Neutronics and Shielding Issues of ADS

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Abstract

Accelerator Driven Systems (ADSs) are hybrid systems consisting of a high-intensity proton accelerator with beam energy in the hundreds of MeV range impinging on a target of a heavy element and coupled to a sub-critical core. The intense (of the order of 10^{15} n/cm^2/s) and fast neutron fluxes produced by the spallation reactions triggered by the impinging protons in the target can be used to induce fission reactions in the actinides and capture reactions in the long-lived fission products in the fuel assemblies in the core of the system.

ADSs have been considered during the last fifteen years as one of the promising technological solutions for the transmutation of nuclear waste, reducing the radiotoxicity of the high-level nuclear waste and hence reducing the burden to the geological repositories.

The European Commission´s Green Paper entitled "Towards a European Strategy for the Security of Energy Supply" clearly pointed out the importance of nuclear energy in Europe. With 145 operating reactors producing a total power of 125 GW$_e$, the resulting energy generation of 850 TWh per year provides 35% of the electricity consumption of the European Union. The Green Paper also points out that the nuclear industry has mastered the entire nuclear fuel cycle with the exception of waste management and for this reason, "focusing on waste management has to be continued".

Amongst the several solutions being studied in recent years, MYRRHA (concept developed at SCK-CEN, Belgium), XADS (design studies co-funded by the European Union in the framework of the 5th Framework Programme) and XT-ADS and EFIT (acronyms standing for an experimental machine and for the long term transmuter to be deployed on an industrial scale, both in the EUROTRANS project of the 6th Framework Programme) have deserved the attention of different communities of specialists in the field of Nuclear Technology and Radioactive Waste Management.

Although these machines have been designed with different parameters, their implementation and deployment have in common the fact that they raise cutting edge scientific and technological issues, associated to the operation of the high-intensity proton accelerator, the high-power (in the multi-MegaWatt range) delivered to the target and the material damage in the target and surrounding structures. The thermal power in the core, the thermal-hydraulic aspects associated to the heat removal in steady state and also in transient mode, the subcriticality level of the system and the efficiency of the transmutation process, are particularly sensitive to the core design (geometry, number of subassemblies, fuel composition, among many other aspects).

Neutronics and shielding issues and the computation and mapping of neutron fluxes and doses are important throughout all stages of design of these systems. In this paper, i) the main characteristics and parameters of the ADS systems previously alluded to will be reviewed ii) the neutronics and shielding calculations of relevance for the design of the ADS systems, for radiation damage and for radiation protection purposes will be extensively described.

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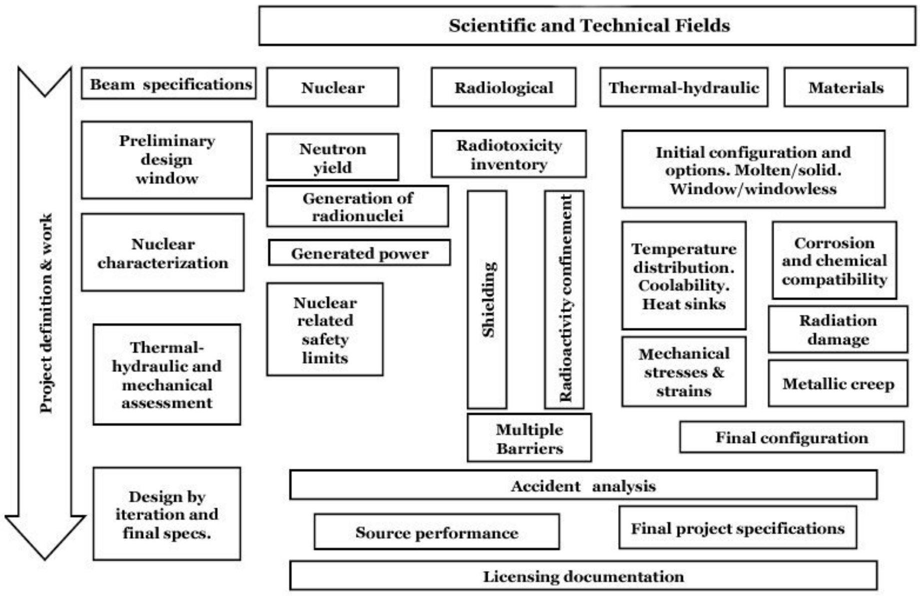
Introduction

A possible representation of the multiple scientific and technical fields involved in the design and deployment of ADS systems are sketched in the following Figure, extracted from reference [1].



Amongst the issues that are directly related to neutronics computations of the target and core components of an ADS systems can be listed: the core design and power distribution, safety criteria and sequences, thermal-hydraulics, radiation damage of structural materials, activation of components and radiotoxicity inventory.
On the other hand, shielding issues are associated to the radioactivity containment, mitigation of radiation damage of structural components and radiation safety and radiological protection of individuals, of relevance for the licensing and operation of the facility.

In the last decade several concepts of accelerator driven systems have been proposed, namely:

- The MYRRHA concept (reference [2]) developed at SCK-CEN, Belgium. It consists of a proton beam with energy of 350 MeV and 5 mA beam current, with a windowless interface between the proton beam tube and the spallation target. A k_{eff} value of 0.95 is considered for the sub-critical core of the system and liquid lead-bismuth (LBE) has been selected as coolant material.
- The XADS concepts (reference [3]) undertaken in the framework of the design studies of the PDS-XADS project co-funded by the European Union in its 5th Framework Programme. Three main concepts have been studied for a 600 MeV proton beam energy and 6 mA beam current with liquid lead-bismuth target and operating with a k_{eff}=0.97 and 80 MW core power:
    - A windowless interface between the proton beam tube and the spallation target, LBE also chosen as the coolant,
    - A 3 mm stainless steel thickness hemi-hemispherical window separating the proton beam tube from the target, LBE also chosen as the coolant and
    - A 3 mm stainless steel thickness hemi-hemispherical window separating the proton beam tube from the target, helium gas cooled

— The XT-ADS (acronym standing for an experimental machine) concept (reference [4]), currently being considered in the framework of the UE co-funded project EUROTRANS in the 6th Framework Programme. It consists of a 600 MeV proton beam energy and 2.3 mA beam current with liquid lead-bismuth target and operating with a k_{eff}=0.95 and 57 MW core power. LBE acts as the material for both the spallation target and coolant.
— EFIT (the long term transmuter to be deployed on an industrial scale) concept (reference [5]), currently being considered in the framework of the UE co-funded project EUROTRANS in the 6th Framework Programme. It consists of an 800 MeV proton beam energy and 20 mA beam current operating with a k_{eff}=0.95 and 400 MW core power. Liquid lead is the selected material for both the spallation target and coolant.

Besides engineering design studies, R&D activities were undertaken in these projects in support of the different scientific and technological aspects involved: accelerator technologies, advanced fuels, materials properties and nuclear data.

Neutronics Issues

Mathematical issues – Boltzmann transport equation, neutron source multiplication factor
In the Boltzmann neutron transport equation written for an ADS, a term has to be added to account for the external source that corresponds to the neutrons produced by spallation reactions triggered by the incoming protons in the target of the system.
In fact, in the sub-critical core of ADS two main categories of neurons exist, according to their production process:
— Neutrons produced by spallation reactions in the target predominantly in the inner region of the core, exhibiting an energy spectrum harder than the typical energy spectrum of fission produced neutrons
— Neutrons produced by fissions in the fissile material of the core assemblies

As explained in reference [6] this led to the definition of a source multiplication factor k_{src} (inspired by the traditional multiplication factor k_{eff} the eigenvalue of the Boltzmann transport equation traditionally used for critical systems) and to the definition of an importance function φ^* that can be written as:

$$\varphi^* = \frac{(1-k_{eff})\cdot k_{src}}{(1-k_{src})\cdot k_{eff}}$$

This function can be interpreted as an assessment of the relative importance of fission produced neutrons compared to the spallation-produced neutrons in the sub-critical core.

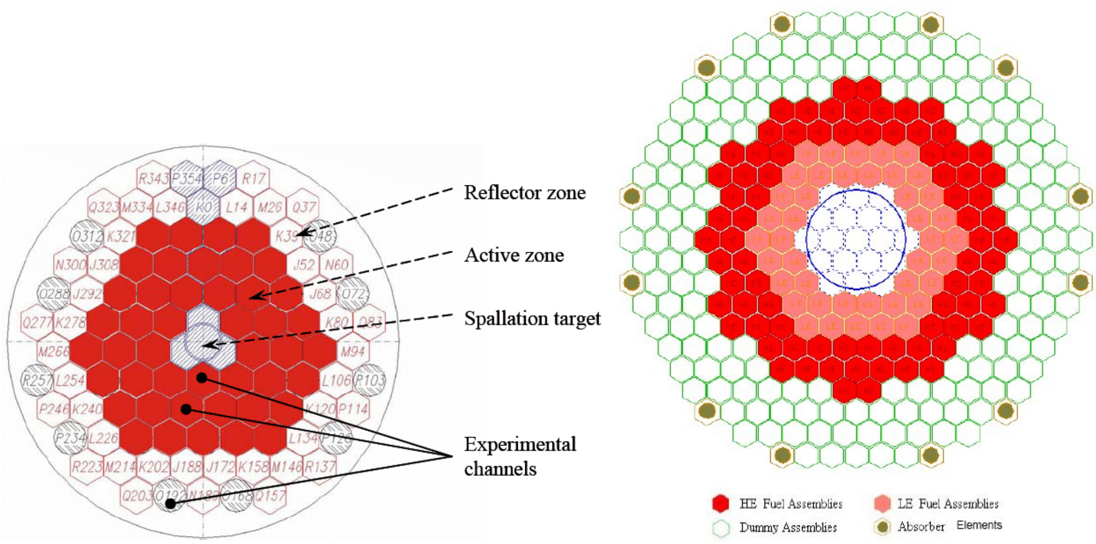
Additionally, considering the higher energy regime, one has to consider the growing importance of the terms corresponding to the (n,xn) normally neglected in the lower than 20 MeV nuclear applications.

Interplay between the different parameters of the system
The equation establishing the interplay between the accelerator parameters (beam current intensity I_P, the parameters characterizing the core (P_{core} – the core power, k_{eff} – the effective multiplication factor, E_f - the mean energy produced per fission in the fuel, ν - the mean number of fission produced neutrons) and the spallation target (Z – the average number of neutrons produced per incident proton, the “yield”, energy dependent) is

$$\underbrace{I_p}_{\text{number of protons per second}} = \frac{1-k_{eff}}{k_{eff}} \cdot \frac{P_{core} \cdot \bar{\nu}}{\bar{E}_f \cdot Z \cdot \varphi^*}$$

Core design

Neutronics issues of relevance in the sub-critical core design of ADS are related to the radial and axial power distribution. Core design must accommodate a number of fuel assemblies in order to provide a fast neutron flux component in excess of 10^{15} neutrons cm^{-2} s^{-1} (typically 1-2 10^{15} n cm^{-2} s^{-1} with energies greater than 0.75 MeV) in order to improve the fission probability in neutron-odd actinide isotopes), at the same time exhibiting a k_{eff} in the range 0.95-0.98 guaranteeing a sufficient safety margin to criticality all this subject to the boundary conditions of aimed proton beam current and core power parameters as displayed in the previous equation.
Studies are normally conducted as a first approximation for the so-called clean core configuration exhibiting a quasi-symmetry in the geometrical placement of the fuel assemblies. However, for some designs (namely MYRRHA and XT-ADS), the clean core configuration is modified in order to include locations accessible from the top of the reactor for on-line monitoring and/or allowing for independent coolant loops. These In-Pile-Positions (IPS) are intended to be used for irradiation and experimental purposes.
The design of the fuel assemblies and of the fuel pins per assembly has to be to carefully established to include considerations such as the inlet and outlet temperatures of the coolant, the temperature of the fuel and cladding and more generally thermal-hydraulics studies associated both to the steady and the transient behavior of the system.
The following Figure extracted from reference [3] depicts the core configurations for the previously described MYRRHA (left) and XADS (right) designs.



Neutron economy

As shown in reference[7] the feasibility of the transmutation process in the ADS can be elegantly assessed by a set of formally simple equations establishing the balance of neutrons in the system as a function of the fuel composition and the system geometry among others. To this end, a set of parameters D_J, the "neutron consumption/fission" of isotope J are used to define the number of neutrons required in the process of its transformation in fission products. These parameters take

into account the nuclear reactions and physics processes (fission, capture, (n,2n), etc.) contributing to the release (“gains”) or absorption (“losses”) of neutrons into the system and establishing the net balance of neutrons resulting in the system due to the isotope *J*. For a given system $D_J > 0$ means net “consumption” whereas $D_J < 0$ translates in net “production”.
As an illustration, being ε_J the fractions of the different transuranium present in the fuel, for a mixing of isotopes

$$D_{TRU} = \sum_J \varepsilon_J^{TRU} D_J^{TRU}$$

and for Plutonium it would become

$$D_{Pu} = \sum_J \varepsilon_J^{Pu} D_J^{Pu} = \varepsilon_{238}^{Pu} \cdot D_{238}^{Pu} + \varepsilon_{239}^{Pu} \cdot D_{239}^{Pu} + \varepsilon_{240}^{Pu} \cdot D_{240}^{Pu} + \varepsilon_{241}^{Pu} \cdot D_{241}^{Pu} + \varepsilon_{242}^{Pu} \cdot D_{242}^{Pu}$$

Then, for a fuel composed of Pu, Am and Cm, it would become

$$D_{FUEL} = \sum_i \underbrace{\varepsilon_i}_{\substack{\text{fraction of Pu,Am,Cm,Np} \\ \text{in the fuel composition}}} D_i$$

As an example, the following Table, extracted from reference[7] displays the parameters *D* for different reactor systems

Table 1.4. *D* (neutron consumption/fission) value for different isotopes in different systems

Isotope	MOX-LWR$^{(1)}$ $r^{(2)} = 1.4$	MOX-LWR$^{(1)}$ $r^{(2)} = 2$	MOX-LWR$^{(4)}$ $r^{(2)} = 2$	MOX-LWR$^{(1)}$ $r^{(2)} = 4$	He-cooled carbide fuel FR$^{(3)}$	SUPER-PHENIX$^{(3)}$	Lead-cooled nitride fuel FR$^{(3)}$	Na-cooled oxide fuel FR$^{(3)}$	Na-cooled metal fuel FR$^{(3)}$
^{235}U	-0.31	-0.38	-0.43	-0.55	-0.84	-0.86	-0.92	-0.95	-1.04
^{238}U	0.104	0.068	-0.06	-0.007	-0.63	-0.62	-0.71	-0.79	-0.90
^{237}Np	0.91	0.93	0.75	0.96	-0.51	-0.56	-0.65	-0.73	-0.88
^{238}Pu	0.014	0.024	-0.16	0.038	-1.25	-1.33	-1.36	-1.41	-1.50
^{239}Pu	-0.60	-0.64	-0.79	-0.73	-1.44	-1.46	-1.58	-1.61	-1.71
^{240}Pu	0.65	0.56	0.14	0.38	-0.93	-0.91	-1.02	-1.13	-1.27
^{241}Pu	-0.26	-0.37	-0.80	-0.58	-1.25	-1.21	-1.26	-1.33	-1.39
^{242}Pu	1.27	1.22	0.73	1.13	-0.65	-0.48	-0.73	-0.92	-1.13
^{241}Am	0.92	0.93	0.71	0.95	-0.56	-0.54	-0.65	-0.77	-0.91
^{242m}Am	-1.55	-1.56	-1.66	-1.56	-2.03	-1.87	-2.08	-2.10	-2.16
^{243}Am	0.44	0.36	-0.15	0.25	-0.84	-0.65	-0.85	-1.01	-1.15
^{242}Cm	0.004	0.014	-0.18	0.026	-1.26	-1.34	-1.37	-1.41	-1.51
^{244}Cm	-0.51	-0.60	-1.12	-0.71	-1.54	-1.44	-1.53	-1.64	-1.71
^{245}Cm	-2.46	-2.46	-2.44	-2.44	-2.70	-2.69	-2.71	-2.74	-2.77

(1) $\varnothing = 1 \times 10^{14}$ n/cm^2.s
(2) r = moderator-to-fuel ratio
(3) $\varnothing = 1 \times 10^{15}$ n/cm^2.s – sodium-cooled burner configuration, GFR and LFR conventional concepts
(4) $\varnothing = 2.5 \times 10^{14}$ n/cm^2.s

In order to ascertain the feasibility of the transmutation process in a particular reactor in terms of the overall neutron balance of the system, reference [7] also suggests the use of a parameter *G*, the neutron economy balance of the system defined as

$$G = \underbrace{S_{ext}}_{\text{external neutron source}} - \underbrace{D_{FUEL}}_{\sum \varepsilon_i \cdot D_i} - (\underbrace{L}_{\text{neutron leakage term}} + \underbrace{C_{par}}_{\text{term of capture in structural materials}} + \underbrace{C_{FP}}_{\text{term of capture in fission products}})$$

If $G > 0$, the transmutation is feasible in the ADS system considered.

Shielding Issues

Shielding issues can be broadly categorized in two main groups according to the ultimate purpose and objective pursued, namely: i) the protection of the structural components elements from the damage caused by high radiation doses and neutron fluxes and ii) the protection of professionals (and members of the public) for radiation protection purposes.
In ADS systems, high instantaneous doses and neutron fluxes are further enhanced by the very high availability and reliability aimed for the performance of the systems.
As for the shielding of structural elements, radiation damage is of concern for, amongst others, two main components of the systems: the core vessel and the core support plate where the fuel assemblies will be hanging from. Considering the very high and fast neutron fluxes, in excess of 10^{15} n cm^{-2} s^{-1}, damage assessment in terms of displacement per atoms (dpa) and gas (helium and hydrogen production) is, in the absence of appropriate shielding, of several dpa / year for the vessel, compromising the lifetime of such crucial system component.
As for the protection of professionals, exposure of personnel has to be kept as low as reasonably achievable, according to the basic principles and foundations of the international radiation protection system. Exposure of professionals can result from the two ADS components: the core and the proton accelerator. Shielding systems have to be designed as containment barriers normally of steel and concrete in order to achieve the attenuation of the high radiation levels and dose rates as low as reasonably achievable and anyhow below the legal dose limits, 20 mSv/year for radiation workers and 1 mSv/year for non-radiation workers or members of the public (reference [8]). The dose rates in the core being extremely high, several meters of concrete have to be implemented in order to guarantee, in case of necessity, that human intervention in nearby areas is possible.

Shielding of the proton accelerator – analytical models

It is commonly accepted that the core of ADS is to be located underground, attenuation of the high radiation doses and fluxes being assured by the surrounding earth. Concerning the accelerator, detailed studies indicate that a linear accelerator is the most favorable solution. The accelerator components will be in an underground tunnel, with several meters of concrete and earth attenuating the radiation doses in the tunnel and making sure that at the surface the dose rates comply with the values imposed by the regulators and licensing authorities.
Accelerator shielding calculations for a proton accelerator are often carried out using the Moyer model, as detailed in reference [9]. The effective dose rate at a distance r from a proton beam target, under an angle θ, behind a shield wall of thickness d is given by the general theoretical expression:

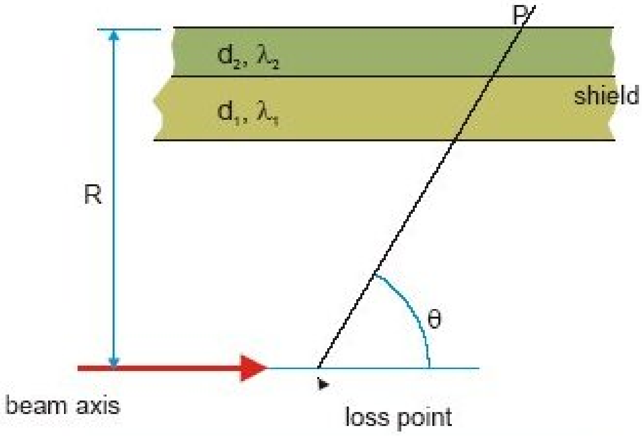
$$\dot{H} = \frac{1}{r^2} \int F(E) B(E,\theta) e^{-d(\theta)/\lambda(E)} \frac{d^2 n(E,\theta)}{dE d\Omega} dE$$

with F(E) the fluence to effective dose conversion factor, B(E,θ) a build-up factor, λ(E) the effective attenuation mean free path and $\frac{d^2n(E,\theta)}{dEd\Omega}$ the differential neutron yield.

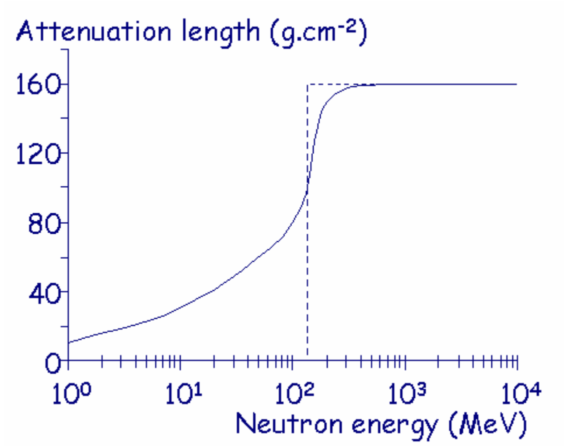
From this general theoretical formula, Moyer derived the simple analytical expression for the shielding of high energy proton accelerators, recognizing that the fluence to effective dose conversion factor is approximately proportional to the primary proton energy, and that at high proton energies the dose outside the shield wall will be caused by high energy neutrons ($E \geq 150$ MeV) and low energy neutrons in radiation equilibrium with them, allowing the use of a single dose attenuation length independent of neutron energy:

$$H(\theta) = \frac{N \cdot \Psi(E_P) \cdot \exp(-\beta\theta) \cdot \exp\left(-{l}/{\sin\theta}\right)}{{R^2}/{\sin^2\theta}}$$

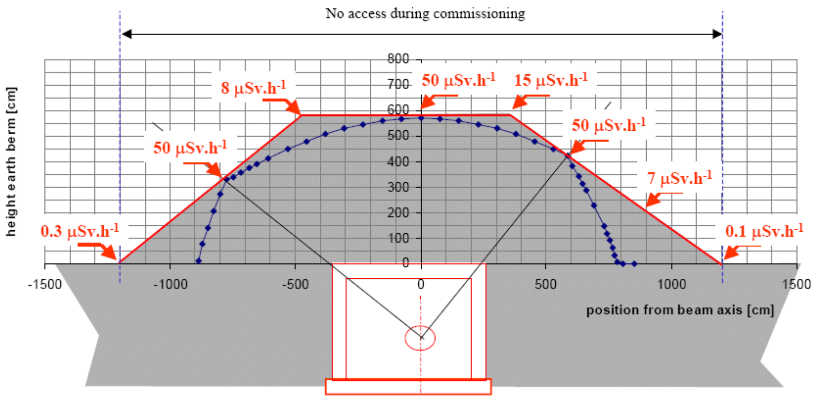
with N the number of protons interacting in the target, β=2.3 rad^{-1}, $\Psi(E_P)=2.8\text{x}10^{-13}\ E_P^{0.8}$ Sv m^2, E_P the beam energy in GeV and $l=\Sigma\ (d_i/\lambda_i)$ being λ_i the neutron attenuation length in material i along the flight path and d_i and R as indicated in the following Figure.



It should be stressed at this point the semi-empirical dependence with energy of the neutron attenuation length in the material, as shown in the next Figure.
Using similar arguments, Sullivan (reference [10]) has developed a model that can be used also for proton energies below 1 GeV. A very enlightening in-depth discussion of the accelerator shielding analytical methodologies using these models is provided in reference [12].



As an example of the application of the analytical computational methodologies previously described, the following picture extracted from reference [11] displays (blue line) the minimum earth profile above a 60 cm thickness concrete tunnel for a proton accelerator of 600 MeV and assuming a beam loss rate of 1 nA m^{-1}, for a residual dose rate of 0.5 µSv h^{-1}. The red line corresponds to a realistic earth profile. The dose rate values shown along the earth profile correspond to the maximum values expected during commissioning or start-up periods, assuming beam losses 100 times higher than during normal operation (100 nA m^{-1}).



Computation of radiation damage and gas production

We refer to the methodology frequently adopted for the computation of the radiation damage based on modeling (references [13],[14], [15]) established several decades ago, on first principles and in some empirical data. The displacement of atoms from the lattice structure following neutron scattering and the development of the cascade of displacements with the creation of Frenkel pairs and the modeling of interstitial creation etc., needs experimental validation, some authors claiming the existing methodology clearly leads to conservative results.

Being σ_{dis} the displacement cross-section for an incident particle of energy E, $\phi(E)$ the incident particle flux and t the irradiation time the number of displacement per atoms is given by

$$dpa = \left(\int \sigma_{dis}(E)\phi(E)dE \right) \cdot t$$

The displacement cross-section can be obtained from the following equation:

$$\sigma_{dis} = \int_{T=E_d}^{T_{\max}} \nu_d(T) \cdot \frac{d\sigma_d(E,T)}{dT} dT$$

With T the PKA energy, T_{max} the maximum value of T, T_d the atomic threshold displacement energy, ν_d the displacement damage function and $d\sigma_d/dT$ the energy differential cross section. The displacement damage function is commonly taken from reference [13] as

$$\nu_d(T) = \left(\frac{\eta}{2E_d}\right) T_{dam}$$

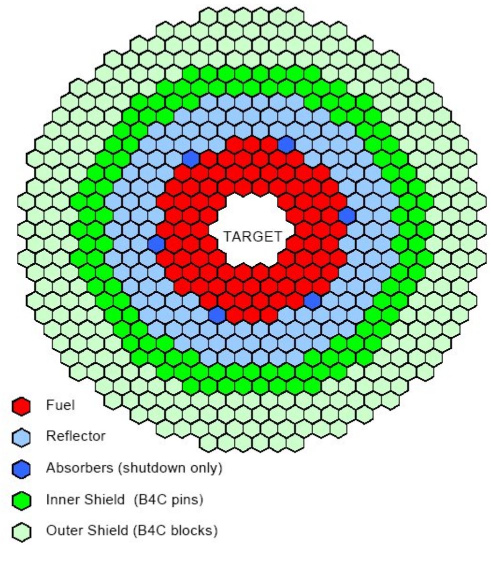
In practice it turns out that the computation of the displacement per atom (dpa per second) is approximately performed using an expression of the type

$$\frac{dpa}{s} = \sum_E \frac{\langle \sigma E \rangle}{2E_d} \eta \cdot \phi(E) \cdot 10^{-24}$$

With E_d in the range 25-40 eV and η=0.8, the quantity <σE> in units *barn-eV* being the damage energy cross-section computed by data processing modules of NJOY (reference [16]) among others.

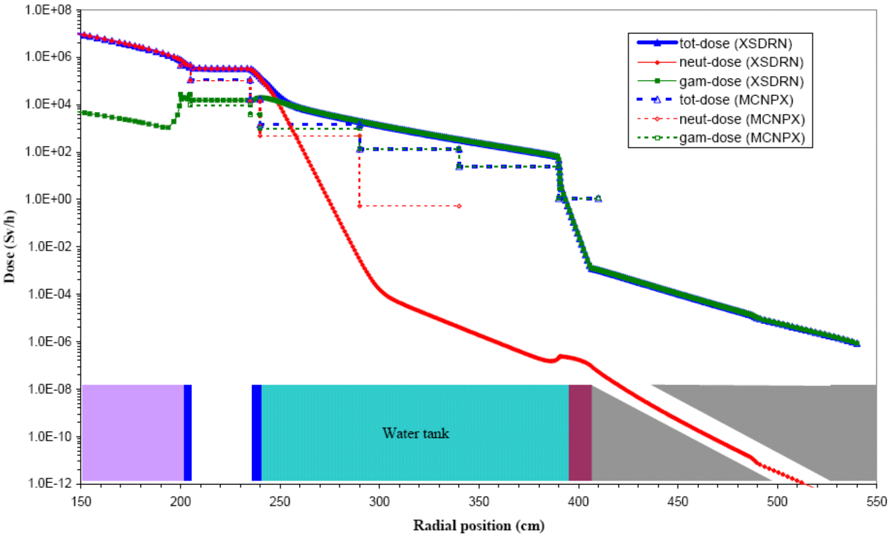
Core shielding

The attenuation of the neutron fluxes at the periphery of the core can be attained by absorbing elements (e.g. B_4C) as well as other materials, in order to protect the core vessel and limit to values close to 1 dpa / year the damage in this structural component which lifetime is expected to remain in the 30-40 years.
The following Figure extracted from reference [3] shows the core configuration for the previously described XADS gas-cooled design, with the inner rings of fuel assemblies followed by reflector positions, inner shield made of B_4C pins and outer shield (outermost positions) of B_4C blocks.



Radiation Protection and Dosimetry issues

It is commonly accepted that the core of ADS is to be located underground, attenuation of the high radiation doses and fluxes being assured by the surrounding earth. Concerning the accelerator, detailed studies indicate that a linear accelerator is the most favorable solution. The accelerator components will be in an underground tunnel, with several meters of concrete and earth attenuating the radiation doses in the tunnel and making sure that at the surface the dose rates comply with the values imposed by the regulators and licensing authorities. The following Figure extracted from reference [17] displays the radial dose distribution for the MYRRHA core in the axial mid-plan.



Nuclear Data Uncertainties

The impact of nuclear data uncertainties on the transmutation of actinides in ADS assemblies has been exhaustively assessed in reference [18]. In this study, an uncertainty analysis based on sensitivity theory has been performed by the authors in order to evaluate the impact of nuclear data uncertainties on a large number of performance parameters of reactor cores dedicated to the transmutation of radioactive wastes and on the radiation damage (dpa, Helium and Hydrogen production) calculations. The results obtained clearly establish which reaction cross-sections, which energy groups and which nuclides (mainly actinides but also isotopes constituents of structural materials) are responsible for the most significant uncertainties, the overall uncertainty, dependent on the parameter under study, ranging from a few percent up to several tens of percent (in the worst cases).

A subset of the main results obtained from the study undertaken can be summarized as follows:

- "As could be anticipated, the most crucial data are fission, capture, and inelastic cross sections of Minor Actinides. However, specific data related to decay heat or β_{eff} assessment are of high relevance. Finally, in the case of a Pb/Bi coolant, the data for these materials should be definitely improved, in particular inelastic and (n,2n) data".
- "High-energy data (E .>20 MeV) uncertainties also play a role, but for the transmutation core, only a few data are relevant. Besides (n, α) and (n, p) data for structural materials, only Pb and Bi high-energy data uncertainties are significant. For the major integral parameters considered, there is no serious impact of MA data at E >20 MeV".

Acknowledgements

A special acknowledgement is due to all the contributors to the engineering design studies in the framework of the MYRRHA, XADS, XT-ADS and EFIT concepts.
The design studies of XADS were carried out in the framework of the EU-funded project PDS-XADS, contract Nr. FIKW-CT-2001-00179 in the framework of the 5th Framework Programme. The XT-ADS and EFIT concepts are currently being conducted in the framework of the EU´s 6th Framework Programme project IP-EUROTRANS, contract Nr. 516520 (FI6W).

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