ACHIEVABLE TRANSMUTATION RATES FOR TRUS AND LLFPS IN MYRRHA

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Abstract

SCK•CEN wants to fulfil a prominent role in the ADS field and is currently establishing a R&D programme to finalise the design of an ADS prototype. Therefore, the ADONIS system, initially intended for dedicated radioisotope production, was revisited to give birth to the Myrrha system. The need for ADS related R&D as well as the extension of current competencies within SCK•CEN and related institutes led to the study of a prototype ADS which would focus primarily on ADS related research, i.e. on materials and fuel research, on the utilisation of liquid metals and associated aspects, on reactor physics, and subsequently on applications such as transmutation and safety research on sub-critical systems. In this respect, the Myrrha system should become a new major research infrastructure for SCK•CEN supporting and enabling the international R&D programmes.

Currently, the study and preliminary conceptual design of the Myrrha system is being finalised and the basic engineering phase has started. This study will also define the final choice of the characteristics of the facility depending on the selected fields of application to be achieved in this machine. The applications which are considered can be grouped in three blocks; i) continuation, and later on extension towards ADS, of the ongoing R&D programmes in the field of reactor materials, fuel and reactor physics research; ii) enhancement and triggering of new R&D activities such as waste transmutation, ADS technology, liquid metal embrittlement; iii) initiation of new competencies such as medical application (proton therapy, PET production,..), neutron beam applications.

Myrrha will use an accelerator delivering a proton beam of (2 mA, 250 or 350 MeV). The spallation target is a liquid Pb-Bi volume circulating in a single closed loop. In its present design the spallation source is a windowless design. The sub-critical assembly is made of two consecutive zones of fuel rod lattices. Due to the main interest in research on the transmutation of TRUs, the first zone, consisting of MOX-type fuel rods with high Pu contents, forms a fast neutron spectrum region. In the present design, the fuel pins are inserted in a lead environment. This lead environment is either a solid block (cooled with a gas-circuit) or liquid lead, depending on the total power to be removed. The fast neutron zone is surrounded by a thermal neutron zone made of LWR UO_2 fuel rods inserted in a water moderator. This thermal zone will allow to increase the multiplication factor to perform experiments related to LWR fuel research, radioisotope production and transmutation of LLFPs.

In this paper we will report on the performances one can achieve in the present design of Myrrha concerning transmutation of TRUs such as Np, Pu, Am, and Cm in the fast neutron zone as well as of LLFPs such as ^{129}I, ^{99}Tc, ^{135}Cs, and ^{93}Zr in the thermal neutron zone.

Introduction

Nuclear energy has to cope with some topics to resolve the economical question of increasing energy demand and more specially the public acceptability requirements:

- Increasing the absolute safety of the installations.
- Managing more efficiently the nuclear waste.

In that respect, the development of a new type of nuclear installation coping with the above constraints as well as with those of technological, social and economical nature is most important for the future of sustainable energy provision. Accelerator driven systems are coping with the above constraints and can pave the way to a more environmentally safe and acceptable nuclear energy production. Fundamental and applied R&D are crucial in the development of these technologies and demand the availability of appropriate prototype installations. These prototype installations have to enable and have to deal with these R&D-issues related to accelerator driven system development.

The Myrrha project aims at investigating the design, development and realisation of a versatile neutron source based on an accelerator driven system. It focuses on the realisation of a radiation source, well-matched to both regional R&D needs and international fundamental research programmes in the field of accelerator driven systems.

The initial project leading to the current Myrrha project focused on the dedicated application of radioisotope production relying on an accelerator driven system. This project, called ADONIS, has been shifted to the Myrrha project to extend the scope of applications towards :

- material irradiation studies;
- fuel research (transient and high burn-up accumulation);
- radioisotope production;
- waste transmutation studies; and,
- ADS system prototyping from the technological point of view.

These studies lead to refocusing the scale of the system for both the sub-critical assembly and the accelerator performances to be considered.

Present Design of Myrrha

Accelerator

The accelerator considered up to now is a six sector cyclotron generating a 2 mA current at 250 or 350 MeV considered in two stages, the first stage being an injector ranging between 40 and 70 MeV. The positive ion (H^+) acceleration technology will be used for this machine. IBA is considered as the most potential partner for the design of the accelerator. The two stage accelerator option is not yet a frozen option: it is kept for potential applications with low energy protons such as radioisotope production using protons or, to a limited extent, proton therapy.

Spallation target

The spallation target is made of liquid Pb-Bi circulating in a double concentric cylindrical circuit with a dump tank at the lower end of the circuit. At the upper part of the target system a free surface is in contact with the incoming proton beam. No conventional window is foreseen between the Pb-Bi free surface and the beam in order to keep the energy losses at their minimum. The Pb-Bi is circulating bottom-top in the outer tube and going down in the central tube leading to the creation of a jet pump effect which will help in trapping the Pb and Po vapours. However, to reduce potential transport of volatile substances from the spallation source into the accelerator and related damage, a very thin He-cooled double walled barrier is placed in the beam trajectory at a certain distance from the spallation source.

The choice of a windowless design has been influenced by the following considerations :

- With 250 MeV, an incident proton delivers 7 MeV kinetic energy per spallation neutron. Almost 85% of the incident energy is lost as « evaporation » energy of the nuclei in the target. The addition of a window would only diminish the fraction of the incident energy delivered to the spallation neutrons.
- A windowless design avoids vulnerable parts in the concept, increasing its reliability.
- The low working temperature in the Pb-Bi eutectic. Indeed, if we consider an inlet temperature of 150°C, we end up with working temperatures of 250°C average temperature in the central part and 170°C in the outer part, with a circulating speed of 1.8 m/s in both sections. These low temperatures will result in very low Pb-Po evaporation rates (within the range of 2. $10^{-8} \sim 10^{-7}$ kg/m².s).

The thermo-hydraulics simulation of the windowless design is foreseen in a first stage to be carried out with hot water which is a good equivalent fluid for the Pb-Bi from the comparison of their dimensionless numbers (Reynolds, Prandtl, Weber)

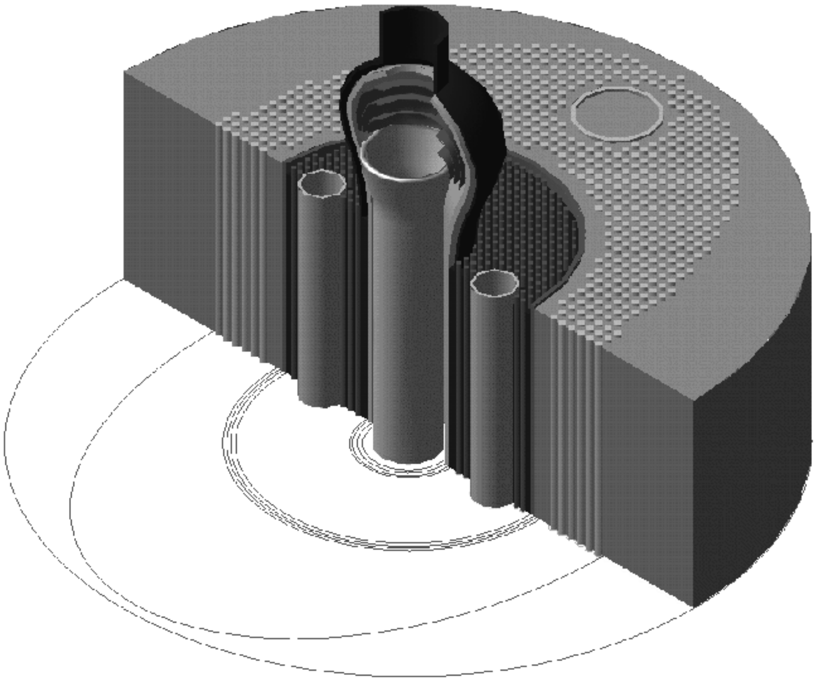
Sub-critical system

The design of the sub-critical assembly to be set up around the spallation source is application-dependent. Indeed, one should meet the neutronic performances as well as the volumes needed by the considered applications. The present design of the Myrrha sub-critical system is shown in Figure 1 below.

In that respect, to meet our goals of material studies, fuel behaviour studies, radioisotopes production, transmutation of MAs and LLFPs studies, we came to the conclusion that the sub-critical system of Myrrha should have two spectral zones ; a fast neutron spectrum zone and a thermal spectrum one. We should have irradiation channels having sufficiently large volumes for housing irradiation rigs such as the ones used in MTRs. Therefore, we should have a spallation source with axial distribution leading to a reasonable axial active length. This resulted in choosing a higher incident proton energy: 350 MeV instead of 250 MeV leading to a spallation-source axial distribution of 15 cm instead of 10 cm.

Besides these considerations, our objectives during this pre-design were to avoid the use of revolutionary technologies from the point of view of the components to be used. The targeted K_{source} value is 0.9: this value could be increased after having performed the necessary safety studies. The resulting thermal power of the sub-critical system is within 25 to 30 MW_{th}. The maximum power density estimated in the fast zone fuel pins is around 180 W/cm, whereas in the thermal zone the value does not exceed 150 W/cm.

Figure 1. **3-D view of the spallation source and the sub-critical system of Myrrha**



Fast zone description

The fast zone is made of MOX FBR-type fuel pins with a Pu content ranging between 20 and 30%, arranged in a square lattice with a 1 cm pitch. The fuel cladding is in stainless steel. The active fuel length is 50 cm. The coolant presently considered is liquid Pb or Pb-Bi or a solid Pb matrix cooled by a circulating gas. This will depend on the total amount of power to be extracted from this zone. In the present configuration, we have foreseen two irradiation channels with an inner diameter of 48 mm, where the radial form factor is nearly 1.0 and the axial form factor is 1.2 for 50 cm length.

Thermal zone description

The thermal zone is made of UO_2 PWR type fuel pins with 4% ^{235}U enrichment cladded with stainless steel. They are arranged in a typical 1.26 cm PWR square lattice. The active fuel length is 50 cm. The fuel pins are arranged presently in a light water pool. Two larger irradiation channels, with 84 mm inner diameter, are considered. Other irradiation channels can be easily accommodated as this zone is large enough.

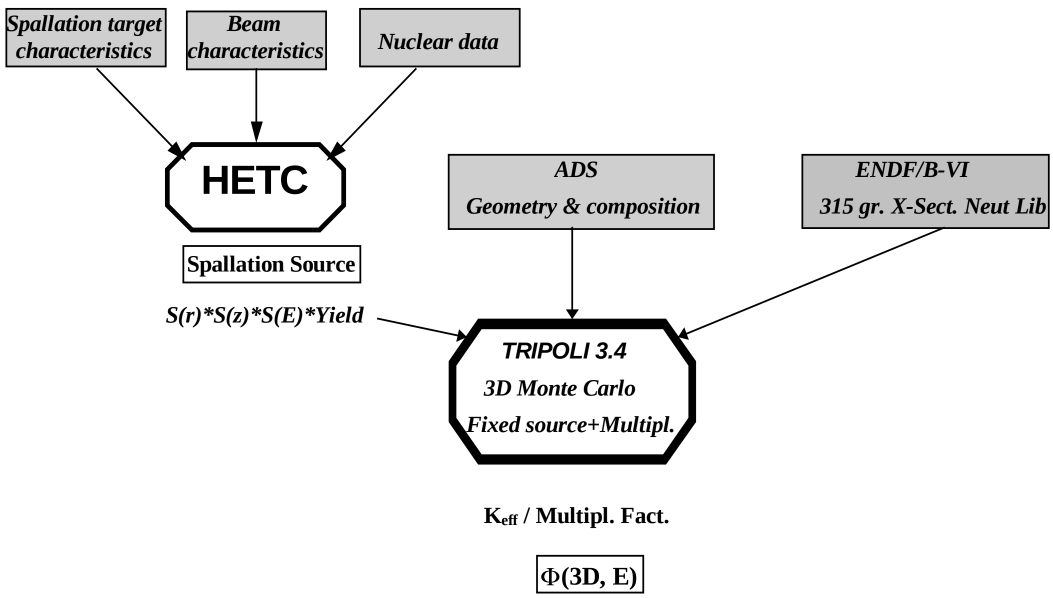
Neutronic performances of Myrrha

Based on our objectives in the project (fast and thermal flux optimisation in the sub-critical system) and our experience in the field of reactor physics, we decided at SCK•CEN to couple the HETC part of the HERMES Program System [1] high energy particle code to the S_N DORT/TORT [2], and to the Monte Carlo TRIPOLI [3] neutron transport codes. The generic calculations are performed using the DORT/TORT code whereas the TRIPOLI Monte Carlo code is used for more precise results as nearly no geometry simplifications are made thanks to the precise geometry description allowed by Monte Carlo codes. The latest resulting calculational scheme as applied in the Myrrha project is summarised in Figure 2.

The HETC code is used to compute the space and energy distribution of the primary spallation neutron source, also including all other particles involved. The high energy cascade is calculated down to 20 MeV neutrons, whereas the neutrons below this energy limit are stored as primary particles (without any interaction in the spallation medium) in the 315 multigroup energy structure of the low energy cross-section neutron library to be used with the TRIPOLI code and will be treated as a fixed neutron source in the Monte Carlo transport code.

A full detailed modelling of the sub-critical system with a heterogeneous representation of the fuel pins (fuel, cladding and coolant around) as well as of the spallation target was introduced into the TRIPOLI code. The code was run in a fixed source (in the spallation target volume) mode with possibility of having multiplication in the sub-critical system. Tallies were defined at various locations in the Myrrha system to record the fast (> 1 MeV or > 0.1 MeV) and thermal (< 0.5 eV) fluxes.

Figure 2 **Computational scheme used for the modelling of Myrrha**



The calculated values are summarised in Table 1 where we are giving also the very well known values of the BR2 materials testing reactor in different irradiation channels.

Table 1 **Comparison of the thermal and fast neutron fluxes in Myrrha and in the BR2 MTR**

Irradiation position	Myrrha Irradiation Positions 1st Fuel FZ	Irr. Chan. FZ	last Fuel FZ	1st Fuel ThZ	Irr. Chan. ThZ	BR2 Irradiation Channels Core Centre	Core Periphery	Reflector
Active Length (cm)	50	50	50	50	50	76.2	76.2	76.2
Diameter (cm)	1	4.8	1	1	8.4	8.4	8.4	8.4
Axial Form Factor	1.2	1.2	1.2	1.2	1.2	1.4	1.4	1.4
$\phi_{> 1\ MeV}$	9.5	2.3	5.4	7.2	1.2	2~3	0.2~0.4	0.03~0.2
$\phi_{> 0.1\ MeV}$	25.1	6.8	13.3	16.1	2.2	4~6	0.5~1	0.1~0.5
$\phi_{> 0.1} / \phi_{> 1}$	2.6	2.9	2.5	2.2	1.8	~2	~2.5	~3
$\phi_{Thermal}$	0.003	0.003	0.13	3.8	34.8	2~5	1~3	1~2

The Flux values given in Table 1 correspond to a K_{source} of 0.85, a proton current of 2 mA and a spallation yield of 2 neutrons/proton (corresponding to the calculated value for protons of 250 MeV). The flux values should be read $\times 10^{14}$n/cm².s.The locations assessed in the Myrrha system correspond to:

- The first fuel pin in the fast zone, the closest to the spallation source.
- The irradiation channel in the fast zone.
- The last fuel pin in the fast zone.
- The first fuel pin in the thermal zone.
- The irradiation channel in the thermal zone.

Achievable transmutation rates of TRUs and LLFPs in Myrrha

The fast neutron flux positions in Myrrha are characterised by fast flux values (ϕ>1.0 MeV) estimated to be of the order of 1.0×10^{15} n/cm^2.s. The burn-out of the MAs ^{237}Np, ^{241}Am, ^{243}Am and ^{244}Cm in targets submitted to a fast neutron flux ($\phi_{>1.0\ MeV}$) of 1.0×10^{15} n/cm^2.s is plotted in Figure 3, as a function of irradiation time (the evolution curves only indicate the consumption of the nuclides concerned and do not indicate the formation of daughter products).

The thermal neutron flux positions in Myrrha, on the other hand, are characterised by thermal flux values ($v_0 \int_0^{0.5} n(E)\, dE$) estimated to range from 3.0×10^{14} to 3.0×10^{15} n/cm^2.s. The burn-out, as a function of irradiation time, of selected LLFPs (^{93}Zr, ^{99}Tc, ^{129}I and ^{135}Cs) in targets submitted to thermal flux values ($v_0 \int_0^{0.5} n(E)\, dE$) of 3.0×10^{14} and 3.0×10^{15} n/cm^2.s is plotted in Figure 4 and Figure 5, respectively (the evolution curves only indicate the consumption of the nuclides concerned and do not indicate the formation of daughter products).

Figure 3 **Evolution of selected MAs in the fast flux positions in Myrrha, with ϕ>1.0 MeV = 1.0×10^{15} n/cm^2.s**

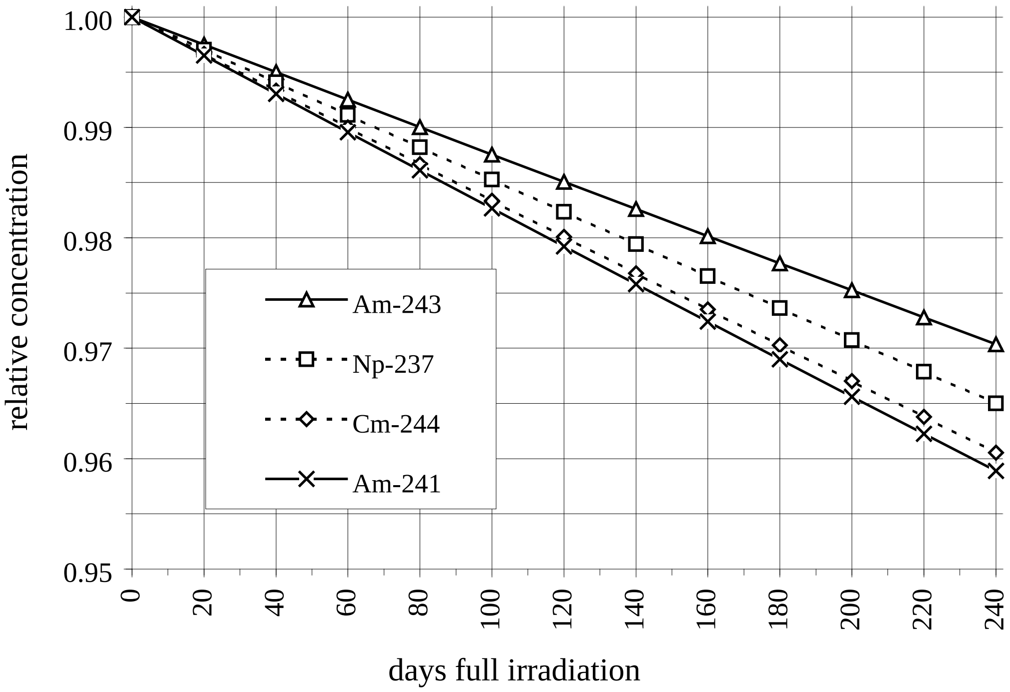


Figure 4 **Evolution of selected LLFPs in low thermal flux positions in Myrrha, with $v_0 \int_0^{0.5} n(E)\, dE = 3.0\times10^{14}$ n/cm^2.s**

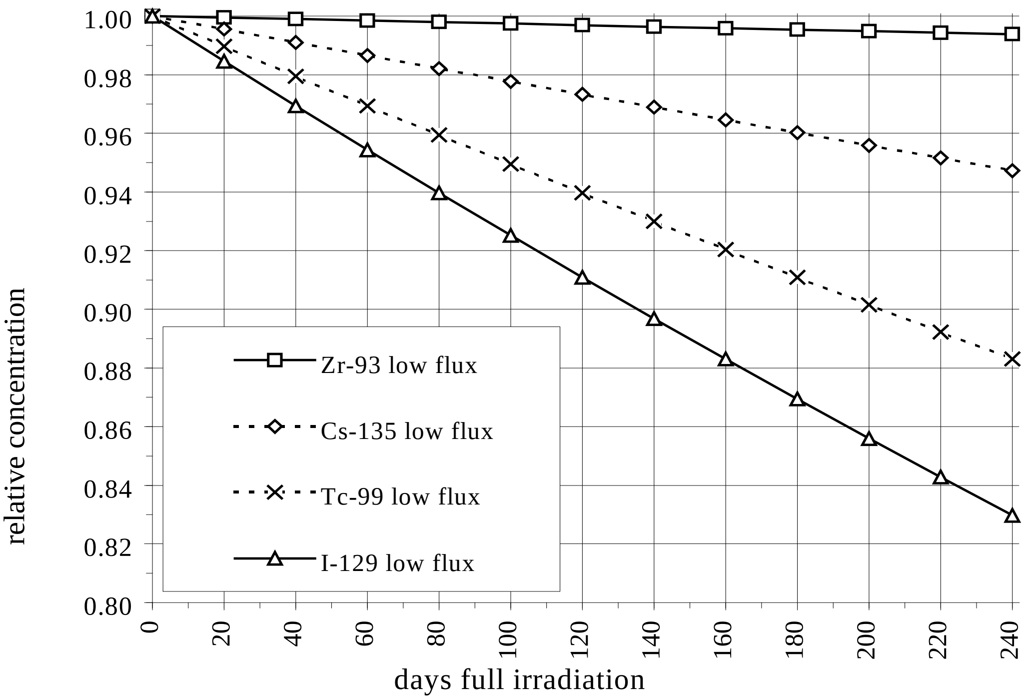
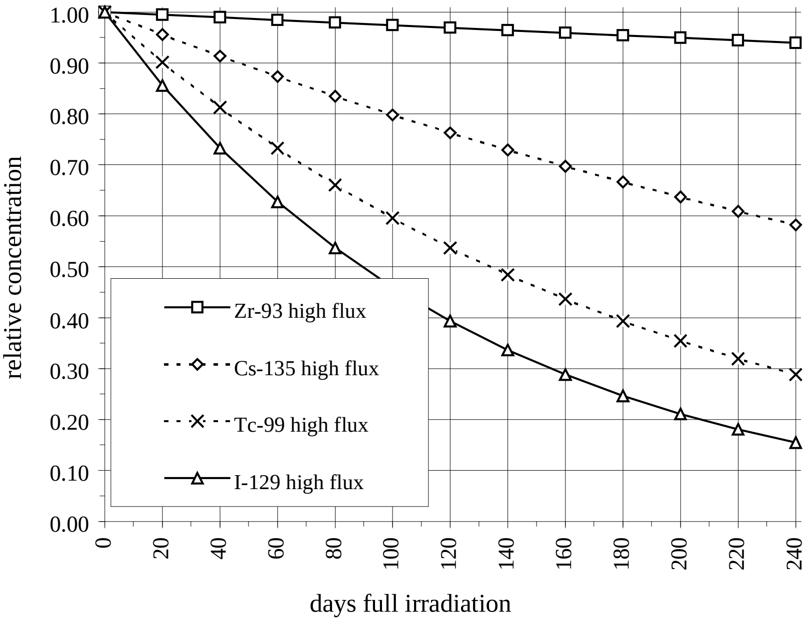


Figure 5 **Evolution of selected LLFPs in high thermal flux positions in Myrrha, with $v_0 \int_0^{0.5} n(E)\, dE = 3.0 \times 10^{15}$ n/cm^2.s**



Conclusions

As our purpose with Myrrha concerning the transmutation research is to provide the possibilities for performing relevant integral demonstration experiments in reasonable irradiation times, one has to look at the radiochemical detection limits and the related depletion rates needed for the different MAs and LLFPs to allow the delivery of relevant data for the validation of the evolution codes. The minimum depletion rates for the ^{99}Tc and the MAs needed by radiochemical labs for delivering 2σ precision data are : 3~5 % whereas for the remaining LLFPs considered here the depletion rate reduces to 1 %.

Given the above limits, we can conclude that transmutation experiments of MAs would be feasible in Myrrha within irradiation times ranging between 100 and 250 days. Whereas for the LLFPs the irradiation time reduces to very short periods of few days (10 to 40 days) if we consider the highest thermal flux position.

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