without the need for a weighting factor \((Z)\). When studying \(\psi^*\) as a function of different system parameters it becomes clear that, choosing a system design that optimizes \(\psi^*\) significantly increases the proton beam amplification and can thus have an important impact on the economy of the operation of an ADS. One of the results from the simulations of a lead-bismuth-cooled core, loaded with plutonium and americium fuel, was that \(\psi^*\) decreases strongly with increasing target radius. Hence, in order to maximize the beam power amplification, and thereby minimizing the proton current needs, a target radius as small as possible should be chosen, without exceeding the limits determined by safety constraints, thermal hydraulics and other target-core characteristics.
Appendix A

Derivation of an Equation for Required Accelerator Current in ADS

The total energy produced by fission in the core \( W \) can be approximately expressed as the product of the total number of fission events and the average available energy released in a fission, according to the following relation;

\[
W = \frac{<F\phi_s>}{\nu} \cdot \bar{E}_f , \tag{A.1}
\]

where \(<F\phi_s>\) is the total production of neutrons by fission. The neutron source efficiency \( \varphi^* \) can be expressed according to [62]

\[
\varphi^* = \left( \frac{1}{k_{\text{eff}}} - 1 \right) \cdot \frac{<F\phi_s>}{<S_n>} , \tag{A.2}
\]

where \(<S_n>\) is the total production of neutrons by the external source. Inserting Eq. (A.2) into Eq. (A.1), yields an expression for the energy produced per source neutron,

\[
\frac{W}{<S_n>} = \frac{\bar{E}_f}{\nu} \cdot \frac{k_{\text{eff}}}{1 - k_{\text{eff}}} \cdot \varphi^* . \tag{A.3}
\]

Now defining the number of source neutrons produced per incident source proton as

\[
Z = \frac{<S_n>}{<S_p>} , \tag{A.4}
\]
where $<S_n>$ is the total number of source protons, we obtain

$$\frac{W}{<S_p>} = \frac{\bar{E}_f}{\nu} \cdot \frac{k_{eff}}{1-k_{eff}} \cdot Z \varphi^* . \tag{A.5}$$

Eq. (A.5) expresses the energy produced per incident source proton. We thus finally obtain the following expression for the proton current $i_p$ required to generate a certain total core power $P_{tot}$ as

$$i_p = \frac{P_{tot}}{\left( W/<S_p> \right)} = \frac{1-k_{eff}}{k_{eff}} \cdot \frac{P_{tot} \cdot \bar{\nu}}{\bar{E}_f \cdot Z \cdot \varphi^*} . \tag{A.6}$$
## Appendix B

### Energy Dependence of the GENEPI-generated Neutron Sources

<table>
<thead>
<tr>
<th>$\theta_{\text{lab}}$ (degrees)</th>
<th>$^2$H(d,n)$^3$He</th>
<th>$^3$H(d,n)$^3$He</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$T_n$ (MeV)</td>
<td>$d\sigma/d\Omega_{\text{lab}}$ (mb/steradian)</td>
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<tr>
<td>0</td>
<td>3.14</td>
<td>7.3</td>
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</table>
References


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INVESTIGATION OF NEUTRON SOURCE EFFECTS IN SUB-CRITICAL MEDIA AND APPLICATION TO A MODEL OF THE MUSE-4 EXPERIMENTS

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ABSTRACT

Monte Carlo simulations have been performed to investigate the neutron source effects in a sub-critical media successively coupled to a (d,d)-source, a (d,t)-source and a spallation source. The investigations have focused on the neutron energy spectra in the fuel and on the source relative efficiency $\phi^*$. The calculations have been performed for three sub-critical configurations, representative of the coming MUSE-4 experiments.

The Monte Carlo codes MCNP and MCNPX have been used to compute $\phi^*$. $\phi^*$ has been found to be low for the (d,d)-source (~1.35 compared to 1.0 for an average fission neutron), while considerably higher for the (d,t)-source (~2.15) and the spallation source (~2.35). The high value of $\phi^*$ for the spallation source has been shown to be due to the fraction of high-energy neutrons (17 % of total source with $E_n > 20$ MeV) born from spallation, which contribute for 50 % to the total number of fission neutrons produced in the core. The variations of $\phi^*$ with neutron importance have also been studied for some spherical configurations with a (d,d)- and a (d,t)-source. For the class of variations considered here, $\phi^*$ was found to remain constant or increase only slightly in the interval $0.70 < k_{eff} < 0.996$.

1 INTRODUCTION

Accelerator Driven Systems (ADS) (Salvatores, 1999) are being investigated as a possible means for reducing the long-term radiotoxicity in the spent fuel from the nuclear industry. In principle, the sub-criticality of ADS allows for dedicated cores with a much higher concentration of minor actinides than what is acceptable in critical reactors. Those dedicated cores could achieve high transmutation rates. Research done on ADS indicates that a waste reduction factor of 50 to 100 is theoretically possible (Delpech et al, 1999).
The basic idea of ADS is to supply a sub-critical reactor core with neutrons generated by an intense external neutron source, usually from spallation reactions in a heavy metal target. This idea is being investigated in the MASURCA experimental facility at CEA Cadarache in the framework of the MUSE experiments (MUItiplication avec Source Externe). Different configurations and several different sub-critical levels are being studied (Salvatores, 1996; Soule, 1997; Lebrat, 1999).

The planned MUSE-4 experiments will not use a spallation source. Instead, a high-intensity pulsed neutron generator GENEPi, constructed by CNRS/ISN/Grenoble, will be used to accelerate a 250 keV deuteron beam towards either a deuterium target \((d,d)\) or a tritium target \((d,t)\) to produce well-characterized neutron sources via fusion reactions. \((d,d)\)-reactions produce neutrons with energies between 2 and 3 MeV, while the \((d,t)\)-reactions produce neutrons between 13 and 15 MeV.

The objective of the present study is to investigate neutron source effects in a MUSE-4-type sub-critical core coupled to a well-known \((d,d)\) or \((d,t)\)-source, and to compare the results with those that would be obtained for a hypothetic spallation source coupled to the same core.

This investigation relies entirely on numerical simulations performed with the MCNP (Briesmeister, 2000) and MCNPX (Waters, 1999) Monte Carlo software packages. The two codes are essentially equivalent for neutron transport below 20 MeV. MCNP is used to simulate the production of the \((d,d)\)- and \((d,t)\)-sources, as well as neutron transport below 20 MeV. MCNPX is used to simulate the production of spallation neutrons and particle transport at all energies.

MCNP and MCNPX Monte Carlo models were set up in which a \((d,d)\)-source, a \((d,t)\)-source and a spallation source were successively coupled to three sub-critical configurations (Sc1, Sc2 and Sc3) representative of the upcoming MUSE-4 experiments.

A description of the MUSE-4 model, the calculation codes and the neutron sources used in this study is given in Section 2. In Section 3, the computed neutron energy spectra in the fuel are compared for the three different sources. In Section 4, we describe investigations of the neutron source efficiency \(\phi^*\). The specific procedure used for calculating \(\phi^*\) with MCNP and MCNPX is described. The differences in the computed values of \(\phi^*\) are analysed, as well as the variations of \(\phi^*\) with neutron importance and reactivity.

2 DESCRIPTION OF THE MUSE-4 MODEL, CALCULATION TOOLS AND NEUTRON SOURCES USED IN THIS STUDY

2.1 The Muse-4 Model

Three homogeneous sub-critical configurations have been studied (Sc1, Sc2 and Sc3 with \(k_{\text{eff}} = 0.99, 0.97\) and 0.95 respectively) representing three configurations planned in the MUSE-4 experiments. The geometry of the Sc2 model is shown in Fig. 1 below. The material compositions of the different regions are listed in Appendix A. The axial (z
direction) dimension of the fuel is 60.96 cm, except in a 21.2 cm wide channel above and below the lead buffer and the accelerator tube (in the y direction), where it was extended by 10.16 cm. The Na/SS reflector (Region 2) ends at \( z = \pm 61.76 \) cm. There is also a 10.16 thick axial shield (Region 3) above and below the Na/SS reflector. The overall dimensions of the whole model, including the reflector and the shields, are 159 \( \times \) 169.6 \( \times \) 143.84 cm.

![Fig. 1 x-y Cross-sectional View of the MUSE-4 Sc2 Sub-Critical Configuration (k-eff = 0.97). The cross shows the position where the neutron spectra have been calculated, see Section 3.](image)

To obtain the two other sub-critical levels, Sc1 and Sc3, fuel cells were added or removed at the core periphery.

2.2 Description of the Calculation Codes

Calculations have been performed with MCNP-4C for models of the three MUSE-4 sub-critical configurations with the \( (d,d) \)- and \( (d,t) \)-sources. MCNPX was used to simulate the system with the spallation source. All simulations relied on the same evaluated nuclear data library, namely ENDF/B-VI.4.

MCNPX is the extended version of MCNP where the major capabilities of LAHET (Prael and Lichtenstein, 1989) and MCNP-4B (Briesmeister, 1997) have been merged together. In MCNP, particle transport relies entirely on nuclear data contained in externally supplied cross section tables \( (E_n < 20 \text{ MeV}) \), which are derived from evaluated nuclear data files. In LAHET, on the other hand, particle transport is accomplished by using various theoretical physics models embedded in the code, covering the energy range up to several GeV. In MCNPX, the table-based data are used whenever they exist, as such data are known to yield the best results. When they do not exist, the code built-in physics models are used.

Several physics models are available for high-energy transport in MCNPX. In the first stage, in which the incident particles interact with the individual nucleons via particle-particle cross sections, the Intranuclear Cascade (INC) and Multistage Pre-
equilibrium (Prael, 1998) Models are used. The INC model used in this study is the Bertini package (Bertini, 1963). In the second stage the nucleus undergoes either evaporation (emitting neutrons and light ions) or fission, while in the final stage the excited nucleus decays by gamma emission, with energies described by a decay library (PHTLIB).

2.3 Description of the Sources

Three different neutron sources have been considered in this study: a \( (d,d) \)-, a \( (d,t) \)- and a spallation source. It should be noted that \( (\alpha,n) \)- or spontaneous fission sources in the fuel have not been considered here.

2.3.1 The Fusion Sources used in MUSE-4

Two different fusion sources can be produced by the GENEPI neutron generator. 250 keV-deuterons are accelerated through the accelerator tube towards either a deuterium or a tritium target. The neutrons are emitted (the fusion reactions themselves are not simulated) from a point at the centre of the core (Fig. 2A). The energy of the emitted neutrons in the laboratory system (derived from basic kinematics) ranges from 2 to 3 MeV for the \( (d,d) \)-neutrons and from about 13 to 15 MeV for the \( (d,t) \)-neutrons, with a maximum emission probability density peaked in the forward direction. The source neutron energy spectrum and angular distribution used in this study are listed in Appendix B.

![Fig. 2](image)

2.3.2 The Spallation Source

For the purpose of producing the spallation source for the numerical simulations, the lead buffer/target in the model was extended by one extra subassembly towards the proton beam, replacing part of the accelerator tube (Fig. 2B). This was done in order to
maximize neutron production near the centre of the core (the same position where the 
\((d,d)\)- and the \((d,t)\)-source neutrons are emitted).

The simulations with the spallation source were divided into two steps. A first 
simulation with the 1000 MeV proton beam (the protons were uniformly distributed 
across the beam of diameter 4 cm) impinging on the lead target, was performed with MCNPX. The properties, in terms of angular, energy and spatial distribution, of the primary neutrons born from the spallation interactions were recorded. In the second step, these primary neutrons were supplied to the MCNPX code as fixed source neutrons for separate simulations.

The spatial range of the primary neutrons was found to be rather limited, most 
neutrons being emitted within a 3 to 4 cm radius around the z-axis and within the first 30 
cm axially, i.e., in the direction of the proton beam. The energy distribution of the neutrons produced from the 1000 MeV protons, integrated over all angles, is shown in 
Fig. 3 (neutrons created from secondary protons will have a slightly softer spectrum). We 
note that 17.3 % of the neutrons have energies higher than 20 MeV and 3.6 % of them 
higher than 150 MeV, and that these are mainly emitted in the forward direction of the incident proton beam. The effect of this high-energy fraction of neutrons on \(\phi^*\) will be discussed in Section 4.5.

The Sc3 sub-critical configuration of the MUSE-4 model with \(k_{\text{eff}} = 0.95\) was 
simulated. The neutron energy spectra resulting from the three different sources were 
calculated in the subassembly centred at 21.2 cm from the centre of the core, indicated 
with a cross in Fig. 1. The neutron spectra of the other sub-critical states (Sc1 and Sc2) 
are not shown here, since they are very similar to the spectra of Sc3.

It is seen in Fig. 4 that the three spectra at this position are very similar to each 
other and that they are largely dominated by the fission multiplication in the fuel. The
two dips in the neutron fluxes caused by the resonances in sodium (∼ 3 keV) and oxygen (∼ 0.4 MeV) can be seen. The fraction of neutrons still having their initial (source) energy is very small (but should not automatically be disregarded) – about 0.2 % of the \((d,t)\)-neutrons are still in the 14 MeV peak and 0.1 % of the spallation neutrons have energies above 20 MeV. Hence, we conclude that, for the purpose of computing neutron spectrum weighted quantities, the presence of the external sources can be considered “forgotten” beyond a few centimetres into the fuel.

![Fig. 4 MUSE-4 Neutron Energy Spectra in the Subassembly Centred at x=21.2 cm, y=-5.3 cm, Resulting from the three Different External Sources.](image)

### 4 SOURCE EFFICIENCY

The relative efficiency \(\phi^*\) of the source neutrons was determined for the three different sources and for the three different sub-critical configurations of the MUSE-4 model.

#### 4.1 Definition of \(\phi^*\)

The neutron flux distribution in a sub-critical core is the solution of the inhomogeneous balance equation:

\[
A\phi_S = F\phi_S + S
\]  

(1)

where \(F\) is the fission production operator, \(A\) is the net neutron loss operator and \(S\) is the external source. The quantity \(\phi^*\), which represents the relative efficiency of external source neutrons, is defined as the ratio of the average importance of the external source neutrons to the average importance of the fission neutrons (Salvatores, 1999), i.e.:

\[
\phi^* = \frac{\langle \phi_0^* , S \rangle}{\langle S \rangle} = \frac{\langle S \rangle}{\langle \phi_0^* , F\phi_S \rangle} = \frac{\langle \phi_0^* , F\phi_S \rangle}{\langle F\phi_S \rangle} \quad (2)
\]
where \( \phi_0^* = \) The adjoint flux (the everywhere positive solution of \( A^* \phi_0^* = \frac{1}{k_{\text{eff}}} F^* \phi_0^* \)) which provides a measure of neutron importance.

\(<F \phi_s^* > = \) Total production of neutrons by fission.
\(<S > = \) Total production of neutrons by the external source.

In the above formula, the brackets imply integration over space, angle and energy.

As some of the integrals in Eq. (2) cannot be directly calculated with MCNP and MCNPX, another procedure was sought to compute \( \phi^* \). By using the balance equation (Eq. 1), the properties of the adjoint flux \( \phi_0^* \), the \( A, F \) operators and their adjoints \( A^*, F^* \), the source efficiency can be expressed equivalently as

\[
\phi^* = \left( \frac{1}{k_{\text{eff}}} - 1 \right) \frac{<F \phi_s^* >}{<S >}
\]

Eq. (3) is a simple formula relating the total fission neutron production \(<F \phi_s^* >\) to the external source, \( \phi^* \) and reactivity \((1 - 1/k_{\text{eff}})\). It shows that, for given values of \( k_{\text{eff}} \) and \(<S >\), the larger \( \phi^* \) the larger the fission power produced in the system.

The quantities in the right hand side of Eq. (3) are standard outputs from MCNP and MCNPX. For simplicity, the production terms will be labelled only \( F \) and \( S \) in the sequel.

### 4.2 Estimation of the Statistical Error in \( \phi^* \)

To get an estimate of the statistical uncertainty in the source efficiency, assume that the errors of \( F \) and \( S \) are \( \Delta F \) and \( \Delta S \) and apply the formula for “propagation of error” (Eq. 4)

\[
\Delta f(x_1, x_2, ...) \approx \left[ \frac{\partial f}{\partial x_1} \Delta x_1 \right]^2 + \left[ \frac{\partial f}{\partial x_2} \Delta x_2 \right]^2 + \cdots + \rho_{12} \frac{\partial f}{\partial x_1} \Delta x_1 \frac{\partial f}{\partial x_2} \Delta x_2 + \cdots
\]

The correlation constant \( \rho \) could be either positive or negative – negative if \( k_{\text{eff}} \) and \( F \) are correlated and positive if they are anti-correlated. However, as a first approximation, \( \rho \) may be assumed to be zero. With the derivatives \( \frac{\partial \phi^*}{\partial k_{\text{eff}}}, \frac{\partial \phi^*}{\partial F} \) and \( \frac{\partial \phi^*}{\partial S} \) inserted in Eq. (4) an expression for the relative error in \( \phi^* \) can be obtained:

\[
\left( \frac{\Delta \phi^*}{\phi^*} \right)^2 \approx \left( \frac{1}{1-k_{\text{eff}}} \cdot \frac{\Delta k_{\text{eff}}}{k_{\text{eff}}} \right)^2 + \left( \frac{\Delta F}{F} \right)^2 + \left( \frac{\Delta S}{S} \right)^2
\]

Eq. 5 will be used in the subsequent sections to estimate the statistical uncertainty in \( \phi^* \).
4.3 Calculations of \( \phi^* \) for the MUSE-4 Model

The multiplication factor \( k_{\text{eff}} \) and the total number of neutrons produced by fission \( (F) \) were calculated for the three different sources and the three different sub-critical configurations. \( F \) was automatically normalised per source neutron, so \( S \) was always equal to 1. The source efficiency was calculated according to Eq. (3) and the corresponding statistical errors (±1 standard deviation) according to Eq. (5). All results including error estimations are listed in Table 1.

Table 1  
MCNP/MCNPX Results for the MUSE-4 Sc1, Sc2 and Sc3 Configurations.

<table>
<thead>
<tr>
<th>Source</th>
<th>( k_{\text{eff}} )</th>
<th>( F ) (( \pm ) %)</th>
<th>( \phi^* ) (( \pm ) %)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Sc1</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(D,D)-Source</td>
<td>0.99045</td>
<td>140.2 ( (\pm 1.6 %) )</td>
<td>1.35 ( (\pm 0.024) )</td>
</tr>
<tr>
<td>(D,T)-Source</td>
<td>( (\pm 8 \text{ pcm}) )</td>
<td>223.2 ( (\pm 1.7 %) )</td>
<td>2.15 ( (\pm 0.040) )</td>
</tr>
<tr>
<td>Spallation Source</td>
<td>0.99040</td>
<td>248.6 ( (\pm 1.8 %) )</td>
<td>2.41 ( (\pm 0.047) )</td>
</tr>
<tr>
<td><strong>Sc2</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(D,D)-Source</td>
<td>0.97007</td>
<td>44.2 ( (\pm 0.9 %) )</td>
<td>1.36 ( (\pm 0.015) )</td>
</tr>
<tr>
<td>(D,T)-Source</td>
<td>( (\pm 14 \text{ pcm}) )</td>
<td>69.9 ( (\pm 1.0 %) )</td>
<td>2.16 ( (\pm 0.024) )</td>
</tr>
<tr>
<td>Spallation Source</td>
<td>0.96992</td>
<td>76.6 ( (\pm 1.1 %) )</td>
<td>2.37 ( (\pm 0.028) )</td>
</tr>
<tr>
<td><strong>Sc3</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(D,D)-Source</td>
<td>0.94982</td>
<td>25.4 ( (\pm 0.6 %) )</td>
<td>1.34 ( (\pm 0.009) )</td>
</tr>
<tr>
<td>(D,T)-Source</td>
<td>( (\pm 14 \text{ pcm}) )</td>
<td>40.1 ( (\pm 0.5 %) )</td>
<td>2.12 ( (\pm 0.013) )</td>
</tr>
<tr>
<td>Spallation Source</td>
<td>0.94993</td>
<td>44.2 ( (\pm 0.7 %) )</td>
<td>2.33 ( (\pm 0.018) )</td>
</tr>
</tbody>
</table>

The energy of the \( (d,d) \)-source neutrons (2-3 MeV, see Appendix B) is only slightly larger than the average energy of a neutron produced by fission. The \( \phi^* \) value for the \( (d,d) \)-source is therefore expected to be equal or slightly larger than 1, which is indeed the case.

In the case of the \( (d,t) \)-source, the reason for the higher values of \( \phi^* \) is the larger fission rate, part of which coming from fissions induced by the neutrons multiplied by \( (n,2n) \)-reactions in the lead buffer. It is seen in Table 1 that the number of fission neutrons per source neutron is large, approximately 58 % larger than for the \( (d,d) \)-source. It is also seen in Fig. 5 that the \( (n,2n) \)-cross section in lead has a threshold at about 7 MeV, which is the reason why this reaction is insensitive to the \( (d,d) \)-source neutrons. At 14 MeV the value of the lead \( (n,2n) \)-cross section is about 2 barns, which is comparable to the fission cross section in Pu-239 and in U-238.

Concerning the spallation source neutrons, the values of \( \phi^* \) obtained in the simulations are somewhat higher than for the \( (d,t) \)-source. This is due to the fraction of neutrons having a very high energy (see Section 2.3.2). Most of the neutrons from the spallation process are born with an energy lower than the \( (n,2n) \)-cross section threshold in lead, but the neutrons with very high energy contribute significantly to \( \phi^* \), as will be shown in Section 4.5.
It is also seen in Table 1 that, for all three sources, \( \phi^* \) remains approximately constant or increases slightly as \( k_{\text{eff}} \) increases. This trend will be further discussed in the following section.

### 4.4 Dependence of \( \phi^* \) on Neutron Importance and \( k_{\text{eff}} \)

The dependence of the source efficiency on neutron importance \( \phi_0^* \) was investigated for a wider range of sub-criticality (\( k_{\text{eff}} = 0.70 \) to 0.996), for a \((d,d)\)- and a \((d,t)\)-source, using a spherical model consisting of a buffer core (lead or U-238 with \( r = 10 \) cm) and MUSE-4 type fuel. Only a limited class of importance variations was considered. The results are plotted in Fig. 6.

![Fig. 6](image)  
**Fig. 6** \( \phi^* \) versus \( k_{\text{eff}} \) for Spherical Configurations with a Buffer of Lead or U-238 \((r = 10 \) cm\) and MUSE-4 type Fuel, Coupled to a \((d,d)\)- and a \((d,t)\)-Source. The neutron importance and \( k_{\text{eff}} \) were changed by varying the fuel radius from about 48 cm \((k_{\text{eff}} \approx 0.70)\) to 68 cm \((k_{\text{eff}} \approx 0.996)\).
Instead of plotting the ratio $\varphi^*/(1/k_{\text{eff}} - 1)$ versus $k_{\text{eff}}$ which would reflect the rapid increase of $<F\phi_S>/<S>$ (and therefore of the fission power) as $k_{\text{eff}}$ approaches unity, we focused instead on the behaviour of $\varphi^*$ versus $k_{\text{eff}}$. The neutron importance (and therefore $k_{\text{eff}}$) was varied by changing the outer radius of the fuel from approximately 48 to 68 cm. It is seen in Fig. 6 that $\varphi^*$ shows the same almost constant or slightly increasing trend in the interval $k_{\text{eff}} = 0.95$ to 0.99, for the spherical configurations with the lead buffer, as already observed for the MUSE-4 model.

The first case (Case 1) is a sphere consisting of a lead core surrounded by fuel with approximately the same material composition as listed in Table 3. $\varphi^*$ increases slightly but constantly in the interval $k_{\text{eff}} = 0.70$ to 0.996. The importance of the $(n,2n)$-effect is also demonstrated by replacing the $(d,t)$-source by a $(d,d)$-source (Case 2), which results in significantly lower values of $\varphi^*$. The curve shows the same increasing trend as for the $(d,t)$-source.

In Case 3, when the lead buffer at the centre of the sphere is replaced by U-238, a large increase in $\varphi^*$ occurs at all sub-criticality levels because of U-238 fissions. The same increasing trend as with the lead buffer is not observed here as $\varphi^*$ remains nearly constant.

The statistical errors of the $\varphi^*$ values are rather small in the range $0.70 \leq k_{\text{eff}} \leq 0.99$ – less than 1 % ($\pm$1 standard deviation), while around 2.5 % for the very last point ($k_{\text{eff}} = 0.996$). In the absolute vicinity of criticality ($k_{\text{eff}} \geq 0.996$) the computation time for calculating $\varphi^*$ grows too large to obtain reliable results.

We conclude that the variations of $\varphi^*$ with neutron importance are rather small in the investigated range $0.70 \leq k_{\text{eff}} \leq 0.996$.

4.5 Decomposition of the Spallation Source

Most reactor code simulations only take into account neutrons with energies lower than 20 MeV. However, a significant fraction of the neutrons produced by spallation have energies higher than 20 MeV (see Fig. 3). The contribution of those high-energy neutrons to the source efficiency needs to be investigated. For this, the spallation source was artificially split into two “low-energy” bins ($S_1$ from 0 to 5 MeV and $S_2$ from 5 to 20 MeV) and two “high-energy” bins ($S_3$ from 20 to 150 MeV and $S_4$ from 150 to 1000 MeV). The study was performed for the third sub-critical level Sc3 ($k_{\text{eff}} = 0.95$) of the MUSE-4 model.

In order to derive a formula for the low- and high-energy contributions to the source efficiency, we start from Eq. (3), applied to each source bin

$$\varphi^*_i = \left( \frac{1}{k_{\text{eff}}} - 1 \right) \frac{<F\phi_i>}{<S_i>}$$

where $\phi_i =$ Flux resulting from each source bin ($S_1 \rightarrow \phi_1, S_2 \rightarrow \phi_2 \text{ etc.}$).
Since \( <F\phi_T> = \sum_{i=1}^{4} <F\phi_i> \), the following relationship for the decomposition of \( \varphi^* \) is readily obtained:

\[
\varphi_T^* = \sum_{i=1}^{4} \varphi_i^* \frac{<S_i>}{<S_T>}
\]

(7)

where \( \varphi_T^* \) = Efficiency of the total source.
\( \varphi_i^* \) = Efficiency of each source bin alone.

The \( \varphi_i^* \) results obtained from the Monte Carlo simulations are listed in Table 2. As expected, for the first low-energy bin, \( \varphi_1^* \) is low (\( \varphi_1^* = 1.25 \)) and close to the value obtained for the (d,d)-source. For the second bin, it is found to be higher (\( \varphi_2^* = 1.71 \)) since many of the neutrons have energies above the lead (n,2n)-cross section threshold. For the two high-energy parts, \( \varphi_i^* \) is very high (\( \varphi_3^* = 4.58 \) and \( \varphi_4^* = 14.4 \)), which is the consequence of fissions induced by neutrons born from (n,xn)-reactions and spallation interactions.

**Table 2**  MCNPX Results for the Sc3 MUSE-4 Model (\( k_{eff} = 0.94993 \)) Obtained from the Decomposition of the Spallation Source.

<table>
<thead>
<tr>
<th>Source Bin</th>
<th>Energy limits (MeV)</th>
<th>( &lt;S_i&gt;/A )</th>
<th>( &lt;F\phi_i&gt;/B )</th>
<th>( \varphi_i^* )</th>
<th>( \varphi_i^* \cdot &lt;S_i&gt;/C )</th>
<th>( &lt;S_T&gt;/D )</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>0 - 5</td>
<td>0.559</td>
<td>23.7</td>
<td>1.25</td>
<td>0.699 (30 %)</td>
<td></td>
</tr>
<tr>
<td>S2</td>
<td>5 - 20</td>
<td>0.268</td>
<td>32.5</td>
<td>1.71</td>
<td>0.458 (20 %)</td>
<td></td>
</tr>
<tr>
<td>S3</td>
<td>20 - 150</td>
<td>0.137</td>
<td>86.9</td>
<td>4.58</td>
<td>0.627 (27 %)</td>
<td></td>
</tr>
<tr>
<td>S4</td>
<td>150 - 1000</td>
<td>0.036</td>
<td>273.1</td>
<td>14.4</td>
<td>0.518 (23 %)</td>
<td></td>
</tr>
<tr>
<td>S_T D</td>
<td>0 - 1000</td>
<td>1.0</td>
<td>44.2</td>
<td>2.33</td>
<td>Sum = 2.30</td>
<td></td>
</tr>
</tbody>
</table>

The superscripts A, B, C and D in Table 2 stand for:
- A: Fraction of the total number of source neutrons in each energy bin (compare Fig. 3).
- B: Neutrons produced by fission in the core.
- C: Contribution to total \( \varphi^* \) (Product of column 3 and 5).
- D: Simulation with the total source, identical as for Sc3 in Table 1.

It is also seen in Table 2 that the two high-energy parts (17.3 % of the total number of source neutrons), contribute for about 50 % of the total \( \varphi^* \), and the highest energy part alone (3.6 % of the total number of source neutrons) for more than 20 %. The sum of the contributions to \( \varphi^* \) from the four different parts in the rightmost column, according to Eq. (7), is 2.30, which is in good agreement with the value obtained from the simulation with the total source (\( \varphi_T^* = 2.33 \)). The statistical 1σ error estimates in the \( \varphi^* \) values are less than 1 %.

The rather high average number of fission neutrons produced per source neutron for the two high-energy bins (87 and 273 respectively) might seem surprising at first. The explanation for this is that most of the high-energy neutrons from the spallation source...
have already been multiplied in the lead (via secondary spallation and \((n,xn)\)-reactions) \textit{before} they enter into the fuel. Each of them gives birth to a number of lower-energy neutrons, which then leak out of the lead and induce fission chain reactions in the fuel. Additional simulations in which the lead target alone was kept showed that only about 5 % of the neutrons leaking out of the lead have energies higher than 20 MeV and about 1 % of them higher than 150 MeV.

We conclude that, although neutron transport in the fuel is largely dominated by neutrons with low energy \((E_n < 20 \text{ MeV})\) which can be well simulated with a number of classical calculation codes such as MCNP, the importance of the high-energy contribution to \(\varphi^*\) indicates the need for further investigating the effects from high-energy spallation neutrons. This could be made easier by extending the capabilities of existing neutronics codes, in particular deterministic codes, for studying high-energy (20-150 MeV) neutron transport.

5 CONCLUSIONS

Numerical simulations have been performed with MCNP and MCNPX to investigate the neutronic properties of a sub-critical core representative of the up-coming MUSE-4 experiments, alternatively coupled with a \((d,d)\)-source, a \((d,t)\)-source and a spallation source. The source-plus-core systems have been studied in terms of neutronic spectra and efficiency \(\varphi^*\) in three different sub-critical configurations \((k_{\text{eff}} = 0.99, 0.97, 0.95)\).

The computed neutron spectra in all cases show that fission multiplication dominates at distances past a few centimetres into the fuel. This implies that, for the purpose of ADS core studies, the presence of the source may be ignored in the calculation of spectrum-weighted quantities, except possibly in the immediate vicinity of the external source.

The relative efficiency of the \((d,d)\)-source is somewhat higher than 1 (~1.35). For the \((d,t)\)-source, it is much larger, around 2.15. This significantly larger value is due to the \((n,2n)\)-multiplication in lead (with an energy threshold at about 7 MeV) and the induced fissions. To analyse the high value of \(\varphi^*\) obtained for the spallation source (~2.35), the source was artificially split into four different energy bins and the efficiency of each bin was determined. It was found that these two high-energy bins \((E_n > 20 \text{ MeV})\) contribute for about 50 % to \(\varphi^*\) and to the total number of fission neutrons produced in the core. This can be explained by the fact that primary neutrons born with high energy from spallation give birth to a large number of lower-energy neutrons, which in turn induce fissions. This rather high fraction indicates the need for extending reactor analysis code capabilities above 20 MeV for more detailed investigations of high-energy spallation neutron effects.

The variations of \(\varphi^*\) with neutron importance (and reactivity) was also investigated for different spherical configurations. It was found that \(\varphi^*\) remains approximately constant or increases slightly in the interval \(0.70 < k_{\text{eff}} < 0.996\).
AKNOWLEDGEMENTS

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REFERENCES


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APPENDIX A

Table 3 Material Composition for the Different Homogeneous Regions of the MUSE-4 Model.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Fuel</th>
<th>Na/SS Reflect</th>
<th>Axial Shield</th>
<th>Radial Shield</th>
<th>Lead Buffer</th>
<th>Accelerator Tube</th>
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<tr>
<td>C</td>
<td>2.75e-05</td>
<td>1.90e-05</td>
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<td>1.47e-03</td>
<td>1.64e-05</td>
<td>9.32e-05</td>
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<td>O-16</td>
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<td>Na-23</td>
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<td>Al-27</td>
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<td>-</td>
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Table 4  Laboratory-system Angular and Energy Dependence (Derived from Basic Kinematics) of the (d,d)- and the (d,t)-Source Neutrons Emitted at the Centre of the Core, Resulting from the GENEPI 250 keV Deuterons Impinging on a Deuterium/Tritium Target.

<table>
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<th>Angle</th>
<th>(D,D)-reaction</th>
<th>(D,T)-reaction</th>
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<tr>
<td></td>
<td>Energy</td>
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<tr>
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<td></td>
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<td>3.020</td>
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<td>2.984</td>
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<td>36</td>
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<td>45</td>
<td>2.876</td>
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<td>54</td>
<td>2.808</td>
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<td>2.734</td>
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<td>2.496</td>
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<td>180</td>
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Spallation Neutron Source Effects in a Sub-Critical System

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Department of Nuclear and Reactor Physics, Royal Institute of Technology, Stockholm, Sweden

R. Jacqmin

CEA Cadarache, SPRC/LEPh Bat. 230, 13108 Saint-Paul-lez-Durance, France

Abstract – Numerical simulations of a sub-critical system coupled to a neutron spallation source (1000 MeV protons impinging on a lead target) have been performed with the Monte Carlo code MCNPX and the deterministic code system ERANOS. The investigations have focused on the determination of the source neutron efficiency, $\phi^*$, i.e. the ratio of the average importance of external source neutrons to the average importance of fission neutrons. The calculations have been performed for a model representative of the MUSE-4 experiments currently underway in the MASURCA facility.

It has been found that the high-energy neutrons ($E_n > 20$ MeV) born from spallation, even though they represent only about 17% of the total neutrons, contribute for a large fraction (50%) to $\phi^*$ and to the total number of fission neutrons produced in the core. It has also been found that codes such as ERANOS, which do not take into account neutrons with energies higher than 20 MeV, largely underestimate $\phi^*$.

I. INTRODUCTION

Accelerator Driven Systems (ADS) (Ref. 1) are being investigated as a possible means for reducing the long-term radiotoxicity of the spent reactor fuel. In principle, the sub-criticality of ADS allows for dedicated cores with a much higher concentration of minor actinides than what is acceptable in critical reactors. Those dedicated cores could achieve high transmutation rates. Research done on ADS indicates that a radiotoxicity reduction factor of 50 to 100 is theoretically possible (Ref. 2).

In an ADS, neutrons generated by an intense external source, usually spallation reactions in a heavy metal target, are supplied to a sub-critical reactor core. This idea is being investigated in the MASURCA experimental facility at CEA Cadarache in the framework of the MUSE experiments (MUItiplication avec Source Externe) (Refs. 3, 4 and 5). Different configurations and several sub-critical levels are being studied.

The on-going MUSE-4 experiments do not use a spallation source. Instead, a high-intensity pulsed neutron generator GENEPI, constructed by CNRS/ISN/Grenoble, is being used to accelerate a 250 keV deuteron beam towards either a deuterium target or a tritium target, producing well-characterized neutron sources via fusion reactions.

In a previous study (Ref. 8), we investigated spallation neutron source effects in a MUSE-4 type sub-critical core coupled to a 1000 MeV proton beam, in particular the contribution of high-energy neutrons ($E_n > 20$ MeV) to the source efficiency ($\phi^*$). The objective of the present study is to complement this past work. Additional numerical simulations have been performed with the Monte Carlo code MCNPX (Ref. 6). The results have been compared to those obtained with ERANOS (Ref. 7), the CEA reference deterministic code system for fast reactor analyses.

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A description of the MUSE-4 model, the MCNPX and ERANOS codes and the spallation neutron source used in this study is given in Section II. In Section III, the specific procedure used for calculating $\phi^*$ with MCNPX and ERANOS is described. A decomposition of the spallation source is performed and analysed. The results of the MCNPX and ERANOS simulations are compared.

II. DESCRIPTION OF THE MUSE-4 MODEL, THE CALCULATION CODES AND THE SPALLATION NEUTRON SOURCE USED IN THIS STUDY

II.A. The Muse-4 Model

A homogeneous model representing one of the sub-critical configurations (SC3) planned in the MUSE-4 experiments ($k_{\text{eff}} = 0.95$) has been studied. The geometry of the model is shown in Fig. 1. The fuel is MOX fuel with 72% U-238, 21% Pu-239 and 5% Pu-240 plus small amounts of some other actinides. The fuel is homogeneously distributed together with the Na-coolant (Ref. 8). The axial (z-direction) dimension of the fuel is 60.96 cm, except in a 21.2 cm wide channel above and below the lead buffer and the accelerator tube (in the y direction), where it was extended by 10.16 cm. The sodium-steel reflector ends at $z = \pm 61.76$ cm. There is also a 10.16 thick axial shield (not shown in the figure) above and below the Na/SS reflector. The overall dimensions of the whole model, including the reflector and the shields, are 159 cm x 169.6 cm x 143.84 cm.

II.B. The Calculation Codes

II.B.1. MCNPX

MCNPX is an extended version of MCNP where the major capabilities of LAHET (Ref. 9) and MCNP-4B (Ref. 10) have been merged together. In MCNP, particle transport relies entirely on nuclear data contained in externally supplied cross section tables ($E_n < 20$ MeV), which are derived from evaluated nuclear data files. In LAHET, on the other hand, particle transport is accomplished by using various theoretical physics models embedded in the code, covering the energy range up to several GeV. In MCNPX, the table-based data are used whenever they exist, as such data are known to yield the best results. When they do not exist, the code built-in physics models are used.

Several physics models are available for high-energy transport in MCNPX. In the first stage, in which the incident particles interact with the individual nucleons via particle-particle cross sections, the Intranuclear Cascade (INC) and Multistage Pre-equilibrium (Ref. 11) Models are used. The INC model used in this study is the Bertini package (Ref. 12). In the second stage the nucleus undergoes either evaporation (emitting neutrons and light ions) or fission, while in the final stage the excited nucleus decays by gamma emission, with energies described by a decay library (PHTLIB).

In this study, all simulations performed with MCNPX relied on the same evaluated nuclear data library, namely ENDF/B-VI.6.

II.B.2. ERANOS

ERANOS is a deterministic fast reactor code system developed by CEA in collaboration with other R&D organizations (Ref. 7). It uses cross-section libraries based on the JEF2.2 evaluated file. The ERANOS code system is well validated for classical sodium-cooled fast reactors. This validation has been recently extended to plutonium burning cores with steel-sodium reflectors and high Pu-content. However, the code is not yet fully validated for systems characterized by large sub-criticalities and the presence of high-energy neutrons from spallation.

In ERANOS, 1-D cell or 2-D subassembly calculations are performed with the ECCO code, while core calculations can be performed with different 2-D or 3-D, diffusion or transport theory modules. In this study, the two-dimensional $S_n$ transport code BISTRO (Ref. 13) was used.
II.C. Description of the Spallation Source and the Primary Spallation Neutrons

The MCNPX simulations calculating the efficiency of the spallation source neutrons were divided into two steps. In the first simulation, a large number of protons (1000 MeV) were accelerated towards the lead target (Fig. 2). The protons were uniformly distributed across the beam of radius 2 cm. The angular, energy and spatial distributions of all neutrons that were created directly from the spallation interactions (primary spallation neutrons) were recorded. After that the neutron trajectories were immediately terminated. This procedure produces the spectrum of primary spallation neutrons, i.e. no secondary neutrons are included.

![Fig. 2. 1000 MeV protons accelerated towards the lead target creating neutrons via spallation interactions. The generated primary neutrons are “frozen” at the moment when they are created, and emitted as fixed source neutrons in a separate simulation.](image)

In the second step, these primary spallation neutrons were supplied to the MCNPX code as fixed source neutrons for separate simulations and the source efficiency was determined.

The spatial distribution where the primary neutrons were created was found to be rather limited. Axially, most of the neutrons were emitted in the upper part of the lead target (77% within the first 20 cm, see Fig. 3A). The radial distribution was found to be very peaked around the axis of the incident proton beam, about 98% of the neutrons were created within a 3 cm radius (Fig. 3B).

![Fig. 3. A) Axial distribution of the primary spallation neutrons. B) Radial distribution of the primary spallation neutrons.](image)

The energy distribution of the primary neutrons produced by the 1000 MeV protons is shown in Fig. 4. We note that 16.8% of the neutrons have energies higher than 20 MeV and 3.3% of them higher than 150 MeV, and that the neutrons with very high energy are mainly emitted in the forward direction of the proton beam, as expected.

![Fig. 4. Energy spectrum of the primary spallation neutrons created by the 1000 MeV protons.](image)

There were about 14.5 primary spallation neutrons produced per 1000 MeV proton. This value should be compared to the total number of neutrons produced in the lead target, i.e. around 21 neutrons per proton. Thus, almost a third of the neutrons that exit the lead target and enter into the fuel are secondary neutrons, most of them created in neutron spallation reactions and \((n,xn)\)-reactions.
It should be noted that, when calculating $\phi^*$ for a spallation system, the results are directly dependent on the definition of the neutron source. Other source definitions are possible (Refs. 14 and 15), which will result in different meanings and values for $\phi^*$.

I. SOURCE EFFICIENCY

III.A. Definition of $\phi^*$

The neutron flux distribution $\phi$ in a sub-critical core is the solution of the inhomogeneous balance equation:

$$A\phi_s = F\phi_s + S$$ (1)

where $F$ is the fission production operator, $A$ is the net neutron loss operator and $S$ is the external source. The quantity $\phi^*$, which represents the relative efficiency of external source neutrons, is defined as the ratio of the average importance of the external source neutrons to the average importance of the fission neutrons (Ref. 1), i.e.:

$$\phi^* = \frac{<\phi^*_{0} S>}{<S>} \frac{<S>}{<\phi^*_{0} F\phi_s> <F\phi_s>$$ (2)

where

$\phi^*_{0}$ = The adjoint flux (the everywhere positive solution of $A^*\phi^*_{0} = 1/k_{eff} F^*\phi^*_{0}$), which provides a measure of neutron importance.

$<F\phi_s>$ = Total production of neutrons by fission.

$<S>$ = Total production of neutrons by the external source.

In the above formula, the brackets imply integration over space, angle and energy.

As some of the integrals in Eq. (2) cannot be directly calculated with MCNPX, another procedure was sought to compute $\phi^*$. By using the balance equation [Eq. (1)], the properties of the adjoint flux $\phi^*$, the $A$, $F$ operators and their adjoints $A^*$, $F^*$, the source efficiency can be expressed equivalently as:

$$\phi^* = \left(\frac{1}{k_{eff}} - 1\right) \frac{<F\phi_s>}{<S>}$$ (3)

Eq. (3) is a simple formula relating the total fission neutron production $<F\phi_s>$ to the external source, $\phi^*$ and reactivity ($1 - 1/k_{eff}$). It shows that, for given values of $k_{eff}$ and $<S>$, the larger $\phi^*$ the larger the fission power produced in the system.

The quantities in the right hand side of Eq. (3) are standard outputs from MCNPX.

III.B. Decomposition of the Spallation Source

Most reactor codes take into account only neutrons with energies lower than 20 MeV. However, a significant fraction of the neutrons produced by spallation have energies higher than 20 MeV (Fig. 4). The contribution of those high-energy neutrons to the source efficiency needs to be investigated. For this, the spallation source was artificially split into two “low-energy” bins ($S_i$ from 0 to 5 MeV and $S_j$ from 5 to 20 MeV) and two “high-energy” bins ($S_l$ from 20 to 150 MeV and $S_m$ from 150 to 1000 MeV), as explained in Ref. 8.

In order to derive a formula for the low- and high-energy contributions to the source efficiency, we start from Eq. (3), applied to each source bin

$$\phi^* = \left(\frac{1}{k_{eff}} - 1\right) \frac{<F\phi_i>}{<S_i>}$$ (4)

where

$\phi_i$ = Flux resulting from each source bin alone ($S_i \rightarrow \phi_i$, $S_j \rightarrow \phi_j$, etc.).

Since $<F\phi_T> = \sum_{i=1}^{4} <F\phi_i>$, the following relationship for the decomposition of $\phi^*$ is readily obtained:

$$\phi^* = \sum_{i=1}^{4} \phi^*_i \frac{<S_i>}{<S_T>}$$ (5)

where

$\phi^*_T$ = Efficiency of the total source.

$\phi^*_i$ = Efficiency of each source bin alone.

III.B.1. Calculations Performed with MCNPX

The $\phi^*_i$ results obtained from the MCNPX simulations are listed in Table I. As expected, for the first low-energy bin, $\phi^*_1$ is relatively low ($\phi^*_1 = 1.24$). For the second bin, it is found to be higher ($\phi^*_2 = 1.63$), since many of the neutrons have energies above the lead $(n,2n)$-cross section threshold (ref. 8). For the two high-energy parts, $\phi^*_3$ is very high ($\phi^*_3 = 4.79$ and $\phi^*_4 = 13.9$), which is the consequence of fissions induced by secondary neutrons born from $(n,xn)$-
reactions and neutron spallation interactions. The statistical relative 1σ error estimates in the \( \phi^* \) values are about 1%.

### TABLE I

<table>
<thead>
<tr>
<th>Source Bin</th>
<th>Energy intervals (MeV)</th>
<th>( &lt;S_1&gt;_A / &lt;S_T&gt; )</th>
<th>( &lt;F\phi_i&gt; &gt; B / &lt;S_i&gt; )</th>
<th>( \phi^*_i )</th>
<th>( \phi^*_i \cdot &lt;S_i&gt; &gt; C / &lt;S_T&gt; )</th>
</tr>
</thead>
<tbody>
<tr>
<td>S_1</td>
<td>0 - 5</td>
<td>0.592</td>
<td>23.5</td>
<td>1.24</td>
<td>0.736 (33%)</td>
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<td>S_2</td>
<td>5 - 20</td>
<td>0.240</td>
<td>31.1</td>
<td>1.63</td>
<td>0.390 (17%)</td>
</tr>
<tr>
<td>S_3</td>
<td>20 - 150</td>
<td>0.135</td>
<td>91.3</td>
<td>4.79</td>
<td>0.647 (29%)</td>
</tr>
<tr>
<td>S_4</td>
<td>150 - 1000</td>
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<td>264.3</td>
<td>13.9</td>
<td>0.458 (21%)</td>
</tr>
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<td>0 - 1000</td>
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<td>42.2</td>
<td>2.21</td>
<td>Sum = 2.23</td>
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</tbody>
</table>

The superscripts \( A, B, C \) and \( D \) in Table I stand for:

- \( A \): Fraction of the total number of source neutrons in each energy bin (compare Fig. 4).
- \( B \): Neutrons produced by fission in the core, per external source neutrons from bin \( i \).
- \( C \): Contribution to total \( \phi^* \) (Product of column 3 and column 5).
- \( D \): Simulation with the total source.

It is also seen in Table I that the two high-energy parts (16.8% of the total number of source neutrons), contribute for about 50% of the total \( \phi^* \), and the highest energy part alone (3.3% of the total number of source neutrons) for more than 20%. The sum of the contributions to \( \phi^* \) from the four different parts in the rightmost column, according to Eq. (5), is 2.23, which is in agreement with the value obtained from the simulation with the total source (\( \phi^*_T = 2.21 \)). Comparisons with additional calculations (Ref. 8) show that this value is slightly higher than the \( \phi^*_d \)-value obtained for a \((d,t)\)-fusion source, coupled to the same model (\( \phi^*_{d,t} = 2.12 \)) and much higher than for a \((d,d)\)-source (\( \phi^*_{d,d} = 1.34 \)).

The rather high average number of fission neutrons produced per source neutron for the two high-energy bins (91 and 264, respectively) might seem surprising at first. The explanation for this is that most of the high-energy neutrons from the spallation source have already been multiplied in the lead (most of them via secondary neutron spallation and \((n,xn)\)-reactions) before they enter into the fuel. Each of them gives birth to a number of lower-energy neutrons, which then leak out of the lead and induce fission chain reactions in the fuel. Additional simulations in which the lead target alone was kept show that only about 5% of the neutrons leaking out of the lead have energies higher than 20 MeV and about 1% of them higher than 150 MeV.

We conclude that, although neutron transport in the fuel is largely dominated by neutrons with low energies (\( E_n < 20 \) MeV) which can be well simulated with a number of classical calculation codes such as MCNP and ERANOS, high-energy neutrons contribute significantly to \( \phi^* \). Further investigating these high-energy effects would be made easier by extending the neutron data libraries of existing codes from 20 MeV to at least 150 MeV.

### III.B.2. Comparisons between MCNPX and ERANOS

In practice, many hybrid system core studies rely on deterministic codes such as ERANOS, which do not model neutrons with energies above 20 MeV. It is therefore interesting to compare the predictions of such codes with MCNPX. While only small differences are expected in reactivity and power shape predictions (see Fig. 5), the results of the previous section suggest that a rather large impact is anticipated on \( \phi^* \).

To verify this conjecture, a small benchmark was defined and calculated with both MCNPX and ERANOS. This benchmark is a simplified two-dimensional \( R-Z \) version of the MUSE-4 model described in Fig. 1. The distribution of the primary source neutrons was slightly simplified to make it possible to use exactly the same sources in both ERANOS and MCNPX.
There are at least two ways of calculating $\varphi^*$ with ERANOS. The first possibility is to use Eq. 2. However, this requires an adjoint calculation. Another, simpler way is to use Eq. 3, which may also be written as:

$$
\varphi^* = \frac{1/k_{\text{eff}} - 1}{1/k_s - 1}
$$

(6)

where $k_s = <F\phi_s>/<A\phi_s>$. We chose this second alternative.

As can be seen in Table II, ERANOS produces values of $\varphi^*$ in fairly good agreement with MCNPX for the two low-energy bins $S_1$ and $S_2$, in spite of differences between the nuclear datasets used by the two codes. The relative statistical 1σ error estimates in the $\varphi^*$ values for the MCNPX-calculations are less than 1%.

### IV. CONCLUSIONS

Numerical simulations have been performed with MCNPX and ERANOS to investigate the neutronic properties of a sub-critical model ($k_{\text{eff}} \approx 0.95$) representative of the on-going MUSE-4 experiments, coupled to a spallation source (1000 MeV protons impinging on a lead target). The system has been studied in terms of source efficiency ($\varphi^*$).

The efficiency of the total spallation source was found to be 2.21, which could be compared to $\varphi^*$ for a $(d,t)$- and a $(d,d)$-source, coupled to the same system ($\varphi^*_{(d,t)} = 2.12$ and $\varphi^*_{(d,d)} = 1.34$, respectively). To analyse this rather high value of $\varphi^*$, the spallation source was artificially split into four energy bins and the efficiency of each bin was determined. It was found that the two high-energy bins ($E_n > 20$ MeV) contribute for about 50% to $\varphi^*$ and to the total number of fission neutrons produced in the core. This can be explained by the fact that primary neutrons born with high energy from spallation give birth to a large number of lower-energy neutrons.

However, since the ERANOS libraries are currently limited to neutrons below 20 MeV, the value of $\varphi^*$ for the total source is much lower ($\varphi^*_{\text{ERANOS}} = 1.42$) than the MCNPX value ($\varphi^*_{\text{MCNPX}} = 2.17$). A large fraction (~ 35%) of the total value of $\varphi^*$ is actually not reflected in the ERANOS results. This is something one should bear in mind when calculating $\varphi^*$ for a spallation source with ERANOS or any other “low”-energy reactor code.

### TABLE II

<table>
<thead>
<tr>
<th>Source Bin</th>
<th>Energy intervals (MeV)</th>
<th>$&lt;S_i&gt; / &lt;S_T&gt;$</th>
<th>$\varphi^*_{i}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$S_1$</td>
<td>0 - 5</td>
<td>0.592</td>
<td>1.20</td>
</tr>
<tr>
<td>$S_2$</td>
<td>5 - 20</td>
<td>0.240</td>
<td>1.56</td>
</tr>
<tr>
<td>$S_3$</td>
<td>20 - 150</td>
<td>0.135</td>
<td>4.56</td>
</tr>
<tr>
<td>$S_4$</td>
<td>150 - 1000</td>
<td>0.033</td>
<td>-</td>
</tr>
<tr>
<td>$S_T$</td>
<td>0 - 1000</td>
<td>1.0</td>
<td>2.17</td>
</tr>
</tbody>
</table>

* When simulating the total source with ERANOS, all neutrons above 20 MeV (16.8%) were placed in the highest energy group (14.2 to 19.6 MeV).
neutrons, which in turn induce fissions. This finding indicates the need for extending reactor analysis code capabilities above 20 MeV for more detailed investigations of high-energy spallation neutron effects.

Comparisons of ERANOS and MCNPX calculations of $\phi^*$ were found to be in good agreement for the energy bins below 20 MeV. However, as ERANOS does not take into account neutrons above 20 MeV, it largely underestimates the total value of $\phi^*$. This effect should be remembered when calculating $\phi^*$ with reactor codes that do not account for neutrons above 20 MeV.

AKNOWLEDGEMENTS

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REFERENCES


III
Definition and Application of Proton Source Efficiency in Accelerator Driven Systems

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Abstract – In order to study the beam power amplification of an accelerator driven system (ADS), a new parameter, the proton source efficiency ($\psi^*$) is introduced. $\psi^*$ represents the average importance of the external proton source, relative to the average importance of the eigenmode production, and is closely related to the neutron source efficiency ($\phi^*$), which is frequently used in the ADS field. $\phi^*$ is commonly used in the physics of sub-critical systems driven by any external source (spallation source, (d,d), (d,t), Cf-252 spontaneous fissions etc.). On the contrary, $\psi^*$ has been defined in this paper exclusively for ADS studies, where the system is driven by a spallation source. The main advantage with using $\psi^*$ instead of $\phi^*$ for ADS is that the way of defining the external source is unique and that it is proportional to the core power divided by the proton beam power, independently of the neutron source distribution.

Numerical simulations have been performed with the Monte Carlo code MCNPX in order to study $\psi^*$ as a function of different design parameters. It was found that, in order to maximize $\psi^*$, and therefore minimizing the proton current needs, a target radius as small as possible should be chosen. For target radii smaller than about 30 cm, lead-bismuth is a better choice of coolant material than sodium, regarding the proton source efficiency, while for larger target radii the two materials are equally good. The optimal axial proton beam impact was found to be located approximately 20 cm above the core center. Varying the proton energy, $\psi^*/E_p$ was found to have a maximum for proton energies between 1200 and 1400 MeV. Increasing the americium content in the fuel decreases $\psi^*$ considerably, in particular when the target radius is large.

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I. INTRODUCTION

Accelerator Driven Systems (ADS) [1, 2, 3, 4] are being investigated as a possible mean for reducing the long-term radiotoxicity of spent reactor fuel. In principle, the sub-criticality of ADS allows for dedicated cores with a much higher concentration of minor actinides than what is acceptable in critical reactors. Such dedicated cores enable multi-recycling of americium and curium, providing thus a potential for reducing the radio-toxicity by a factor of 50 to 100 [5].

In ADS, a high-power particle accelerator is used to accelerate protons to energies of the order of 1000 MeV. The protons impinge on a heavy metal target, generating a large number of neutrons via spallation. The spallation neutrons leak out from the target, after different kinds of interactions with the target nuclei, and are subsequently multiplied in the surrounding sub-critical blanket.

An important factor when designing an ADS is to optimize the beam power amplification, given that the reactor is operating at a certain sub-critical reactivity (with sufficient safety margins to criticality). Optimizing the source efficiency, and thereby minimizing the proton current needs, can have an important impact on the overall design of an ADS and on the economy of its operation. The neutron source efficiency parameter \( \varphi^* \) is commonly used to study this quantity, since it is related to the number of fissions produced in the core (which is closely related to the core power), by an average external source neutron.

However, calculating \( \varphi^* \) for an accelerator driven system introduces some complications, since the actual source particles are protons, and not neutrons. In order to determine \( \varphi^* \), the external neutron source first has to be defined and then the efficiency of this neutron source can be determined. A drawback with using \( \varphi^* \) is that the neutron source can be defined in several different ways, and the results are directly dependent on the choice of definition. Therefore, completely different values of \( \varphi^* \) are often observed [6, 7, 8], due to different choices of external neutron source definition. Another complication associated to the neutron source efficiency is that, studying \( \varphi^* \) as a function of a certain system parameter might change the neutron source distribution and the number of neutrons produced per source proton. In this case, in order to represent the beam power amplification, \( \varphi^* \) needs to be weighted with the number of source neutrons produced per source proton.

With the motivation of simplifying the concept of source efficiency, we introduce in this paper a new parameter \( \psi^* \), which refers to the number of fission neutrons produced in the system by each source proton. The advantages with using the proton source efficiency instead of the neutron source efficiency is that there is no ambiguity in how to define the external source, and that it is proportional to the beam power amplification, without the need of a weighting factor.

In this paper, \( \psi^* \) has been studied as a function of different system parameters, in order to find the optimal design conditions for maximum proton beam amplification, within the given safety constraints. For this purpose, a model of a nitride fuelled and lead-bismuth cooled ADS was used. First, the neutron source efficiency is defined and discussed (Section II) and then the proton source efficiency parameter is introduced (Section III). Section IV describes the reference model used in this study. In Section V.A, \( \psi^* \) and \( \varphi^* \) are studied as functions of the target radius and coolant material. In the following sections, \( \psi^* \) is studied as a function of the axial proton beam impact (V.B) and the proton beam energy (V.C). Finally, an americium based fuel has been compared with the plutonium based reference fuel (V.D).

II. NEUTRON SOURCE EFFICIENCY \( \varphi^* \)

II.A. Definition of the Neutron Source Efficiency

The neutron flux distribution \( \phi_n \) in a sub-critical core is the solution to the inhomogeneous steady-state neutron transport equation

\[ A \phi_n = F \phi_n + S_n \]

where \( F \) is the fission production operator, \( A \) is the net neutron loss operator and \( S_n \) is the external neutron source.

The neutron source efficiency [9, 10], usually denoted \( \varphi^* \), represents the efficiency of the external source neutrons and can be expressed according to the following equation [11]:

\[ \varphi^* = \left( \frac{1}{k_{\text{eff}}} - 1 \right) \frac{<F\phi_n>}{<S_n>} \]

which is valid in the range \( 0 < k_{\text{eff}} < 1 \). \( <F\phi_n> \) is the total production of neutrons by fission and \( <S_n> \) is the total production of neutrons by the external source. In the above formula, the brackets imply integration over space, angle and energy. Eq. (2) relates the total fission neutron production \( <F\phi_n> \) to the external neutron source, \( \varphi^* \) and the reactivity \( (1 - 1/k_{\text{eff}}) \). It shows that, for given values of \( k_{\text{eff}} \) and \( <S_n> \), the larger \( \varphi^* \) the larger the fission power produced in the system.

II.B. Definition of the External Neutron Source

Since the actual source particles in an accelerator driven system are protons and not neutrons, it is not obvious which
is the best way to define the neutron source. The procedure to calculate \( \varphi^* \) is usually divided into two steps – the first generating the source neutrons, produced from the proton beam interacting with the target, and the second one determining the efficiency of these source neutrons. Different source definitions are possible and they will result in different values and meanings of \( \varphi^* \). A brief summary of four different definitions of the external neutron source that have been used in the ADS field is given in [12]. Among these, the two most frequently used definitions, the target neutron leakage source and the energy cut-off source, will be discussed in the following sections. The other two definitions are the fission source, consisting of the first generation of fission neutrons, and the primary neutron source, which is the collection of neutrons that are created directly from proton induced spallation (primary spallation neutrons) [8].

II.B.1. The Target Neutron Leakage Source

This approach uses the neutrons that leak out radially from the target as source neutrons [6, 13]. The method consists of, in the first step, transporting the high-energy protons and the secondary particles that they produce in the target. The neutrons that leak out from the target are defined as the source, and their properties, in terms of position, direction and energy, are written to a source file. Only the target is present in the first simulation, so no once-leaked neutrons re-entering the target are included in the source definition. In the second step, the leakage neutrons are reemitted as fixed source neutrons in a separate run and the efficiency \( \varphi^* \) of them is determined. Since the target neutron leakage spectrum includes a high-energy tail, both step 1 and 2 need to be simulated with a high-energy transport code (which can simulate neutrons with energy up to the incident proton energy).

Since the neutron source is generated by a proton beam/target simulation, the distribution of the source neutrons is dependent on the target properties and the proton beam properties. This might induce complications when trying to optimize \( \varphi^* \) and the beam power amplification, by varying different system parameters. If a change in the studied system parameter changes the distribution of the source neutrons, \( \varphi^* \) has to be weighted by the number of neutrons produced per source proton. With the target neutron leakage definition, examples of these parameters are the target dimension, the proton beam energy or the axial proton beam impact position. Other system parameters, such as the core coolant material, the fuel composition or the core dimensions, are independent of the target region and do not affect the neutron source.

II.B.2. The Energy Cut-off Neutron Source

The other way to define the neutron source is to collect the neutrons that fall below a certain cut-off energy (usually 20 or 150 MeV) [7, 14, 15]. In the first step, a high-energy code is used to transport the accelerated protons and the secondary high-energy particles. The neutrons that are produced are either killed if they are born below the cut-off energy or transported until they fall below this energy. The properties of the killed neutrons are written to a source file. In the second step, these neutrons are reemitted as fixed source neutrons in a separate run and \( \varphi^* \) is determined.

An advantage of this approach is that the second step can be simulated with a low-energy transport code. The cut-off energy is set to the upper energy of the cross section library that will be used in the second step calculation. This is desirable since many reactor codes systems are limited to the upper energy limit of the cross-section data library (e.g. 20 or 150 MeV).

It has been shown in [7] that, in contrast to the target neutron leakage source, the neutron source distribution in this case is rather insensitive to changes in the target radius. However, substituting the coolant material or changing the fuel composition will affect the distribution of the neutron source.

III. PROTON SOURCE EFFICIENCY \( \psi^* \)

III.A. Introduction of the Proton Source Efficiency

In order to simplify the concept of source efficiency, a new parameter, called “proton source efficiency” and denoted \( \psi^* \), which represents the product of \( \varphi^* \) and the number of source neutrons generated per source proton \( (S_n/S_p) \), is introduced in this paper. We thus have the following relation between the proton source efficiency \( \psi^* \) and the neutron source efficiency \( \varphi^* \):

\[
\psi^* = \varphi^* \cdot \frac{<S_n>}{<S_p>} \quad (3)
\]

This parameter could also, in analogy with \( \varphi^* \), be expressed in terms of \( k_{en} \) and the total number of neutrons produced by fission in the core, for each source proton. Inserting Eq. (2) in Eq. (3) it is expressed in the same way as \( \varphi^* \), only with the replacement of \( S_n \) by \( S_p \).

\[
\psi^* = \left( \frac{1}{k_{eff}} - 1 \right) \cdot \frac{<F\psi_s>}{<S_p>} \quad (4)
\]

\( <F\psi_s>/<S_p> \) is the total production of neutrons by fission over the total number of source protons.
Considering $\psi^*$ and $\varphi^*$ (according to the target neutron leakage definition) as functions of the target radius and of coolant material illustrates the discussion in the previous section about which design parameters that affect and do not affect the neutron source distribution. Changing the coolant material, $S_n/S_p$ does not change, so $\psi^*$ and $\varphi^*$ vary in exactly the same way. When varying the target radius, on the other hand, $S_n/S_p$ changes, so $\psi^*$ and $\varphi^*$ varies in different ways. Consequently, if one wants to use the neutron source efficiency parameter $\varphi^*$, it needs to be weighted with $S_n/S_p$ when it is studied as a function of the target radius, whereas this is not necessary when comparing different core coolant materials. However, with the introduction of the proton source efficiency and always referring to $\psi^*$, none of this has to be considered, and the procedure is simplified.

III.B. Relationship between $\psi^*$ and the Core Power

The total power produced by fission in the core ($P_f$) can be expressed as the product of the total number of fission events and the average available energy released in a fission, according to the following relation;

$$P_f = \frac{<F\phi>_v}{\nu} \cdot \bar{E}_f\quad (5)$$

Inserting Eq. (4) in Eq. (5), we obtain

$$\frac{P_f}{<S_p>/v} = \frac{\bar{E}_f}{\bar{\nu}} \cdot \frac{k_{\text{eff}}}{1-k_{\text{eff}}} \cdot \psi^*\quad (6)$$

For a given fuel composition, $\bar{E}_f$ and $\bar{\nu}$ can, for the purpose of this study, be considered to be constant. Even though there are high-energy neutrons entering into the fuel (the neutron yield $\nu$ is not constant with respect to neutron energy), the fraction of fissions in the core that are induced by high-energy neutrons is very small. Therefore, a change in the neutron yield for these fission events will have very little impact on $\bar{\nu}$. The variations of $\bar{E}_f$ and $\bar{\nu}$ have been calculated for the series of different studies presented in this paper and were found to be much smaller than the statistical errors in the simulations (except in the case where the fuel composition was modified). When the fuel composition changes, $\bar{\nu}$ might also change, which should be kept in mind in Section V.D, where two different fuels are compared.

If we further make the approximation that the energy produced by fission is proportional to the total core power produced in the core, we find that $\psi^*$ is proportional to the total power divided by the source intensity (output/input power).

IV. SYSTEM MODELING

A homogenized model representing a nitride fuelled and lead-bismuth cooled ADS (maximum 800 MWth) has been studied. The height of the active core in the reference model (Fig. 1) is 100 cm and the outer radius is 70 cm. The inner radius is 20 cm, which is also the boundary of the lead-bismuth target. The accelerator tube has a radius of 15 cm and the axial position of the proton beam impact is 25 cm below the top of the core. The radius of the radially uniform 1000 MeV proton beam is 7.5 cm. Above and below the active zone of the core, plena for accommodation of gas release are included, having lengths of 100 and 50 cm, respectively. The radial reflector is assumed to consist of 90 % steel and 10 % lead-bismuth.

![Fig. 1. RZ-view of the homogenised reference model. The 1000 MeV protons are guided through the accelerator tube and impinge on the Pb-Bi target. The different regions in the model are pure Pb-Bi (1), the plena (2), the active core (3) and the reflector (4).](image-url)

The relative fraction of fuel, cladding and coolant material used in the homogenized model correspond to a pin radius of 2.5 mm and P/D = 1.72. The nitride actinide fuel is in solid solution with ZrN. The volume fraction of ZrN was adjusted to 83 %, in order to obtain a $k_{\text{eff}}$ of about 0.95. The fuel consists of 80% plutonium and 20% americium (the actinide vectors are listed in Table I). The spallation target and the core coolant consist of lead-bismuth eutectic and the fuel pin cladding of 10% chromium and 90% iron.
TABLE I
Relative Fraction of Actinides in the Reference Fuel. The Pu vector corresponds to that of spent LWR MOX fuel after 7 years of cooling and the Am vector to a mixture of spent UOX (Uranium Oxide Fuel) and MOX (Mixed Oxide Fuel) fuel.

<table>
<thead>
<tr>
<th>Plutonium</th>
<th>80 %</th>
<th>Americium</th>
<th>20 %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-238</td>
<td>5 %</td>
<td>Am-241</td>
<td>67 %</td>
</tr>
<tr>
<td>Pu-239</td>
<td>38 %</td>
<td>Am-243</td>
<td>33 %</td>
</tr>
<tr>
<td>Pu-240</td>
<td>30 %</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu-241</td>
<td>13 %</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu-242</td>
<td>14 %</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The Monte Carlo code MCNPX [16] (Version 2.3.0), in coupled neutron and proton mode, was used for all simulations, relying on the evaluated nuclear data library ENDF/B-VI.8 (limited to 20 MeV). The Intranuclear Cascade model used by MCNPX was the Bertini package [17].

V. THE PROTON SOURCE EFFICIENCY AS A FUNCTION OF DIFFERENT SYSTEM PARAMETERS

The proton source efficiency $\psi^*$ has been studied as a function of a number of system parameters, such as the target radius, coolant material, axial proton beam impact position, proton beam energy and fuel composition. The starting point for each parameter study is the reference model. In each case when a parameter has been changed, the reactivity has been re-adjusted in order to keep $k_{\text{eff}}$ constant at about 0.95.

V.A. $\psi^*$ as function of Target Radius

In this section, $\psi^*$ has been computed for different target radii, for the reference lead-bismuth cooled core and for a sodium cooled core. The neutron source efficiency $\psi^*$, using the target neutron leakage definition, has also been determined. $k_{\text{eff}}$ was kept constant at 0.95 by adjusting the outer radius of the core.

Since changing the geometry of the core might affect the results, $\psi^*$ has also been studied as a function of the outer core radius, varying from 60 to 90 cm. It was found that an increase of the core radius leads to a slight increase of $\psi^*$, on average 0.26% per cm. The explanation for this is that the radial neutron leakage into the reflectors decreases as the core radius increases. This dependence of $\psi^*$ (rather small but not automatically negligible) on the core radius should be kept in mind in the parameter studies where the core radius is varying.

$\psi^*$ was computed for the reference model for different target radii, both as the product of $\phi^*$ and $S_p/S_n$ (Eq. 3) and directly according to Eq. (4), in order to verify the consistency between the two different expressions. $\phi^*$ was calculated using the target neutron leakage definition. The results are listed in Table II and we see that they are in good agreement with each other, the differences being within the statistical uncertainty. Hence, using a two-step simulation procedure, $\psi^*$ can be obtained according to Eq. (3), independently of the choice of neutron source definition. If MCNPX is used, $\psi^*$ can be obtained directly according to Eq. (4). $\psi^*$, $\phi^*$ and $S_p/S_n$ are also plotted as functions of target radius later on in this section.

TABLE II
Computation of $\psi^*$ according to Eq. (3) and Eq. (4) (1σ-error ~ 0.75 %). For the 10 cm target radius, the radius of the accelerator tube was decreased to 10 cm. The results are also plotted below.

<table>
<thead>
<tr>
<th>Target Radius</th>
<th>$\phi^*$</th>
<th>$&lt;S_n&gt;$</th>
<th>$&lt;S_p&gt;$</th>
<th>$\phi^*&lt;S_n&gt;$</th>
<th>$&lt;S_p&gt;$</th>
<th>$\psi^*$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>1.81</td>
<td>21.9</td>
<td>39.8</td>
<td>39.6</td>
<td></td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>1.35</td>
<td>26.8</td>
<td>36.3</td>
<td>35.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>1.13</td>
<td>29.0</td>
<td>32.9</td>
<td>32.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>0.99</td>
<td>30.0</td>
<td>29.6</td>
<td>29.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>50</td>
<td>0.89</td>
<td>30.2</td>
<td>26.7</td>
<td>27.0</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Concerning the dependence on target radius, we see that $\psi^*$ decreases considerably when the radius increases. There are mainly two reasons for this behaviour. One of them is the softening of the radial neutron leakage spectrum from the target (the spectrum of the neutrons that enter into the fuel), when the target is enlarged. The probability to induce fission for the source neutrons strongly decreases with decreasing energy, especially when the core is loaded with even-neutron number actinides, as will be shown below. Lower neutron energy also inhibits other possible neutron multiplication reactions, such as $(n,xn)$-reactions and secondary spallation. The number weighted mean energy of the radial neutron leakage spectrum decreases by a factor of 4 when the target radius is increased from 20 cm to 50 cm and the fraction of neutrons above 20 MeV is only 0.5% for a radius of 50 cm, while 3.2% for a 20 cm radius. The radial neutron leakage spectra for different target radii are plotted in Fig. 2 and the fraction of leakage neutrons above some energy thresholds are listed in Table III.
Fig. 2. Radial neutron leakage spectra from the target for different target radii.

Table III
Fraction of Neutrons Leaking out Radially from the Target that have Energies above 1, 7, 20 and 150 MeV.

<table>
<thead>
<tr>
<th>Target Radius</th>
<th>&gt;1 MeV</th>
<th>&gt;7 MeV</th>
<th>&gt;20 MeV</th>
<th>&gt;150 MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>62.4%</td>
<td>13.4%</td>
<td>6.9%</td>
<td>1.1%</td>
</tr>
<tr>
<td>20</td>
<td>45.2%</td>
<td>6.1%</td>
<td>3.2%</td>
<td>0.45%</td>
</tr>
<tr>
<td>30</td>
<td>31.0%</td>
<td>3.0%</td>
<td>1.7%</td>
<td>0.22%</td>
</tr>
<tr>
<td>40</td>
<td>20.7%</td>
<td>1.6%</td>
<td>0.9%</td>
<td>0.12%</td>
</tr>
<tr>
<td>50</td>
<td>13.5%</td>
<td>0.8%</td>
<td>0.5%</td>
<td>0.07%</td>
</tr>
</tbody>
</table>

The other reason for the decrease in $\psi^*$ is that the axial target neutron leakage (the neutrons that leak out from the target axially and never enter the active core) increases significantly with increasing target radius. The fraction of axial target leakage neutrons relative to the total number of neutrons exiting the target is approximately 5% for the 20 cm radius target while about 28% for the 50 cm radius target. The major part of the axial leakage is in the backward direction, through the accelerator tube.

On the other hand, increasing the target radius increases the neutron multiplication inside the lead target, which leads to a higher number of neutrons created per source proton ($S_n/S_p$ increases from 26.8 for $r=20$ cm to 30.2 for $r=50$ cm). The multiplicative effect of $(n,xn)$-reactions and secondary spallation in the lead target enhances the proton source efficiency. Consequently, with increasing target radius, there are more neutrons for each source proton that enter into the fuel, though the efficiency of these neutrons is strongly reduced. Accordingly, we see in Table II that $\psi^*$ decreases less rapidly than $\varphi^*$ with increasing target radius, due to the increase in $S_n/S_p$. These two competing factors in $\psi^*$ are thus well represented by Eq. (3).

We conclude that, in order to optimize the proton source efficiency and the output/input power, a target radius as small as possible should be chosen (increasing the radius from 20 cm to 50 cm decreases $\psi^*$ by about 25%). These results are in good agreement with other similar studies [7]. However, it has also been shown that, reducing the target radius has some undesirable effects, for instance higher fluence/burnup ratio (lower maximum burnup) and more severe high-energy damages. Moreover, for a large-scale ADS, the target must be sufficiently large to be able to remove the heat from the high-power accelerator beam. It is thus clear that, in order to optimally design a cost-efficient ADS with high fuel performances, a trade-off between several different aspects arises.

V.A.2. A Sodium Cooled Model

Replacing the lead-bismuth coolant with sodium reduces the reactivity by about 5000 pcm, so in order to maintain a $k_{eff}$ of 0.95, the outer core radius was increased by 11 cm. As is shown in Fig. 3, the proton source efficiency for small target radii is lower for the sodium cooled core than with the lead-bismuth coolant (approximately 7% lower for $r = 20$ cm).

The main reason for this is that there is no neutron multiplication in the sodium coolant, in contrast to lead-bismuth. The differences between the two curves indicate that the contribution from $(n,xn)$-multiplication in the Pb-Bi coolant is significant for target radii smaller than about 30 cm. As long as there is a fraction of neutrons with energy higher than about 7 MeV (the $(n,2n)$-threshold in lead) there will be $(n,xn)$-neutron multiplication in lead. When the target radius is small, this high-energy fraction is rather high (6.1% have energies higher than 7 MeV for $r=20$ cm.

Fig. 3. Proton source efficiency $\psi^*$ versus target radius for a Pb-Bi cooled and a Na cooled model ($1\sigma$-error $\sim$ 0.75%).

The main reason for this is that there is no neutron multiplication in the sodium coolant, in contrast to lead-bismuth. The differences between the two curves indicate that the contribution from $(n,xn)$-multiplication in the Pb-Bi coolant is significant for target radii smaller than about 30 cm. As long as there is a fraction of neutrons with energy higher than about 7 MeV (the $(n,2n)$-threshold in lead) there will be $(n,xn)$-neutron multiplication in lead. When the target radius is small, this high-energy fraction is rather high (6.1% have energies higher than 7 MeV for $r=20$ cm.
compared to 0.8% for r=50 cm). When the target radius increases, the fraction of high-energy neutrons decreases, and at radii above 40 cm the $\psi^*$ values are essentially the same. Another difference between the two coolant materials is that lead-bismuth has better spallation-neutron production characteristics. This enhances $\psi^*$ for small target radii, in which cases there are still a significant fraction of very energetic neutrons leaking out into the fuel region. The difference between the Pb-Bi and the Na coolant are further illustrated in Fig. 4, where the neutron source efficiencies are depicted, together with the number of neutrons per source proton.

![Fig. 4. Neutron source efficiency $\psi^*$ (according to the target neutron leakage definition) and number of neutrons per source proton ($S_n/S_p$), versus target radius for the Pb-Bi cooled and the Na cooled model (1σ-error ∼ 0.75 %).](image)

V.B. $\psi^*$ as function of Axial Beam Impact Position

The proton source efficiency has been studied as a function of the axial beam impact position, varying from the center of the core ($z = 0$) to the top of the core ($z = 50$ cm). It is seen in Fig. 5 that, for the reference model (with an accelerator tube radius of 15 cm) the maximum in $\psi^*$ is obtained at about 20 cm above the core center. Moving away from the optimum impact position, $\psi^*$ decreases, due to increasing axial neutron leakage from the target.

![Fig. 5. Proton source efficiency $\psi^*$ versus axial beam impact position (cm above the core center) for a tube radius of 15 cm and of 10 cm (1σ-error ∼ 0.50 %).](image)

As is shown in Fig. 5, $\psi^*$ and its dependence of the axial beam impact is rather sensitive to the accelerator tube radius. For a tube radius of 10 cm, the beam impact position that maximizes $\psi^*$ is moved downwards in the core, with the optimum at about 13 cm above the core center. The maximum value of $\psi^*$ also increases somewhat (~3%) when the tube radius is changed from 15 cm to 10 cm.

When the tube radius decreases there are mainly two effects appearing. One is that the axial neutron leakage in the backward direction decreases (increasing $\psi^*$) and the other one is that the energy spectrum of the neutrons entering into the fuel is softened (decreasing $\psi^*$). When the beam impact is close to the core center, the impact of the leakage on the neutron balance is more pronounced than the softening of the neutron spectrum. On the contrary, when the beam impact is close to the top of the core, the softening effect is dominant and $\psi^*$ is lower for the 10 cm tube radius.

An accidental scenario that has been considered in an ADS is that the accelerator tube is filled (partially or fully) with the target material [18], which would increase $k_{\text{eff}}$. For the reference model, filling the tube from 20 cm above the core center till the top of the core, this increase is about 600 pcm. For an initial reactivity level of $k_{\text{eff}} = 0.95$, and not taking the variations in $\psi^*$ into account, this would increase the core power by 14%. For the 10 cm tube radius, the change in $k_{\text{eff}}$ is smaller and also the effect on the power. However, the rapid decrease in $\psi^*$ above $z = 20$ cm will tend to reduce the core power. Adding these two opposite effects together, according to Eq. (6), it is found that the core power decreases when the tube channel is filled with lead-bismuth coolant. Filling the tube from $z = 20$ cm to $z = 50$ cm decreases the core power by 7% for the 15 cm tube radius and by 20% for the 10 cm tube radius.
However, if the reactor is operating at a reactivity level closer to criticality, the effect on the core power from a reactivity insertion will be higher and might be more important than the decrease in $\psi^*$. One way to increase the safety margins to prompt criticality, is to set the proton beam impact at the top of the core ($z = 50$ cm). However, the loss in proton source efficiency, by moving the impact from the optimal position to the top of the core would be large - about 18% for the 15 cm tube radius and 24% for the 10 cm tube radius.

V.C. $\psi^*$ as function of Proton Beam Energy

The proton source efficiency was calculated for different proton beam energies, varying from 400 MeV to 2.5 GeV, for a 20 cm and a 50 cm radius target. The $\psi^*$ values divided by the proton energy ($\psi^*/E_p$) are displayed in Fig. 6.

![Proton Source Efficiency](image)

Fig. 6. Proton source efficiency divided by proton energy in GeV ($\psi^*/E_p$) versus proton beam energy, for target radii of 20 cm and 50 cm (1σ-error ~ 0.50 %). Energy gain $G$ from CERN experiments [19] for a set-up configuration of $k_{eff} = 0.895$.

In coherence with Fig. 3, $\psi^*$ is higher for the 20 cm radius than for the 50 cm radius, for all proton energies. However, the shape of the two curves is very similar. $\psi^*/E_p$ increases strongly with increasing proton energy up to about 1000 MeV. The maximum is reached at about 1200-1400 MeV. Above 1400 MeV the curves decrease slightly. The reason for this is that the range of the protons increases with increasing energy and a larger fraction of the spallation induced neutrons will be created far away from the center of the core (or even below the core). This increases the axial neutron leakage from the target in the downward direction. For instance, the range of 1 GeV protons impinging on a lead target is about 53 cm, while about 95 cm for 2 GeV protons [18]. The distance from the top of the target to the bottom of the core was 75 cm (Fig. 1).

The proton source efficiency divided by the proton energy ($\psi^*/E_p$) is closely related to the energy gain (beam power amplification) of a source driven sub-critical system. The energy gain ($G$), as defined in [19], represents the total power produced in the core over the accelerator power

$$G = \frac{P_{tot}}{P_{acc}} = \frac{G_0}{1 - k_{eff}}$$  \hspace{1cm} (7)

where $G_0$ relates to the efficiency of the spallation regime. Similarly, using Eq. (6) and the fact that $P_{acc} = < S_p > \cdot E_p$, $P_{tot}/P_{acc}$ can be expressed as

$$\frac{P_{tot}}{P_{acc}} = \frac{P_{tot}}{< S_p > \cdot E_p} \propto \frac{k_{eff}}{1 - k_{eff}} \left( \frac{\psi^*}{E_p} \right)$$  \hspace{1cm} (8)

Hence, for given $k_{eff}$, $\psi^*/E_p$ is proportional to the energy gain $G$. In the experiments performed at CERN [19], the energy gain was studied as function of the proton beam energy. The results are re-plotted in Fig. 6. We see in the figure that the MCNPX simulated values of $\psi^*$ have similar relative dependence on the proton energy, as the results from the CERN experiments. This comparison confirms that the $\psi^*$ parameter represents well the beam power amplification of an ADS.

The $k_{eff}$ of the sub-critical assembly in the CERN experiments was 0.895. In order to obtain $G$ for $k_{eff} = 0.95$, the values have to be multiplied by $(1 - 0.895)/(1 - 0.95) = 2.1$. For $E_p = 1000$ MeV the energy gain becomes 61.

V.D. $\psi^*$ for an Americium Based Fuel

Finally, $\psi^*$ has been studied for an americium based fuel (40% Pu and 60% Am), as a function of the target radius. The volume fraction of the ZrN matrix was re-adjusted to 63%, in order to keep a $k_{eff}$ of about 0.95. The results are displayed, together with the results for the reference model, in Fig. 7. We see that the introduction of americium decreases the source efficiency significantly and makes it more sensitive to the target radius.
This is expected, since the fission cross-section of Am-241 and Am-243 decreases rapidly for neutron energies below 1 MeV. Fig. 8, where the fission cross-section over the absorption cross section is plotted for Am-241 and Pu-239, shows that the fission probability of Am-241 is very sensitive to neutron energies between 0.1 and 1 MeV, whereas for Pu-239, it is high everywhere in this energy range. Above 1 MeV the fission probability for plutonium and americium are of the same order of magnitude. The other even-neutron number actinides, Pu-238, Pu-240, Pu-242 and Am-243, have similar cross-section dependence as Am-241, while the Pu-241 cross-section is similar to that of Pu-239.

Hence, the neutrons entering into the fuel with energy below 1 MeV will have less probability to induce fission in the fuel, when the core is loaded mainly with americium. As was shown in Fig. 2 and in Table III, the energy distribution of the radial target leakage neutrons is very sensitive to the target radius. For a 10 cm radius, only 38% of the leakage neutrons are below 1 MeV and $\psi^*$ is not so sensitive to the americium/plutonium ratio, which is also seen in Fig. 7. However, with increasing target radius, the fraction of neutrons above 1 MeV decreases rapidly, with a corresponding loss in proton source efficiency. Thus, increasing the americium concentration from 20% to 60%, reduces $\psi^*$ by 11% for the 20 cm target radius and by 29% for the 50 cm target radius.

In Section III.B, it was shown that, for a fixed fuel composition, $\psi^*$ is approximately proportional to the core power divided by the source intensity. However, increasing the fraction of americium in the fuel increases $\nu$ by about 5%. Therefore, normalising to the same core power, the difference between the two curves would be slightly larger (by 5%) than in Fig. 7.

VI. CONCLUSIONS

Instead of using the neutron source efficiency parameter $\phi^*$, in order to study the beam power amplification, a new parameter $\psi^*$, representing the efficiency of the source protons, has been introduced. The proton source efficiency $\psi^*$ is related to the number of fissions produced in the system, which in turn is approximately proportional to the total core power. By introducing $\psi^*$, ambiguities in defining the external neutron source when calculating the source efficiency are avoided. Another advantages with $\psi^*$ over $\phi^*$ is that it is proportional to the core power divided by the proton beam power, independently of the neutron source distribution. Maximizing the proton source efficiency minimizes the proton current needs and relaxes the constraints on the construction of the high-power accelerator. Studying $\psi^*$ when optimising different system parameters therefore becomes an important factor in the overall design of an ADS.

Studying the radius of the spallation target, it was found that $\psi^*$ decreases strongly with increasing target radius (by 25% when the radius is changed from 20 cm to 50 cm). The two main reasons for this behaviour is that, the axial neutron leakage from the target increases and that the energy spectrum of the neutrons that enter into the fuel is softened, when the target is enlarged. These effects are only partly compensated by the increased neutron multiplication inside the target. Hence, in order to maximize the beam power amplification, a target radius as small as possible should be chosen, without exceeding the limits determined by safety constraints and other target-core characteristics (e.g. high-energy particle fuel damages and beam power heat removal). Substituting the lead-bismuth coolant by sodium decreases $\psi^*$ for target radii smaller than 30 cm, whereas
for larger target radii, the two coolant options perform equally good.

The axial position of the proton beam impact that maximizes $\psi^*$ was, for the reference model (tube radius = 15 cm), located approximately 20 cm above the core centre, but with relatively small variations between 0 and about 35 cm. However, the dependence of $\psi^*$ on the impact position is sensitive to the accelerator tube radius, and for a tube radius of 10 cm, the maximum was found at about 13 cm above the core centre. Reducing the tube radius from 15 to 10 cm also increases the maximum of $\psi^*$ by 3%.

Investigating the proton source efficiency divided by the proton energy ($\psi^*/E_p$) as a function of the proton energy showed that a maximal accelerator power amplification is obtained for proton energies of about 1200 to 1400 MeV, but with rather small changes between 1000 and 2000 MeV. Finally, increasing the americium content in the fuel from 20% to 60%, decreases $\psi^*$ considerably, especially for larger target radii. Due to the sharp decrease in fission cross-section below 1 MeV, americium is more sensitive than plutonium to the softening of the energy spectrum of the neutrons that enter into the fuel.

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Proton Source Efficiency for Inert Matrix Fuels in Accelerator Driven Systems

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Abstract – In order to study the beam power amplification of an accelerator-driven system (ADS), a new parameter, the proton source efficiency ($\psi^*$) was introduced in a previous study. $\psi^*$ represents the average importance of the external proton source, relative to the average importance of the eigenmode production, and is closely related to the neutron source efficiency ($\phi^*$), which is frequently used in the ADS field. The main advantage with using $\psi^*$ instead of $\phi^*$ for ADS is that the way of defining the external source is unique and that it is proportional to the core power divided by the proton beam power, independently of the neutron source distribution.

It has been shown that the source efficiency can vary considerably for different reactor core systems. Studying $\psi^*$ for different system parameters is therefore of interest when designing an ADS. In this paper, numerical simulations have been performed with the Monte Carlo code MCNPX in order to study $\psi^*$ as a function of spallation target radius for different inert matrix fuels. It was found that, in order to maximize $\psi^*$, and thereby minimizing the proton current needs, a target radius as small as possible should be chosen. A ZrN or an YN matrix, mixed with the plutonium and americium mixed nitride fuel, appears to be a slightly better choice than a HfN matrix, considering only the proton source efficiency. It was also found that a power flattened double-zone core, compared to a single-zone core, decreases $\psi^*$ by about 5% for the ZrN matrix and by about 10% for the HfN matrix.

I. INTRODUCTION

Accelerator-Driven Systems (ADS) are being investigated as a possible mean for reducing the long-term radiotoxicity of spent reactor fuel. These systems allow for a much higher concentration of minor actinides than what is acceptable in critical reactors.

Optimizing the output/input power, without exceeding the limits determined by safety constraints and other target-core characteristics can play an important role in the overall design of an ADS and on the economy of its operation. The neutron source efficiency parameter $\phi^*$ is commonly used to study this quantity, since it is related to the number of fissions produced in the core by an average external source neutron. However, calculating $\phi^*$ for an accelerator-driven system involves some complications, since the actual source particles are protons, and not neutrons. With the motivation of simplifying the concept of source efficiency, we have introduced in a previous paper a new parameter $\psi^*$, which refers to the number of fissions produced in the core by each source proton. The advantages with using $\psi^*$ instead of $\phi^*$ is that there is no ambiguity in how to define the external source, and that it is proportional to the beam power amplification, without the need of a weighting factor.

In order to guarantee the stability of uranium free fuels at high temperatures, the use of inert matrices is foreseen. Different safety parameters of several possible inert matrix fuels have been studied. In the present paper, three inert matrices (ZrN, YN and HfN), mixed with a plutonium and americium mixed nitride fuel, have been investigated in terms of $\psi^*$. In Section II, the neutron source efficiency is first defined and discussed and then the proton source efficiency parameter is introduced. Section III describes the lead-bismuth cooled ADS modelled in this study. In Section IV.A, $\psi^*$ is studied as a function of target radius for the different matrices, while in Section IV.B, $\psi^*$ has been compared for a single-zone core and a power flattened double-zone core.

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II. SOURCE EFFICIENCY $\phi^*$

II.A. Definition of the Neutron Source Efficiency

The neutron flux distribution $\phi_s$ in a sub-critical core is the solution to the inhomogeneous steady-state neutron transport equation

$$ A\phi_s = F\phi_s + S_n $$

(1)

where $F$ is the fission production operator, $A$ is the net neutron loss operator and $S_n$ is the external neutron source.

The neutron source efficiency $\phi^*$, usually denoted $\phi^*$, represents the efficiency of the external source neutrons and can be expressed according to the following equation:

$$ \phi^* = \left( \frac{1}{k_{eff}} - 1 \right) \frac{<F\phi_s>}{<S_n>} $$

(2)

which is valid in the range $0 < k_{eff} < 1$. $<F\phi_s>$ is the total production of neutrons by fission and $<S_n>$ is the total production of neutrons by the external source. Eq. (2) shows that, for given values of $k_{eff}$ and $<S_n>$, the larger $\phi^*$ the larger the fission power produced in the system.

II.B. Introduction of the Proton Source Efficiency

A new parameter, called “proton source efficiency” and denoted $\psi^*$, which represents the product of $\phi^*$ and the number of source neutrons generated per source proton ($S_n/S_p$), has been introduced. We have the following relation between the proton source efficiency $\psi^*$ and the neutron source efficiency $\phi^*$:

$$ \psi^* = \phi^* \cdot \frac{<S_n>}{<S_p>} $$

(3)

For a fixed system (constant $S_n/S_p$), we see that $\psi^*$ is proportional to $\phi^*$. However, when studying a change in a system design, $S_n/S_p$ might also change. In this case, $\psi^*$, but not $\phi^*$, is proportional to the output/input power. The $\psi^*$ parameter could also, in analogy with $\phi^*$, be expressed in terms of $k_{eff}$ and the total number of neutrons produced by fission in the core, for each source proton. Inserting Eq. (2) in Eq. (3) it is expressed in the same way as $\phi^*$, only with the replacement of $S_n$ by $S_p$,

$$ \psi^* = \left( \frac{1}{k_{eff}} - 1 \right) \frac{<F\phi_s>}{<S_p>} $$

(4)

$<F\phi_s>/<S_p>$ is the total production of neutrons by fission over the total number of source protons.

III. SYSTEM MODELING

A homogenized model of a uranium free nitride fuelled and lead-bismuth cooled ADS was used. Simulations have been performed for two different fuel compositions (curium free), with the ratio of plutonium to americium set to 80/20 and 40/60. For all matrix-fuel combinations, the fraction of inert matrix was adjusted in order to obtain a $k_{eff}$ of about 0.95. The volume fractions of the different matrix fuels are displayed in Table I. We note that the plutonium based fuels enable a much larger fraction of inert matrix than the americium based ones.

<table>
<thead>
<tr>
<th>Matrix</th>
<th>Volume fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu/Am = 80/20</td>
<td>Pu/Am = 40/60</td>
</tr>
<tr>
<td>ZrN</td>
<td>82.9%</td>
</tr>
<tr>
<td>YN</td>
<td>83.4%</td>
</tr>
<tr>
<td>HfN</td>
<td>67.0%</td>
</tr>
</tbody>
</table>

The height of the active core of the reference model, depicted in Fig. 1, is 100 cm and the outer radius is 70 cm. The inner radius is 20 cm, which is also the boundary of the lead-bismuth target. The accelerator tube has a radius of 15 cm and the axial position of the proton beam impact is 25 cm below the top of the core. The radius of the radially uniform 1000 MeV proton beam is 7.5 cm. Above and below the active zone of the core, plena for accommodation of gas release are included, having lengths of 100 and 50 cm, respectively. The radial reflector is assumed to consist of 90% steel and 10% lead-bismuth.
IV. THE PROTON SOURCE EFFICIENCY FOR DIFFERENT INERT MATRIX FUELS

The proton source efficiency $\psi^*$ has been studied for different inert matrix fuels. The starting point for each parameter study is the reference model, described in the previous section.

IV.A. $\psi^*$ as function of Target Radius for different Inert Matrix Fuels

In this section, $\psi^*$ has been computed as a function of target radius for the three different inert matrix fuels. Each time the target radius was changed, $k_{\text{eff}}$ was kept constant at 0.95 by adjusting the outer radius of the core. We see in Fig. 2, where the results for the plutonium based fuel have been plotted, that $\psi^*$ decreases considerably when the target radius increases. There are mainly two reasons for this behaviour. One of them is the softening of the radial neutron leakage spectrum from the target, when the target is enlarged. The probability to induce fission for the neutrons entering the active core strongly decreases with decreasing energy, especially when the core is loaded with even-neutron number actinides. The other reason for the decrease in $\psi^*$ is that the axial target neutron leakage increases with increasing target radius. The major part of the axial leakage is in the backward direction, through the accelerator tube.

Comparing the different matrix fuels, it is also shown that, for the ZrN and the YN matrices, $\psi^*$ has similar dependence on the target radius. This is expected, since Zr and Y have similar neutron cross-sections and similar mass numbers. However, for the HfN matrix fuel, $\psi^*$ decreases faster with increasing target radius. The reason for this is that Hf has higher absorption and scattering cross-sections for low-energy neutrons (below a few keV), and therefore it is more sensitive to the softening of the neutron energy spectrum. This effect increases with increasing target radius, as the energy spectrum of the neutrons leaking out from the target is softer for larger target radii. For instance, for the 10 cm target radius, the $\psi^*$ values for the different matrices are very similar, since only a very small fraction of the target leakage neutrons have energy of the order of a few keV or lower. On the other hand, for the 50 cm radius, $\psi^*$ is lower by about 23% for the HfN matrix than for the ZrN and YN
matrices, due to the much softer target neutron leakage spectrum.

For the americium based fuel (Fig. 3), there are mainly two different effects arising, compared to the plutonium based fuel. Firstly, the decrease in $\psi^*$ with increasing target radius is faster, due to lower fission probability (mainly for neutrons below $\sim 1$ MeV) for the americium isotopes than for the plutonium isotopes.\(^5\) Secondly, the difference between the HfN matrix fuel and the two other fuels is smaller ($\sim 13\%$ for the 50 cm target radius). The explanation for this is that the fractions of matrix are lower and consequently a switch of matrix has smaller impact on $\psi^*$.

**Fig. 3.** $\psi^*$ versus target radius for different americium based matrix fuels (40% Pu + 60% Am).

We conclude that, in order to optimize the proton source efficiency and the output/input power, a HfN matrix fuel is slightly less favorable than a ZrN or a YN matrix fuel, in particular for large target radii. However, a HfN matrix is desirable for other reasons, e.g. its high melting point and the hard spectrum it induces, which leads to less production of curium. Therefore, HfN might still be a good choice of inert matrix, despite the loss in proton source efficiency, in particular if an americium based fuel with relatively small target radius is used.

**IV.B. $\psi^*$ for a Single-zone Core compared to a Power Flattened Double-zone Core.**

The proton source efficiency $\psi^*$ has been studied for the reference model (single-zone core), compared to a model where the core has been divided into two zones with different matrix fractions, with the purpose of lowering the power peaking. The single-zone and the double-zone power densities, calculated for the plutonium based ZrN matrix fuel, are depicted in Fig. 4. For the double-zone core, the matrix fractions have been adjusted in order to obtain the same maximum power density for the two zones.

**Fig. 4.** Power density profiles for a single-zone core and a power flattened double-zone core, calculated for the plutonium based ZrN matrix fuel. Target radius = 20 cm.

The comparison of $\psi^*$ for the single-zone core and the double-zone core has been performed for the plutonium based and the americium based fuel, dispersed with either ZrN or HfN matrices. The calculations were performed for a target radius of 20 cm and the results are listed in Table II. We see that $\psi^*$ decreases for the power flattened double-zone core, by about 5-6% for the ZrN matrix fuels and about 10-11% for the HfN matrix fuels. If a double-zone core is assumed, the loss in $\psi^*$ by substituting the ZrN matrix by a HfN matrix is approximately 12% for the plutonium based fuel and 8% for the americium based one. Hence, a trade-off arises, where one has to consider whether the advantages with a power flattened core or the wish of using a HfN matrix fuel is worth the loss in proton source efficiency.

**TABLE II**

$\psi^*$ calculated for a Single-zone Core (1Z) and a Double-zone Core (2Z). Target radius = 20 cm.

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Matrix</th>
<th>1Z</th>
<th>2Z</th>
<th>Diff.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu/Am = 80/20</td>
<td>ZrN</td>
<td>36.0</td>
<td>34.3</td>
<td>-4.8%</td>
</tr>
<tr>
<td></td>
<td>HfN</td>
<td>34.0</td>
<td>30.3</td>
<td>-10.7%</td>
</tr>
<tr>
<td>Pu/Am = 40/60</td>
<td>ZrN</td>
<td>32.0</td>
<td>30.1</td>
<td>-5.9%</td>
</tr>
<tr>
<td></td>
<td>HfN</td>
<td>30.7</td>
<td>27.6</td>
<td>-9.9%</td>
</tr>
</tbody>
</table>
There are mainly two effects arising when the double-zone is introduced and the content of matrix in zone 1 increases. Firstly, the absorption and slowing-down of the neutrons increases, in particular for low-energy neutrons. For the studied 20 cm radius target model, the energy spectrum of the neutrons entering into the fuel is softer than the average fission neutron spectrum in the core. Therefore, since the source efficiency ($\psi^*$ or $\phi^*$) relates the ratio between the source neutrons and the fission neutrons, the increase of the matrix fraction has a larger impact on the source neutrons than on the fission neutrons. Consequently, $\psi^*$ decreases. As Hf has higher absorption cross-sections for low-energy neutrons than Zr, this effect on $\psi^*$ is stronger for the HfN matrix, as is seen in Table II. It also increases with increasing target radius, since the energy of the neutrons that leak out from the target decreases with increasing target radius, while the average fission neutron energy is unchanged.

Secondly, increasing the matrix content in zone 1 suppresses the (n,2n)-multiplication in the inner parts of the active core, which will also tend to decrease $\psi^*$. This effect is strong for small radii when the target neutron leakage spectrum is hard, but decreases as the target radius increases.

V. CONCLUSIONS

Instead of using the neutron source efficiency parameter $\phi^*$, in order to study the beam power amplification, a new parameter $\psi^*$, representing the efficiency of the source protons, has been introduced in a previous study. Maximizing the proton source efficiency minimizes the proton current needs and relaxes the constraints on the construction of the high-power accelerator. Studying $\psi^*$ when optimising different system parameters therefore becomes an important factor in the overall design of an ADS.

It has been shown that the proton source efficiency decreases significantly with increasing target radius. Thus, in order to maximize $\psi^*$ and the output/input power within the safety limits, a target radius as small as possible should be chosen.

Moreover, it was found that the HfN matrix fuel yields a lower $\psi^*$ than the ZrN and the YN matrix fuels. However, for the americium based fuel and in particular for small target radii, the difference is relatively small. Due to other favourable properties of HfN, it is therefore still an interesting option of inert matrix material, despite the loss in proton source efficiency.

It was also found that $\psi^*$ is lower for a power flattened double-zone core, compared to a single-zone core. The differences are about 5% for the ZrN matrix fuels, while about 10% for the HfN matrix fuels. Comparing the ZrN matrix with the HfN matrix, assuming a double-zone core, the difference in $\psi^*$ is larger for the plutonium based fuel (∼12%) than for the americium based fuel (∼8%).

We conclude that, when designing an ADS, there is a trade-off arising between several different aspects. Various parameters, such as optimal target radius, power flattening and choice of inert matrix material together with other target-core characteristics and the various safety limitations, have to be weighted against the advantage of optimizing $\psi^*$ and minimizing the proton current needs.

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