# Design of an Accelerator-Driven System for the Destruction of Nuclear Waste

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#### **Abstract**

Progress in particle accelerator technology makes it possible to use a proton accelerator to produce energy and to destroy nuclear waste efficiently. The Energy Amplifier (EA) proposed by Carlo Rubbia and his group is a sub-critical fast neutron system driven by a proton accelerator. It is particularly attractive for destroying, through fission, transuranic elements produced by present nuclear reactors. The EA could also transform efficiently and at minimal cost long-lived fission fragments using the concept of Adiabatic Resonance Crossing (ARC) recently tested at CERN with the TARC experiment.

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#### 1 Introduction

Because particle physicists, interested in discovering the ultimate structure of matter, have pushed particle accelerator technology as far as they have, it is possible today to consider using a proton accelerator to drive a new type of nuclear system, with very attractive properties.

Today, the world is facing an extremely difficult challenge, that of producing sufficient energy to sustain economic growth without ruining the ecological equilibrium of the planet. The massive use of fossil fuels has allowed the Western World to reach an unprecedented level of wealth. Unfortunately, if the rest of the Earth's population were to carry out the same energy policy, the entire planet would be in serious trouble. There is, therefore, a moral obligation for developed countries to provide new energy sources for the entire world in order to minimize global warming and other effects of pollution.

If an acceptable solution is found, it will certainly be the result of systematic R&D, and in this context, nuclear energy should be part of this R&D. The present nuclear energy programme is meeting growing public opposition in Europe and other parts of the world because of three main reasons: (a) the association with military use and the fear of nuclear weapon proliferation; (b) the fear of accidents such as Chernobyl (1986 prompt-supercritical reactivity excursion) and Three Mile Island (1979 loss-of-coolant accident resulting in a core meltdown); (c) the issue of the back-end of the fuel cycle (nuclear waste management: at this time only deep geological storage is seriously envisaged).

Obviously, without these drawbacks, nuclear power would be ideal as it releases neither greenhouse gases nor chemical pollutants ( $NO_x$ ,  $SO_x$ , etc.), and less radioactivity than a coal-fired generating station (coal ashes contain uranium and thorium). Therefore, the real question facing scientists today is: Is it possible to change nuclear energy production in such a way as to make it more acceptable to society? Nuclear energy is a domain that has essentially seen no significant fundamental R&D since the end of the 1950s when the first civil power plants came into operation. There have been many technological improvements, mainly with the purpose of improving safety. However, we have seen that even these were not sufficient.

The concept of the EA [1] was proposed by C. Rubbia and his group specifically as an answer to the concerns raised by current nuclear energy production. The present EA version is optimized for the elimination of nuclear waste, as it is considered to be the most pressing issue in the Western World. In developing countries such as China and India, where there is virtually no nuclear waste, a version of the EA optimized for energy production, adapted to the detailed needs of the country and with minimized waste production, is the more appropriate solution.

#### 2 Nuclear Waste

Transuranic elements (TRU) and fission fragments (FF) are the two main components of nuclear waste, representing respectively 1.1% and 4% of spent nuclear fuel. TRU, which are produced by neutron capture in the fuel eventually followed by decay, can only be destroyed by fission, while FF can only be destroyed by neutron capture; therefore, different methods will have to be used to eliminate them. As the long-term radio-toxicity of waste (Figure 1) is clearly dominated by TRU, the EA has been designed to destroy them with the highest efficiency.

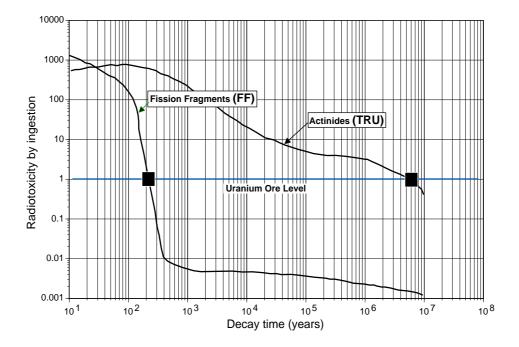


Figure 1: Time evolution of the potential radio-toxicity (relative to uranium ore) of the two main components of nuclear waste for PWR spent fuel.

At present most of the nuclear waste is kept under surveillance in shallow depth storage facilities (e.g. cooling ponds in nuclear power plants). However, concerns about leaks in the biosphere and proliferation risks implies that this can only be a standby solution to be followed either by permanent storage in deep geological repositories and/or transmutation into "harmless" nuclear species.

# 2.1 Deep geological storage

It is proposed to store permanently LWR waste directly from the reactor in bentonite (USA) or after reprocessing and vitrification (Europe). This would result in a large decay heat inside the storage cave ( $\approx 130$  °C). There will still be some concerns about possible leaks in the biosphere of <sup>99</sup>Tc and <sup>129</sup>I but after long times. The possibilty of intrusions and the proliferation risks that this would generate can not be discarded, specially when waste plutonium is converted to bomb grade plutonium after some 20 thousand years.

This option is technically possible but not trivial and may turn out to be very expensive ( $\approx 1000 \text{ } \text{s/kg}$  of spent fuel). The estimated costs of geologic storage are shown in Figure 2.

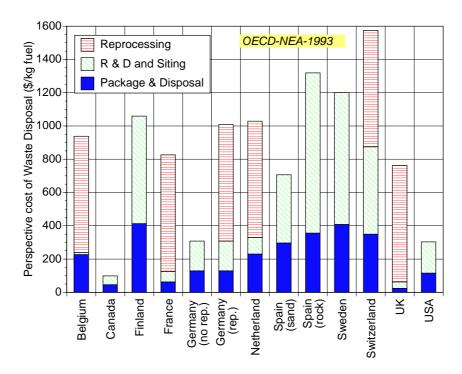


Figure 2: Estimated costs of geologic waste disposal for unit fuel mass. The actual package and Disposal and the Preparation and R & D costs are indicated. Reprocessing is added, whenever appropriate (700 \$/kg).

Underground storage may be performed either with or without reprocessing. Reprocessing is presumed to offer added insurance to containment, since it allows vitrification and optimal packing according to the nature of each element, but at

extra cost. We remark the large amount of R & D and Siting costs before the actual Package and Disposal and the added costs due to Reprocessing. In our view these figures are in many instances underestimates, since for instance a full benchmark study of all the consequences of deep storage of sub-critical mixtures has not been performed and the public opposition is still hard to quantize in terms of added costs.

The actual cost of the Geologic Repository is therefore rather uncertain, as evidenced by the spread figures given in Figure 2. At the estimated cost of 800 \$/kg<sup>1</sup> for Spain (sand solution and no reprocessing) this represents a projected cost of about 7.7 B\$.

#### 2.2 Transmutation before storage?

It is a fortunate circumstance that 99.995% of the long lasting (> 500 years) radio-toxicity is in a few elements and about 1% of the spent fuel ( $\approx 300$  kg/year/GWe). The spent fuel contains several elements which are resilient to further burning in a LWR. Therefore, full fission + transmutation cannot be done in an ordinary reactor (using thermal neutrons). In order to eliminate them more efficiently, fast neutrons are necessary.

Very fast neutrons are produced by protons accelerated to a medium energy ( $\sim 1~GeV$ ) by a dedicated, compact, high-current accelerator. This device called Accelerator-Driven System, ADS, is:

- 1. non critical, since the process rate is fully controlled by the current of the accelerator,
- 2. preferable to a fast breeder reactor, for higher safety, flexibility and efficiency.

The transmutation goals are to eliminate 99.9% of the TRU and up to 95% of the long-lived fission fragments ( $^{99}TC$  and  $^{129}I$ ).

• **Direct use of spallation neutrons.** The cost (in terms of energy) of incinerating a given fraction of the fission fragments produced is given by:

$$E_{fp} = q_{fp} \cdot \frac{E_p}{\eta_{sp}} \cdot \frac{1}{\eta_b \eta_T} \qquad [MeV]$$

where,  $q_{fp}$  = fraction of FP to be transmuted ( $^{99}$ Tc,  $^{129}$ I,  $^{135}$ Cs,  $^{90}$ Sr,  $^{93}$ Zr  $\approx 28\%$ )  $E_p$  = incident proton energy ( $1000 \ MeV$ )

<sup>&</sup>lt;sup>1</sup> It should be pointed out that cost estimates vary widely from country to country. For instance in the case of Switzerland, where only 5 reactors are in operation, the much higher figure of 14 B SWF (11 B\$) has been given, for a complete storage by year 2061.

 $\eta_{sp}$  = spallation neutron yield ( $\approx 30$  for Pb target)

 $\eta_b$  = electrical efficiency for accelerating protons ( $\approx 50\%$ )

 $\eta_T$  = thermal efficiency ( $\approx 33\%$ )

Hence,

$$E_{fp} = 0.28 \times \frac{1000}{30} \times \frac{1}{0.5 \times 0.33} \approx 60 \quad MeV$$

This would represent  $60/200 \approx 30\%$  of the total fission energy produced which is prohibitive from an economical point of view.

• In an ADS. The neutron economy is enhanced by further multiplying the spallation neutrons in a sub-critical medium. In such a system, the fission power extracted,  $P_{fi}$ , is given by:

$$P_{fi} = \eta_{sp} \cdot \frac{\varphi^* \cdot k}{\nu(1-k)} \cdot \frac{i}{C} \cdot E_f$$

where, k = neutron multiplication factor

 $\varphi^*$  = source importance ( $\approx 1.5$ )

 $\nu$  = neutrons emitted per fission ( $\approx 2.5$ )

 $E_f$  = energy generated per fission ( $\approx 3.1x10^{-10}$  W)

i = accelerator current

 $C = \text{charge of a proton} (= 1.6x10^{-19} C)$ 

$$E_{fp} = \frac{\eta_{sp} \cdot \frac{k}{\nu(1-k)} \cdot E_f - \frac{E_p}{\eta_b \cdot \eta_T}}{\eta_{sp} \left[ (1-k) \cdot \eta_{fp} + \frac{k}{(1-k)} \cdot \left( (1-\frac{k}{\nu}) \cdot \eta_{fp} - \frac{q_{fp}}{\nu} \right) \right]}$$

In order for the process to be self-sufficient,

$$k \ge \frac{1}{1 + \frac{\eta_{sp} \cdot \eta_b \cdot \eta_T \cdot E_f}{\nu \cdot E_p}} \approx 0.7$$

#### 3 Historical Background of Accelerator-Driven Systems

#### 3.1 Early history of ADS

In 1941, Glenn Seaborg produced the first man-made plutonium using an accelerator. However, the first practical attempts to promote accelerators to generate potential neutron sources were made in the late 40's by E.O. Lawrence in the United States, and W.N. Semenov in the former Soviet Union. During the period 1950-54, the MTA (Materials Testing Accelerator) programme [2] at Lawrence Livermore (at that time the Livermore Research Laboratory) investigated in detail the use of accelerators to produce fissionable material. This project was soon abandoned when high-grade uranium ores were discovered in the United States.

Almost concurrently in Canada, Lewis realised the value of accelerator breeding in the power programme and initiated spallation neutron yield measurements with the McGill cyclotron. The Canadian team at Chalk River [3] has always been a strong proponent of such a producer of fissile material which could be used in conjunction with a conversion-efficient CANDU reactor.

A materials production accelerator – the Electronuclear Reactor – was patented in 1960 by Lawrence et al. to provide adequate quantities of material which can only be produced artificially. The targets considered were natural uranium and thorium and the artificially produced materials were <sup>239</sup>Pu and <sup>233</sup>U respectively. This concept of accelerator breeding was also studied by Russian scientists. Under the guidance of V.I. Goldanski, R.G. Vassylkov [4] made a neutron yield experiment in depleted uranium blocks using the accelerator in Dubna. Later studies (1975-88) on the Fertile-to-Fissile Conversion (FERFICON) Programme [5] – a collaborative effort with various laboratories – investigated the energy dependence, up to 800 MeV, of the fertile-to-fissile conversion efficiency using standardised target materials and geometries.

A relatively realistic concept of an "Accelerator Driven System" (ADS) in the present meaning, where safety issues and transmutation of waste play an important role, was developed in the late eighties by a research group at Brookhaven National Laboratory lead by H. Takahashi and G. Van Tuyle [6] and is now carried out in Japan as part of the OMEGA programme [7].

The first detailed design of a transmutation facility using thermal neutrons was published by C. Bowman's Los Alamos group in 1991 introducing a common name The Accelerator Transmutation of Waste (ATW) [8].

In 1993 a group of CERN scientists led by Carlo Rubbia presented the basic concepts of a so-called "Energy Amplifier", a sub-critical nuclear system based on U-Th cycle, fed by a high intensity proton accelerator having the purpose to produce energy with very small amount of minor actinide (MA) and long-lived fission fragment (LLFP) production. Later on the scientific feasibility and the

verification of the principle of energy amplification by a high energy cascade were proven in experiments such as FEAT (autumn 1994) and TARC (1997-1998).

FEAT [9], an experiment carried out at CERN under the leadership of Carlo Rubbia, with the participation of research groups from France, Greece, Italy, Spain and Switzerland, stands for First Energy Amplifier Test, and was an experiment based on a sub-critical core of 3.5 tons a metallic natural uranium driven by an intense neutron source activated by a powerful beam of protons coming from the PS accelerator at CERN. Both natural uranium and lead targets were used in the experiments, where power, flux and temperature distributions and time evolution were recorded.

TARC [10] represented a second series of experiments which was carried out at CERN by the same team in order to study the adiabatic resonance crossing of neutrons in a matrix of lead with some samples of specific material, particularly <sup>99</sup>Tc. The TARC experiment (from Transmutation by Adiabatic Resonance Crossing) was conclusive to demonstrate that an appropriate neutron spectrum is shaped in a large lead matrix in order to enhance neutron capture in any significant resonance. This was the case for <sup>99</sup>Tc, which was transmuted into <sup>100</sup>Tc, rapidly decaying into <sup>100</sup>Ru (stable). The experiments showed that the ARC method is a powerful neutron technique for burning any type of nuclei showing resonances (which is the case for all offending nuclei in nuclear waste management).

#### 3.2 Recent developments

In 1998 the Research Ministers of France, Italy and Spain, recognising the potentialities of Accelerator Driven Systems for the transmutation of long-lived nuclear waste, decided to set up a Group of Advisors (Ministers' Advisors Group – MAG) to define a common R&D European platform on ADS.

A Technical Working Group (TWG) under the chairmanship of Carlo Rubbia was also established with the task of identifying the critical technical issues in which R&D, in such a demonstration programme is needed. In October 1998, the TWG issued an Interim Report [11] which, in particular, highlighted a) the need for a demonstration programme, b) the basic components and the different options for the proposed facility, and c) the R&D directly relevant to the realisation of such a facility.

In September 1999, the ETWG – composed of representatives of Austria, Belgium, Finland, France, Germany, Italy and Spain – issued a new technical report [12] aimed at providing an overview of the different ongoing activities on ADS in various European countries.

At the beginning of 2000 the ETWG (further enlarged to representatives of the JRC, Portugal and Sweden), recognising that the R&D programme on ADS

has reached a turning point with regard to programme co-ordination and resource deployment in Europe and taking also into account the substantial recent progress on the subject in the United States and in Japan, issued a so called "four-page document" [13] on a strategy for the implementation of an ADS programme in Europe. In particular, the document called for the urgent definition of a consensual European "Roadmap" towards the demonstration of feasibility of a European waste transmutation facility.

In the European Roadmap towards the experimental demonstration of ADS [14], several experiments are indicated which should allow to validate the separated components of an ADS. This is the case for the accelerator (IPHI, TRASCO), the target (e.g. the MEGAPIE experiments), the sub-critical core (the FEAT, TARC and MUSE experiments [15]). The coupling of the components will be performed in a new system, with all innovative features included, with the exception of the sub-critical core fuel, which should be of a well-proven type. However, the case for the licensing of such a system can present some difficulty, in absence of a preliminary coupling experiment at power. A way out is simply to realise an experiment where an "existing" (low) power reactor, with well-known safety features, is made sub-critical and coupled with an accelerator which should provide the needed protons to induce spallation reactions on a target hosted inside the core.

As far as coupling, such experiment will not need a high neutron yield from spallation. In fact it could be run even with a neutron per proton production rate as low as one, since an optimisation in terms of efficiency or transmutation will not be a requirement.

The domain of interest of such experiment will be to show a reliable operation of the system, from start-up to nominal power level, up to shutdown, in presence of thermal reactor feedback effects. The presence of control rods in the system will allow to verify different modes of operation during fuel irradiation and the determination and monitoring of reactivity levels with "ad-hoc" techniques. The joint cooling of the target and of the sub-critical core will be demonstrated, together with the solution of some practical engineering problems of generic interest for an ADS, such as the configuration of the beam ingress into the core.

The possibility to run the experiment at different levels of sub-criticality (realised e.g. with appropriate fuel loading patterns), will allow to explore experimentally the transition from an "external" source-dominated regime, to a core thermal feedback-dominated regime. This transition is relevant, in particular to understand the dynamic behaviour of an ADS, which, in the future full scale demonstrations of transmutation, could have both a very low  $\beta_{eff}$  and very low Doppler reactivity effect.

Carlo Rubbia proposed that this pilot experiment, which would be the first example of ADS component coupling "at real size", could be carried out on the TRIGA reactor at the ENEA Casaccia Centre (Italy), an existing pool reactor of 1 MW thermal power, cooled by natural convection of water in the reactor pool. The TRADE project [16] is based on the coupling of an upgraded commercial proton cyclotron with a tungsten solid target, surrounded by the TRIGA reactor in a sub-critical configuration. Indeed, the flexibility offered by the pool reactor is eminently suited for the conversion into that configuration.

#### 4 Physical Features of Accelerator-Driven Systems

The basic process of accelerator-driven systems is nuclear transmutation. This process was first demonstrated by Rutherford in 1919, who transmuted  $^{14}N$  to  $^{17}O$  using energetic  $\alpha$ -particles. I. Curie and F. Joliot produced the first artificial radioactivity in 1933 using  $\alpha$ -particles from naturally radioactive isotopes to transmute boron and aluminum into radioactive nitrogen and oxygen. It was not possible to extend this type of transmutation to heavier elements as long as the only available charged particles were the  $\alpha$ -particles from natural radioactivity, since the Coulomb barriers surrounding heavy nuclei are too great to permit the entry of such particles into the atomic nuclei.



The invention of the cyclotron by E.O. Lawrence in 1939 removed this barrier and opened quite new possibilities. When coupled with the spallation process, high power accelerators can be used to produce large numbers of neutrons, thus providing an alternative method to the use of nuclear power reactors for this purpose (Figure 3).

Spallation offers exciting new possibilities for generating intense neutron fluxes for a variety of purposes.

One way to obtain intense neutron sources is to use a hybrid sub-critical reactor-accelerator system called just Accelerator-Driven System (ADS) as illustrated in Figure 3.

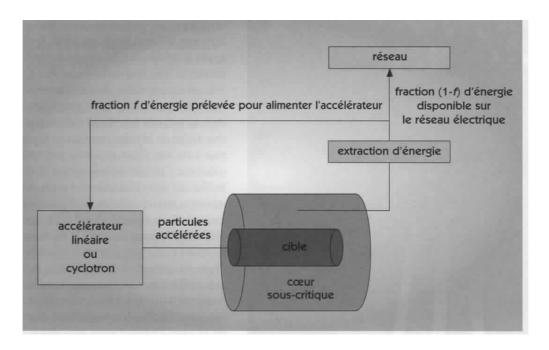


Figure 3: Standard scheme of an ADS (courtesy of CEA).

The accelerator bombards a target with high-energy protons which produces a very intense neutron source through the spallation process. These neutrons can consequently be multiplied in the sub-critical core which surrounds the spallation target.

The original idea of exploiting the spallation process to transmute actinide and fission products directly (that is to use only the spallation neutrons generated in the target) was soon abandoned. The proton beam currents required were much larger than the most optimistic designs that an accelerator could achieve, which are around 300 mA.

#### 4.1 The spallation process

Several nuclear reactions are capable of producing neutrons (Table 1). However, the use of protons minimises the energetic cost of the neutrons produced.

Nuclear Reactions	Incident Particle & Typical Energies	Beam Currents (part./s)	Neutron Yields (n/inc.part.)	Target Power (MW)	Deposited Energy Per Neutron (MeV)	Neutrons Emitted (n/s)
(e,γ) & (γ,n)	e (60 MeV)	$5\times10^{15}$	0.04	0.045	1500	$2 \times 10^{14}$
H <sup>2</sup> (tn)He <sup>4</sup>	H <sup>3</sup> (0.3 MeV)	$6\times10^{19}$	10 <sup>-4</sup> — 10 <sup>-5</sup>	0.3	$10^4$	10 <sup>15</sup>
Fission			≈ 1	57	200	$2 \times 10^{18}$
Spallation (non-fissile target)	p (800 MeV)	1015	14	0.09	30	$2\times10^{16}$
Spallation (fissionable target)	р (800 Меч)	10	30	0.4	55	$4\times10^{16}$

Table 1: Nuclear reactions capable of producing neutrons.

There is no precise definition of spallation: this terms covers the interaction of high energy hadrons (e.g. protons, neutrons, pions, etc.) or light nuclei (deuterons, tritons, etc.), from a few tens of MeV to a few GeV, with nuclear targets. It corresponds to the reaction mechanism by which this high energy projectile pulls out of the target some nucleons and/or light nuclei, leaving a residual nucleus (spallation product). Depending upon the conditions, the number of emitted light particles, and especially neutrons, may be quite large. This is of course the feature of outermost importance for the so-called ADS.

At these energies it is no longer correct to think of the nuclear reaction as proceeding through the formation of a compound nucleus. The initial collision between the incident projectile and the target nucleus leads to a series of fast direct reactions (Intra-Nuclear Cascade,  $\sim 10^{-22}~sec$ ) whereby individual nucleons or small groups of nucleons are ejected from the nucleus. At energies above a few GeV per nucleon, fragmentation of the nucleus can also occur (Pre-Compound Stage,  $< 10^{-18}~sec$ ). After the intra-nuclear cascade phase of the reaction, the nucleus is left in an excited state. It subsequently relaxes its ground state by "evaporating" nucleons, mostly neutrons.

The spallation process is depicted in Figure 4, showing two stages of the process (intra-nuclear cascade and evaporation). For thick targets, high energy secondary particles can undergo further spallation reactions (inter-nuclear cascade) as illustrated in Figure 5.

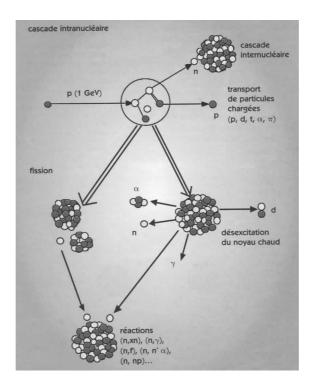


Figure 4: Illustration of the spallation process in thick targets, with evaporation competing with high energy fission (courtesy of CEA).

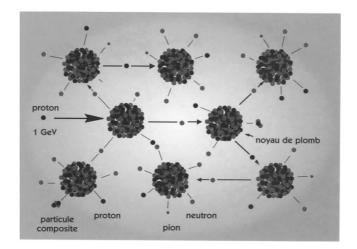


Figure 5: Illustration of the inter-nuclear cascade in thick targets (courtesy of CEA).

For some target materials, low energy spallation neutrons can enhance neutron production through low energy (n,xn) reactions. For heavier nuclei, high energy fission can compete with evaporation (e.g. tungsten and lead). Some spallation targets such as thorium and depleted uranium can be further fissioned by low energy spallation neutrons ( $> 1 \, MeV$ ).

## 4.2 The spallation target

The function of the spallation target in the ADS is to convert the incident high energy particle beam to low energy neutrons. Its main characteristics can be summarized as follows:

• **High neutron production efficiency** (high spallation neutron yield), which determines the requirement in terms of the accelerator power (i.e. current and energy of the incident proton beam). The number of emitted neutrons varies as a function of the target nuclei and the energy of the incident particle, reaching saturation around 2 *GeV* as shown in Figure 6.

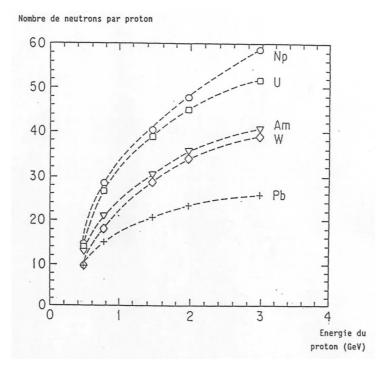


Figure 6: Variation of the number of emitted neutrons as a function of incident proton energy for different thin targets.

Deuterons and triton projectiles produce more neutrons than protons in the energy range below 1-2~GeV. However, the high yield of neutrons among the low energy deuterons and tritons can easily contaminate the low-energy part of the accelerator with radioactivity from these spilled charged particles. Figure 7 shows the neutron yield when deuterons or tritons are injected into a thick uranium target.

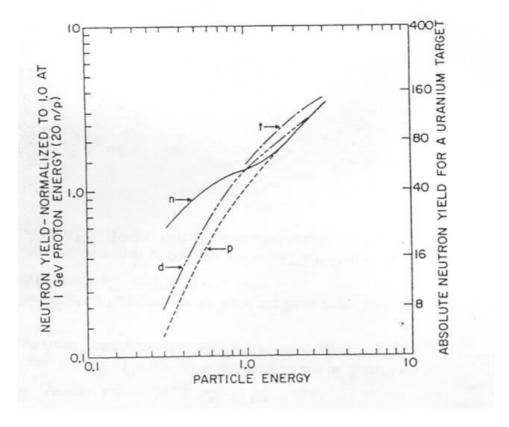


Figure 7: Neutron yield vs. energy for proton, deuteron, triton and neutron particles.

• Damage and activation of the structural components (beam window and target) must be kept at tolerable values for a safe and low hazard operation (reliable and low maintenance operation). The energy distribution of spallation neutrons evaporated from an excited heavy nucleus bombarded by high energy particles is similar to that of fission neutrons, slight shifting towards higher energies ( $E_n \approx 3 - 4 \, MeV$ ) as illustrated in Figure 8.

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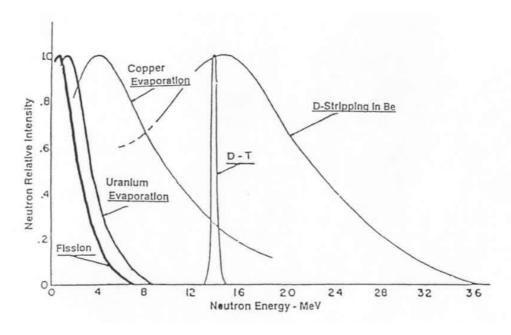


Figure 8: Variation of the energy distribution of emitted neutrons for different nuclear reactions.

3. Heat deposition in the beam window and target, which determines the thermal-hydraulic requirements (cooling capabilities and nature of the target), must be consistent with a high power operation of the order of 1 to 10 MW. An example of the heat deposition of a proton beam in a lead target is shown in Figure 9. Increasing the energy of the incident particle affects considerably the power distribution in the lead target. Indeed one can observe that, while the heat distribution in the axial direction extends considerably as the energy of the incident particle increases, it does not in the radial direction, which means that the proton tracks tend to be quite straight. Heat deposition is largely contained within the range of the protons. But while at 400 MeV the energy deposit is exactly contained in the calculated range (16 cm), this is not entirely true at 1 GeV where the observed range is about 9% smaller than the calculated ( $r_{calc} = 58$  cm,  $r_{obs} \sim 53$  cm). At 2 GeV the difference is even more relevant ( $r_{calc} = 137$  cm,  $r_{obs} \sim 95$  cm). This can be explained by the rising fraction of nuclear interactions with increasing energy, which contribute to the heat deposition and shortens the effective proton range.

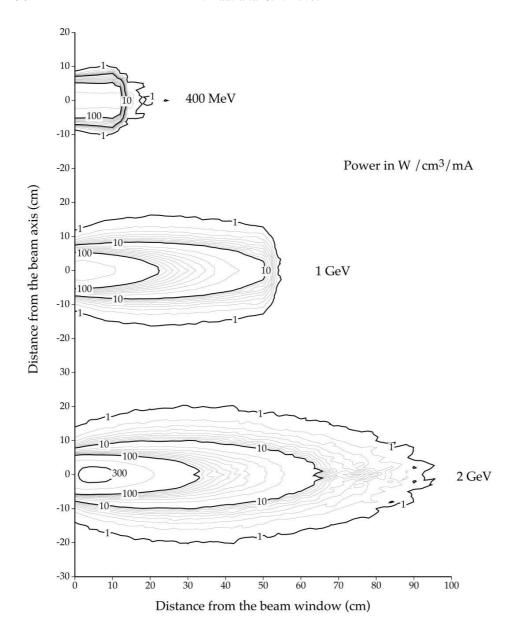


Figure 9: Heat deposition in a beam window and a lead target for various incident proton energies. The calculations take into account not only the electromagnetic interactions, but all kinds of nuclear reactions induced by both protons and secondary particles (including neutrons down to 20~MeV) and  $\gamma$ .

• The contribution of the spallation products to the waste stream must be very small. The spallation product distribution varies as a function of the target material and incident proton energy. Figure 10 shows that it has a very characteristic shape: At high masses it is characterized by the presence of two peaks corresponding to (i) the initial target nuclei and (ii) those obtained after evaporation. Three very narrow peaks corresponding to the evaporation of light nuclei such as (deuterons, tritons, <sup>3</sup>He and α). Finally an intermediate zone corresponding to nuclei produced by high-energy fissions

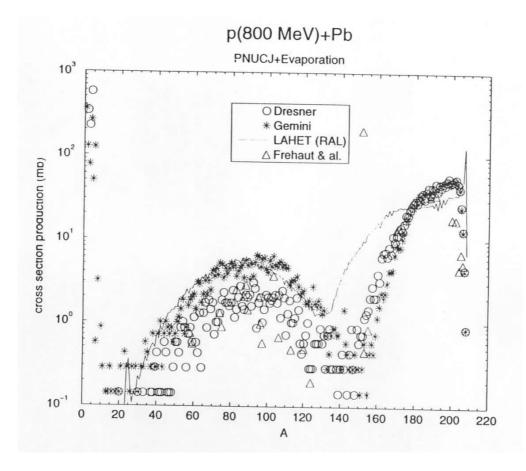


Figure 10: Spallation product distribution from 800 MeV protons impinging on a thick lead target. Simulations carried out with different high energy transport codes are compared with measurements by Frehaut et al. Large discrepancies are observed for mass numbers ranging between 140 and 160 and those corresponding to nuclei produced by high energy fission.

It is believed today that molten lead or lead-bismuth eutectic (LBE) are the best choices to meet most of these requirements. A significant problem with LBE, however, is the production of radioactive and highly mobile polonium from high-energy proton and neutron reactions on bismuth. Lead, on the other hand, has a much reduced polonium production, but higher operating temperatures.

#### 4.3 The sub-critical core

In an ADS, the sub-critical blanket surrounding the spallation target can multiply the spallation neutrons. In quantitative terms, the total number of fissions  $N_{fiss}$  in the sub-critical assembly can be expressed by:

$$N_{fiss} = N_h \Gamma_h \frac{k_{eff}}{\left(1 - k_{eff}\right)} v$$

where  $N_{fiss}$  is the total number of fissions,  $N_h$  the total number of fissions by high-energy proton reactions,  $\Gamma_h$  the number of neutrons produced by high-energy reactions per fission in the blanket,  $\nu$  is the number of neutrons per low-energy fissions and  $k_{eff}$  is the multiplication factor for low-energy fission neutrons. Hence, by increasing the  $k_{eff}$  value of the sub-critical core one can reduce the proton current required to incinerate the waste.

The important issue in designing an ADS is its inherent sub-criticality and stability of reactivity. This feature can significantly improve the safety of an ADS. Contrary to a critical reactor (Figure 11), an ADS operates in a non self-sustained chain reaction mode, which minimizes criticality and power excursions.

The ADS is operated in a sub-critical mode and remains sub-critical whether the accelerator is on or off, providing thus an extra level of safety against criticality accidents.

The accelerator provides a control mechanism for sub-critical systems, which is by far more convenient than control rods in critical reactors. Control rods are not only a safety concern, they degrade the neutron economy of the system as well. Good neutron economy is crucial for ADS since it determines the power and consequently the cost of the accelerator.

The ADS provides a decoupling of the neutron source (spallation source) from the fissile fuel (fission neutrons). It accepts fuels that would not be acceptable in critical reactors, such as Minor Actinides, fuels with a high Pu content, and even long-lived fission fragments (e.g. <sup>99</sup>Tc and <sup>129</sup>I).

ADS Design

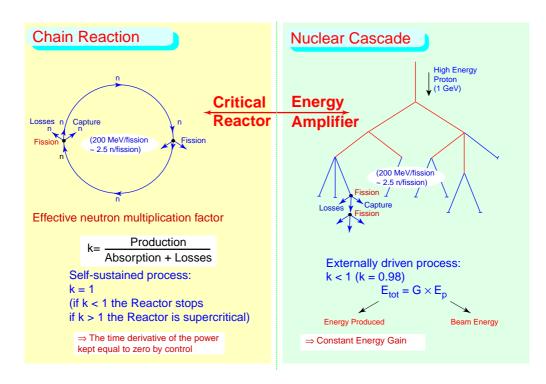


Figure 11: Illustration of the nuclear cascade that drives an ADS as opposed to the self sustained chain reaction driving a critical fission reactor.

• The neutron multiplication factor. The main parameter characterizing the neutron economy of an accelerator driven sub-critical fission device is the factor M by which the "source" spallation neutrons are multiplied by the fission dominated cascade. A related quantity is the multiplication coefficient  $k_{src} = (M-1)/M$ , that is the average ratio of the neutron population in two subsequent generations of the source-initiated cascade.

Such a factor  $k_{src}$ , depending on both the properties of the source and of the medium, is in general conceptually and numerically different from the effective criticality factor  $k_{eff}$ , commonly used in reactor theory, which is in fact only relevant to the fundamental mode of the neutron flux distribution, and is independent on the source.

The effective criticality factor  $k_{eff}$  is however a meaningful measure of the actual safety characteristics of the device, that is 1-  $k_{eff}$  is a proper gauge of the distance from criticality.

In the classic theory of one group diffusion for the uniform reactor, we can write the neutron transport equation as:

$$\frac{1}{v}\frac{d\psi}{dt} = D\left(\nabla^2 + B^2\right)\psi(x,t) \tag{1}$$

If we suppose to factorise the solution as  $\psi(x,t) = \phi(x) \varphi(t)$ , we are left with a classical eigenvalue problem. The eigenvalues that give the correct boundary conditions are:

$$B^{2} - n\frac{\pi^{2}}{a^{2}} = \frac{v\Sigma_{f} - \Sigma_{a}}{D} - n\frac{\pi^{2}}{a^{2}}$$

where a is the size of the reactor. To every value of n corresponds an eigenfunction. The time dependent part is then:

$$\frac{1}{vD}\frac{d\varphi(t)}{dt} = (B^2 - n\frac{\pi^2}{a^2})\varphi(t)$$

and to every value of *n* corresponds a time component:

$$\varphi_n(t) = e^{Dv(B^2 - n\pi^2/a^2)t}$$

If  $B^2 < \pi^2 a^{-2}$ , the reactor is sub-critical and the fluence dies away. If  $B^2 > \pi^2 a^{-2}$ , the reactor is supercritical and it diverges. In case the reactor is not critical, an associated critical reactor is defined where  $B_0^2$  is defined as

$$B_0^2 = \frac{(v/k_{eff}) \Sigma_f - \Sigma_a}{D} = \frac{\pi^2}{a^2}$$

which acts as a definition of  $k_{eff}$ . This is the correction that we have to apply to the average number of neutrons produced per fission to make the reactor critical. Solving for  $k_{eff}$  we have:

$$k_{eff} = \frac{v \Sigma_f}{\Sigma_a + D \pi^2 / a^2}$$

where  $k_{eff}$  can be interpreted as the number of fission neutrons produced for each neutron absorbed. If the system is in the eigenstate relative to  $B^2$  and  $\underline{\text{not}}\ B_0^2$  then the net multiplication factor due to fission is:

$$M_{eff} = \frac{1}{1 - k_{eff}}$$

This simple theory can be generalised in different ways to a more realistic situation, but two aspects are neglected since the start, i.e. the nonfission multiplicative processes and the possible presence of an external source. The nonfission multiplication could be taken into account as a modification of  $\Sigma_a$ , and still the previous development would hold.

In this development  $k_{eff}$  is an intrinsic property of the system. If the fluence distribution is not an eigenstate of the operator, the net multiplication factor will be different, but this will not change the value of  $k_{eff}$ . We can still define formally a value of k as  $k_{src} = 1 - 1/M_{src}$  but it will depend on the fluence as well as on the system. In particular, in the presence of an external source, this value will depend on the position and spectrum of the source neutrons. We will indicate hereon with  $k_{src}$  the value of k calculated from the net multiplication factor  $M_{src}$  in the presence of an external source.

• Neutron source importance. By definition a constant power operation requires  $v/k_{eff}$  neutrons per fission, which means that an external source has to provide a number of neutrons per fission which is

$$\mu_{eff} = \nu \left(\frac{1}{k_{eff}} - 1\right) = \frac{\nu}{M_{eff} - 1}$$

if they are distributed exactly as the eigenfunction of the stationary problem. In the case of an arbitrary external source, this number becomes:

$$\mu_{src} = v \left( \frac{1}{k_{src}} - 1 \right) = \frac{v}{M_{src} - 1}$$

The ratio is known as the importance of source neutrons:

$$\frac{\mu_{eff}}{\mu_{erc}} = \frac{(1 - k_{eff})/(k_{eff}/\nu)}{(1 - k_{erc})/(k_{erc}/\nu)} = \varphi *$$
 (2)

The operational safety margin on  $k_{\infty}$  for an ADS with multiplication M (Figure 12), can be written as

$$\Delta k_{\infty}^{\text{crit}}(M) = k_{\infty}(M) \frac{\varphi^*}{M-1}$$

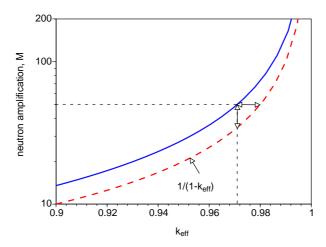


Figure 12: Neutron amplification versus the criticality factor  $k_{\it eff}$ . The dashed line is the amplification that one would compute using  $k_{\it eff}$  instead of  $k_{\it src}$ . The vertical and horizontal arrows indicate, respectively, the difference in the amplification at given  $k_{\it eff}$  and the difference in  $k_{\it src}$  (and hence in the safety margin) at given amplification, due to the distinction between the two criticality factors,  $k_{\it src}$  and  $k_{\it eff}$ .

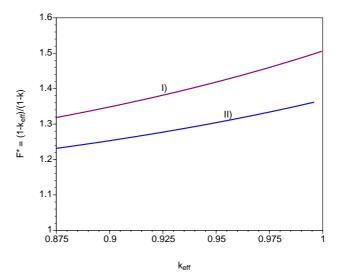


Figure 13: Function  $\varphi^*$ , ratio of the actual safety margin to  $(1 - k_{src})$  vs  $k_{eff}$ , as computed by a simple diffusive model for two cases with geometry shematizing that of a typical ADS: I) the fissile core is surrounded by a breeder; II) the core is surrounded by a diffuser. In both cases  $\varphi^*$  is well above unity and grows with  $k_{eff}$ .

In general, it is found that  $\varphi^*$  grows with k (Figure 13). At a given k,  $\varphi^*$  increases with:

- 4. the "containment" of the neutron source;
- 5. the ratio of the neutron diffusion length to the size of fissile core;
- 6. the presence of an absorbing medium, "enclosing" the fissionable core, which, in a sense, limits the "widening" of the neutron flux distribution as *k* is increased.
- **Neutron Spatial Distribution**. While the neutron distribution inside a reactor is determined primarily by the boundary conditions, in an ADS the geometry of the initial high energy cascade is dominant. The two spatial distributions are expected to differ substantially (Figure 14). The flux distribution is of fundamental importance in order to determine the generated power distribution and the uniformity of the burning of the fuel, both of major relevance when designing a practical device.

To compute the neutron flux and neutron current we use diffusion theory, according to which the current is given by

$$J = -D\nabla \phi$$

where  $D=l_{tr}/3$  is the diffusion coefficient,  $l_{tr}$  is the neutron transport mean free path, given by  $l_{tr}=(\Sigma_t-\overline{\mu}\ \Sigma_s)^{-1}$ , where  $\Sigma_t$ ,  $\Sigma_s$ , and  $\Sigma_a$ , are respectively the macroscopic total cross section, the scattering cross section and the absorption cross section, and  $\overline{\mu}$  is the average value of the cosine of the scattering angle in the laboratory system . Since in an EA the fuel is cooled (and the neutrons diffused and moderated) by a high-Z material, then one can take  $l_{tr}\cong \left(\Sigma_a+\Sigma_s\right)^{-1}$ .

The neutron flux is the solution of the equation

$$\nabla^2 \phi + B_M^2 \phi + \frac{C}{D} = 0$$

where C is the contribution of the external source (neutrons per unit volume and unit time),  $B_M$  is the so-called material buckling

$$B_{\rm M}^2 = \frac{k_{\infty} - 1}{L^2}$$

 $k_{\infty}$  and L are, respectively, the infinite multiplication coefficient and the diffusion length:

$$k_{\infty} = \frac{\nu \Sigma_{\rm f}}{\Sigma_{\rm a}}$$
 ,  $L = \sqrt{\frac{D}{\Sigma_{\rm a}}}$ 

 $\nu$  is the average fission multiplicity, and  $\Sigma_a$  is the macroscopic cross section.

As it is well known, if we consider a finite, system, with vanishing flux at the (extrapolated) boundaries, and a source also vanishing at and outside the boundaries, we can write the solution in terms of the eigenvectors  $\psi_n$  of the characteristic "wave equation"

$$\nabla^2 \mathbf{w} + \mathbf{B}^2 \mathbf{w} = 0$$

which form a complete orthonormal basis, each eigenvector  $\psi_n$  corresponding to an eigenvalue  $B_n$ . We normalize the eigenfunctions in such a way that  $\int \psi_n^2 \, \mathrm{d} V = 1$ ; and introduce the volume integrals of the eigenfunctions:

$$\Psi_n = \int_V \psi_n(\mathbf{x}) dV$$

We then write the (known) outer source as

$$C(\mathbf{x}) = \sum_{n=1}^{\infty} c_n \psi_n(\mathbf{x})$$

with the expansion coefficients given by

$$c_n = \int_V C(\mathbf{x}) \psi_n dV$$

so that the space integrated source neutron rate can be written as

$$Q = \int_{V} C(x) \ dV = \sum_{n=1}^{\infty} c_n \Psi_n$$

The (unknown) neutron flux can be expanded in the same basis, too,

$$\phi(x) = \sum_{n=1}^{\infty} \phi_n \psi_n(x)$$

and a straightforward solution is found for a homogeneous medium. Indeed, in this case, by substituting the expansions for the source and the flux we obtain an independent equation for each n, giving the coefficient of the flux as a function of that of the source :

$$\phi_n = \frac{1}{\Sigma_a} \frac{c_n}{1 - (k_\infty - B_n^2 L^2)} = \frac{c_n}{\Sigma_a (1 + B_n^2 L^2)} \frac{1}{1 - k_n}$$

where

$$k_n = \frac{k_{\infty}}{1 + B_n^2 L^2}$$

As anticipated in the introduction, we see that if all  $k_n$ 's are smaller then unity, then the flux is given by a linear superposition of eigenmodes; as soon as  $k_1$ =1 the system becomes critical; the source is no more needed to sustain the system, and the only surviving mode is the fundamental one.

If  $\frac{k_{\infty} - 1}{L^2} > 0$ , the solutions are of sinus form:

$$\Psi_{lmn} = \sqrt{\frac{8}{abc}} \sin \pi \frac{lx}{a} \cdot \sin \pi \frac{my}{b} \cdot \sin \pi \frac{nz}{c};$$

If 
$$\frac{k_{\infty}-1}{L^2} = -\gamma^2$$
, the solutions are of exponential form: 
$$\begin{cases} \Psi(x) = A_1 e^{-x} + B_1 e^{x} \\ \Psi(y) = A_2 e^{-x} + B_2 e^{x} \end{cases}$$
$$\Psi(z) = A_3 e^{-x} + B_3 e^{x}$$

#### SPATIAL DISTRIBUTION OF THE NEUTRON FLUX

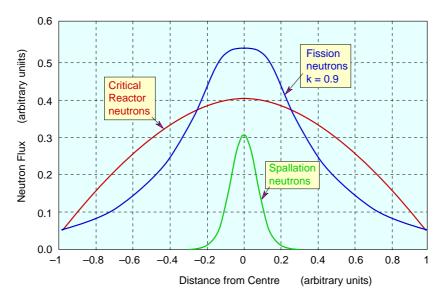


Figure 14: Spatial distribution of the neutron flux depending on the value of k.

• **Neutron Source "Amplification"**. In an accelerator driven, sub-critical fission device, like the Energy Amplifier (EA) [1], the "primary" (or "source") neutrons produced via spallation by the interaction of the proton beam with a suitable target, initiate a cascade process. The source is then "amplified" by a factor  $M^2$  and the beam power is "amplified" by a factor  $G = G_0 M$ .

If we assume that all generations in the cascade are equivalent, we can define an average criticality factor k (ratio between the neutron population in two subsequent generations), so that

$$M = 1 + k + k^2 + k^3 + \dots = \frac{1}{1 - k}$$

and then G can be computed from k, according to

$$G = \frac{G_0}{1-k}$$

The  $G_0$  constant contains the spallation process information (position and energy distribution of the spallation source), such that:  $G_0 \sim 3$  for uranium and  $\sim 2.7$  for lead for instance (typical for protons with kinetic energy of 1 GeV, as predicted by simulations [17] and confirmed by the FEAT experiment [9]).

In the case of the Energy Amplifier, a 1 GeV proton incident on a lead target produces ~ 30 spallation neutrons, which in turn produce (for k = 0.98, and 1/1-k = 50)  $30 \times 50 = 1500$  neutrons from fission.

Typically, 40% of these neutrons also produce fissions (600 fissions, the remaining 900 neutrons are captured or escape the system), the energy produced in the EA is then:  $600\times200~(MeV/fissions) = 120~000~MeV \equiv 120~GeV$ , and the energy gain is : G = 120/1 = 120. We check that  $G_0 = G\times(1-k) = 120\times0.02 = 2.4$  for lead.

If  $\eta_{th\rightarrow el} \sim 45\%$ , the corresponding electric energy is  $120\times0.45 \sim 54$  GeV, i.e. more than sufficient to drive the proton accelerator which uses 2 GeV of electric energy (assuming a 50% efficiency in converting electric energy to proton kinetic energy).

This aspect has been studied in the First Energy Amplifier Test (FEAT) experiment [9] at CERN where it was shown that this energy gain is well understood and that, not only is it independent of the proton beam intensity, but it

<sup>&</sup>lt;sup>2</sup> The quantity M measures the multiplication of the source neutrons by the cascade process. Since, on the other hand the term multiplication is usually employed with a different meaning in reactor theory (where the infinite multiplication factor  $k_{\infty}$ , and the effective multiplication factor  $k_{eff}$  are introduced), here we refer to M as to the "neutron source amplification factor", and to k as to the "criticality factor".

is also independent of the beam kinetic energy if above about 900 MeV as shown in Figure 15.

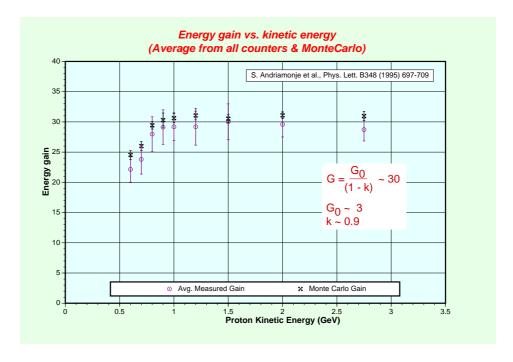


Figure 15: Average energy gain measured in the FEAT experiment carried out at CERN as a function the proton beam kinetic energy.

• The FEAT Experiment. The core of the concept of the First Energy Amplifier Test (FEAT) experiment [9] is the production of substantial amounts of energy, over and above the kinetic energy brought in by the accelerator beam. From that stems the concept of an "energy gain" G. In conditions of practical interest, the gain is predicted to be G = 30 to 60, which, taking into account the relevant efficiencies, was easily shown to be much more than what is needed to power the accelerator.

The main purpose assigned to the test is therefore to ascertain that there is such a gain and that its magnitude is in agreement with the value predicted by the simulation (Figure 15).

• The beam intensity that we used (of the order of 10<sup>8</sup> protons/second) was much smaller (by five orders of magnitude) than the one normally delivered by the CERN Proton Synchrotron (PS);

- The power produced during the test was 1 Watt, i.e. nine orders of magnitude less than that of a fully fledged 1000 MW EA unit necessitating a dedicated high intensity accelerator (typically, a few mA of proton beam at 1 GeV);
- The total energy release in the volume of the assembly is calculated by taking the heat release measured at the different points by the thermometers and the variation with distance of the energy release to perform the integration over the volume. The thermometers register the complete energy release not only from fission fragments but also from  $\gamma$ s following neutron capture and from radioactive decays.

A schematic representation of the FEAT assembly is shown in Figure 16.

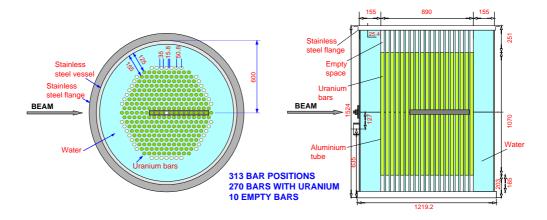


Figure 16: Top and side views of the FEAT assembly along the T7 beam line from the CERN/PS.

The test was performed with an existing sub-critical assembly of natural uranium and water. It consists of small cylindrical rods of natural uranium metal, with aluminum claddings, immersed in ordinary water which has the function of moderator (Figure 17). We note that with such choices of moderator and target, the device can never become critical. Its "infinite multiplication factor" is  $k_{\infty}$ = 0.97.

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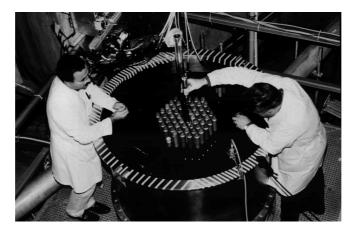


Figure 17: Picture of the FEAT assembly with its 3.6 tons of natural uranium.

The neutronic behavior of the assembly has been calibrated with the help of a 58 GBq neutron source (Am-Be) inserted in the centre of the device. The neutron flux measured with a boron loaded counter is shown in Figure 18, and confirms the expected exponential behavior as a function of the distance from the source. We find a neutron multiplication factor for a point-like centred source of  $k = 0.915 \pm 0.010$ . This is in good agreement with EAMC calculations which give  $k = 0.920 \pm 0.005$ .

The comparison indicates that one cannot use the "critical reactor" formalism to describe a sub-critical system, since all the ortho-normal modes of the "buckling" equation representing the neutron flux distribution must be evaluated, and not only the fundamental mode. The use of the fundamental mode alone, results in an underestimation of the neutron multiplication factor, k, since the escape probability is enhanced by the "cosine-like" distribution of the fundamental mode with respect to the real distribution which is exponential. In particular, the data in Figure 18 show that Monte Carlo calculations carried out in the "reactor mode" with MCNP-4B [18] give large disagreements with measurements (e.g.,  $k = 0.868 \pm 0.002$ ).

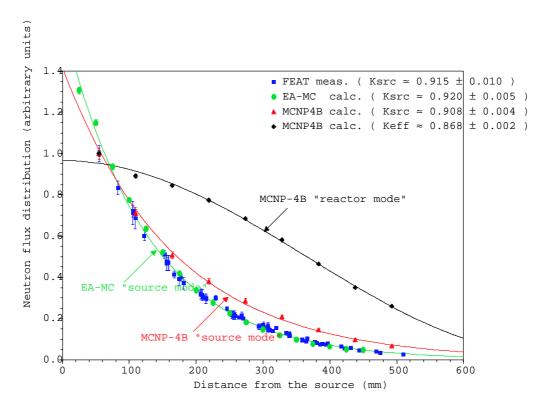


Figure 18: Comparison of the neutron flux distribution between measured data and Monte Carlo calculations carried out (i) in the "source mode" with EAMC and MCNP-4B, and (ii) in the "reactor mode" with MCNP-4B.

## 5 The Energy Amplifier

The Energy Amplifier is a sub-critical, fast neutron system, driven by a proton accelerator (Figure 19). A complete description of all the features of the EA can be found in Ref. [1]. One of the main characteristics is the presence of  $10^4$  tons of molten lead used as a target for the protons to produce neutrons by spallation, as a neutron moderator, as a coolant to extract heat by natural convection, and as a radioactivity containment medium.

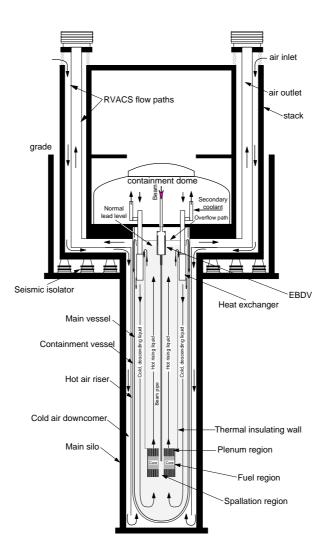


Figure 19: Schematic of the  $1500 \, MW_{th}$  Energy Amplifier standard unit [1]. The main vessel is about  $25 \, m$  high and  $6 \, m$  in diameter. The proton beam is injected vertically, through a vacuum pipe to produce spallation neutrons at the level of the core.

# 5.1 Why fast neutrons?

Lead was chosen as the neutron moderator to obtain the hardest possible neutron energy spectrum. This is dictated by the need to optimize the fission probability of

TRU. Indeed, in the fast neutron flux provided by the EA all TRU can undergo fission, a process which eliminates them, while in a PWR thermal neutron flux many TRU do not fission and thus accumulate as waste (Figure 20).

In addition, as the capture cross section of neutrons on FF is smaller for fast neutrons than for thermal neutrons (Figure 21), and since neutron capture on FF is the main limitation to long burnups, in a fast neutron system the efficiency with which the fuel can be used will be much higher than in a PWR. Typically, it is hoped to reach burnups of  $150~GW \times day/t$  (a larger burnup of  $200~GW \times day/t$  was already achieved in the fast EBR2 reactor at Argonne National Laboratory).

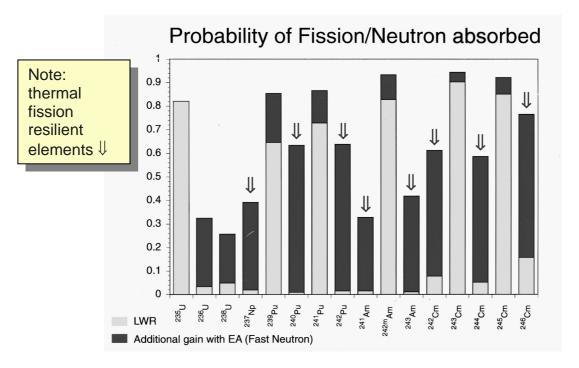


Figure 20: Comparison of fission and capture probabilities of actinides for thermal and fast neutron fluxes. In contrast to a thermal neutron flux, in a fast neutron flux, all TRU can fission.

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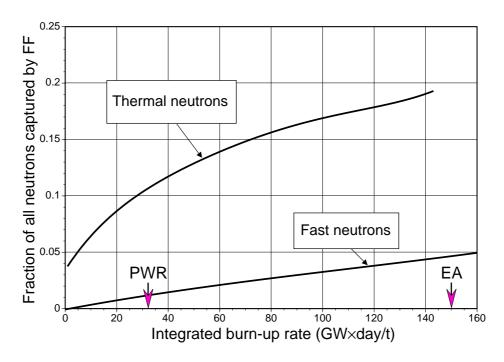


Figure 21: Fraction of neutron captures on fission fragments (FF) for thermal and fast neutron fluxes, as a function of burnup. The maximum burnup for a PWR and the typical burnup which one hopes to achieve with an EA are indicated.

# 5.2 Sub-criticality and the accelerator

The proposed system [1] has a neutron multiplication coefficient (k) of 0.98. The sustainability of the nuclear fission reactions is made possible because of the presence of an external source of neutrons provided by the proton beam. The working point is far below criticality, which ensures that the system remains subcritical at all times, implying that, by construction, accidents of the Chernobyl type are impossible (Figure 22). The traditional  $k_{eff}$  of the system itself (with beam turned off) is even smaller than k (approximately 0.97). The energy amplification in the system, defined as the ratio between the energy produced in the EA and the energy provided by the beam, can be parametrized as  $G_0/(1-k)$ , where  $G_0$  is a constant characterizing the spallation process. This aspect of the system has been studied in the First Energy Amplifier Test (FEAT) experiment [9] where it was shown that this energy gain is well understood and that, not only is it independent of the proton beam intensity, but it is also independent of the beam kinetic energy if above about  $900 \ MeV$ .

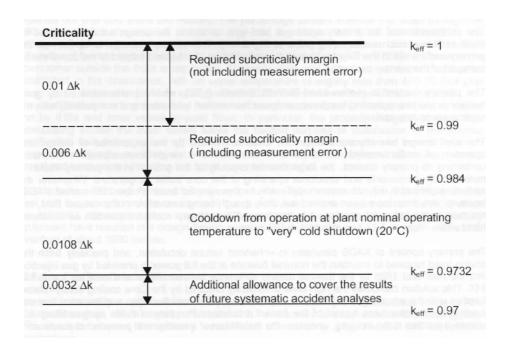


Figure 22: Sub-criticality conditions for a safe operation of the EA.

This fortunate feature means that the accelerator can be of relatively modest size (Figure 23). Experts agree that present accelerator technology can provide the required beam power (10 to 20 mA at 1 GeV) with either linac or cyclotron solutions [19]. Examples already exist of suitable high-power accelerators which are planned or have been considered in various parts of the world:

- the PSI (Switzerland) cyclotron now running at 1.4 mA, 590 MeV, 0.826 MW [20];
- the proton linac for the Los Alamos Neutron Science Center (LANSCE) running up to 1.5 mA, 0.8 GeV and 1 MW of average power [21];
- both the USA and Europe had projects to build linacs to produce tritium: (TRISPAL [22] at CEA (France): 600 MeV, 40 mA, 24 MW and APT [23] at LANL (USA): 1 GeV, 100 mA, 100 MW). Even though tritium is no longer officially on the agenda, accelerator developments are continuing for other applications;
- Japan is also considering a high-intensity proton source as part of their new Neutron Science Project [24].

The system needed to drive an EA represents only a reasonable extrapolation of what has already been achieved in current accelerator technology.

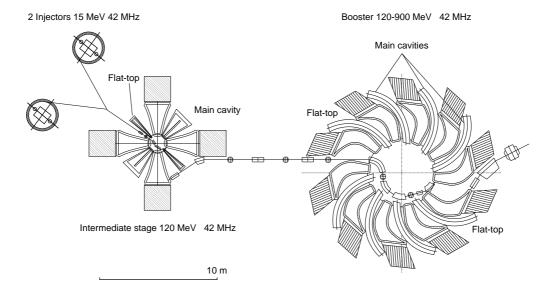


Figure 23: The full, high-intensity, cyclotron accelerator layout proposed to drive a k = 0.98 Energy Amplifier [1].

In practice, the choice of accelerator technology may be coupled with the strategy for the utilization of EA systems. If the main purpose is to destroy waste on a nuclear power plant site, then the cyclotron with its smaller size (Figure 23) has a clear advantage (no need to extend the power plant site, and simpler control and safety of the accelerator, all resulting in better cost effectiveness).

Several other technical advantages can be found in favour of a cyclotron as compared to a linac:

- One should be able to achieve high efficiency (50%), as the current in Radio Frequency (RF) cavities would be about 100 times (100 separated turns) the extraction current, implying that most RF power goes to the beam while copper losses become relatively small. Today, state-of-the-art RF cavities have reached 70% efficiency (mains to RF). The power needed for the magnet and for all other equipment is small compared to the RF power.
- There is no need for Super Conducting (SC) cavities, keeping the technology simple. In a SC linac, niobium-coated cavities, such as those developed at CERN, can be used down to  $\beta \sim 0.7$ . Below that, it is necessary to develop another cavity technology.

- In a warm linac, the efficiency is small and the small aperture is a problem for the beam losses, which in addition are not localized. In a cyclotron, the magnet aperture is relatively large (7 to 8 cm) and the beam losses may only be significant at the beam extraction. An extraction efficiency of ≥ 99.9% is the goal. However, even if losses turn out to be larger than one would hope, they will only activate a limited region of the machine. Most machine elements could still be accessible as soon as the cyclotron comes to a halt (this is presently the case at PSI).
- Reliability may be better than in linacs which need many more control elements (reliability decreases strongly with an increase in the number of parts).

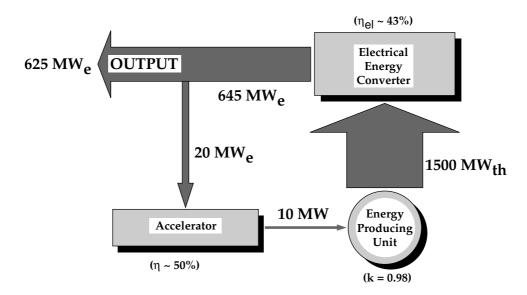


Figure 24: The energy production scheme in the standard EA system as proposed in Ref. [1].

An important achievement of the FEAT experiment was the validation of the innovative simulation of energy amplification in accelerator-driven sub-critical systems developed by the Emerging Energy Technology (EET) group at CERN. This gives confidence in the choice of the main parameters of a system where less than 5% of the electric power needs to be recirculated during its operation (Figure 24).

# **5.3** Target for the protons

The spallation target has to provide the highest possible neutron yield, be transparent to neutrons, and at the same time sustain a large beam power of 10 to 20 MW. In this respect, molten lead is almost an ideal candidate since it has also excellent thermodynamics properties and can participate in cooling. The use of liquid targets is a tendency which is presently developing in the design of spallation neutron facilities (for instance, ESS [25] and SNS [26] are developing liquid mercury targets and SINQ [27] is planning an upgrade to a liquid lead-Bismuth target). Tungsten, although acceptable from the point of view of spallation, is not favourable to neutron transport (neutron absorption and activation) and would clearly have to be used in solid form since its melting temperature is very high (3422 °C) with the additional difficulty that it can break (very brittle above 600 to 700 °C) or even explode if the proton source is pulsed.

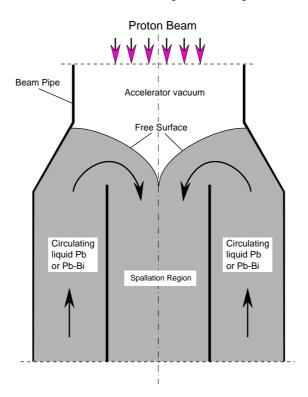


Figure 25: Sketch of a windowless interface between the accelerator and the spallation region. Because of the low vapour pressure of lead, the molten lead can be in direct contact with the beam vacuum. A cold trap type of device (not shown) can capture residual vapours.

From the point of view of neutronics, both lead and eutectic lead-bismuth (Pb-Bi) mixtures are satisfactory. Pb-Bi has the advantage of allowing operation at a lower temperature, and might be chosen in a first stage for the design of an EA demonstrator. The maximum temperature of the window in a 6 mA, 600 MeV beam Pb-Bi system is about 500 °C which can be handled with presently available materials such as ferritic, 9% chromium steel. Going to pure lead would increase that maximum window temperature by about 200 °C, which requires developing new materials through technological R&D.

Because Pb-Bi targets produce significantly more radiotoxic elements (<sup>210</sup>Po) than pure lead, the long-term preferred solution is pure lead. We refer the reader to a discussion of these effects in the first item in Ref. [1] pp. 77 to 82. One assumes that through proper R&D, materials will be developed which can accommodate the high lead temperature including corrosion effects.

The target is presently an area where intense R&D is being carried out in Europe, within the 5<sup>th</sup> Framework Programme of the European Union. The Benchmark Working Group, a collaboration between 16 institutes (see for instance Ref. [28]), is particularly active in this domain. All of this implies a careful design of the interface (window) between the accelerator and the effective target. The very low vapour pressure of lead, makes it possible for liquid lead to be compatible with direct exposure to the accelerator beam pipe vacuum which opens the possibility of a windowless solution for that interface [29] (Figure 25).

## 5.4 Destruction of nuclear waste: TRU

The general strategy consists of using as fuel thorium mixed with TRU as opposed to uranium with plutonium as proposed in fast critical reactors, such as SuperPhenix.

The availability of an external neutron source, thanks to the accelerator, and the availability of a fast neutron energy spectrum, thanks to the choice of lead as moderator, allows the sustained operation of a sub-critical device with wide flexibility in the choice of fuel. For reasons which will become clear later, the preference is for fuels based on thorium rather than uranium. Pure thorium does not fission, but <sup>233</sup>U bred from <sup>232</sup>Th can produce energy through fission. In practice, seeds of fissionable material are needed to provide fissions at the startup of the system, and for this purpose any fissionable element will do: <sup>233</sup>U from a previous EA fuel load, <sup>235</sup>U extracted from natural uranium, military <sup>239</sup>Pu or simply TRU, which is precisely the main component of the waste we wish to destroy. In this way, it is possible, in an EA, to destroy TRU by fission, a process which produces energy and makes the method economically attractive. The energy contained in the TRU in PWR waste is about 40% of the amount extracted in the PWR.

Thorium is an attractive fuel because it exists in relatively large quantities in the Earth's crust (at least five times more abundant than uranium) and it is isotopically pure so that natural thorium can be used in the EA as compared to only the 0.7% of <sup>235</sup>U in natural uranium from which PWR fuel is manufactured. Thorium is about 5 neutron captures away from the TRU one wants to destroy (Figure 26), ensuring that it can more easily work in a mode where it destroys more TRU than it produces (lower equilibrium concentrations for TRU).

# CHOICE OF FUEL: Thorium [ $^{232}$ ThO $_2$ (+ $^{233}$ UO $_2$ )] $n + ^{232}$ Th(1.4×10 $^{10}$ a) $\rightarrow ^{233}$ Th(22.3 m) $\rightarrow ^{233}$ Pa(27 j) $\rightarrow ^{233}$ U(1.6×10 $^{5}$ a) Among the 60% of neutrons not used for fission, 20% are lost and 40% are used to breed 233-U from 232-Th. In this way, new fissile material replaces what is used for fission. $(n, \gamma)$ capture $(n, \gamma)$ capture (n, 2n) (n, 2n) (n, 2n) $(E_n: \geq 6 \text{ MeV})$ Note the difference between 232-Th and 238-U in terms of TRU access!

Figure 26: Chart of the actinides.

It is easy to see why a thorium system would be much more practical than a uranium system for the destruction of TRU. The high equilibrium concentration (15%) of plutonium in uranium type systems (Figure 27a) forces the use of extremely large plutonium enrichment, which would make these systems extremely delicate to operate, while in an EA, equilibrium concentrations of the order of 10<sup>-5</sup> (Figure 27b) naturally ensure a high burning rate for reasonable TRU concentrations.

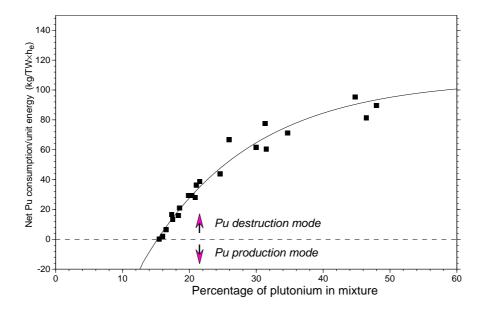


Figure 27a: Net plutonium consumption per unit energy in a uranium-plutonium fast breeder (CAPRA [30]) as a function of plutonium concentration. Note that the unit is  $kg/TW \times h$  electric and not thermal.

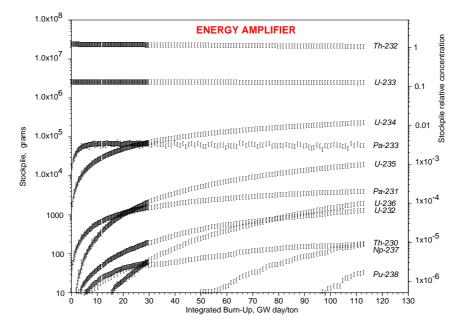


Figure 27b: Evolution as a function of burnup of the stockpile of the main elements present in the EA fuel [1].

A study [31] carried out for the Spanish government, based on a practical example, showed that a 1500  $MW_{th}$  EA could destroy a net amount of 298 kg of TRU per  $GW \times year$  of thermal energy produced. In comparison, a PWR produces 123 kg of TRU per  $GW \times year$ .

It is expected that the reprocessing needed to extract TRU from spent fuel should be much simpler than what is needed to extract plutonium from spent fuel for MOX, as performed, for instance, in the La Hague factory (PUREX process). A pyroelectric reprocessing method [32] developed at Argonne National Laboratory in the United States collects all TRU on a single electrode; this is sufficient since all of them fission in an EA-spectrum flux and they do not need to be separated from one another.

# 5.5 Why not a critical system using thorium

Critical reactors using thorium fuel have worked in the past [33], motivated by the prospect of a high neutron yield per neutron absorbed which <sup>233</sup>U offers over the whole neutron energy range (Table 2), only slightly surpassed by <sup>239</sup>Pu for fast neutrons. However, there is a price to pay for breeding <sup>233</sup>U. It is the production of <sup>233</sup>Pa which has a large neutron capture cross-section and must be compensated by a higher enrichment in fissile material. Also, <sup>233</sup>U fissions produce more <sup>135</sup>Xe (direct yield 1.4 % for <sup>233</sup>U versus 0.3% for <sup>235</sup>U) and samarium precursors (<sup>147</sup>Nd, <sup>149</sup>Pm) than <sup>235</sup>U. These isotopes represent a significant fraction of the total neutron absorption by fission products. At mid-cycle they account for more than 50% of the total fission product absorption.

	FAS'	T NEUTRO	ONS	THERMAL NEUTRONS		
	$^{235}U$	$^{235}U$	<sup>239</sup> Pu	$^{235}U$	$^{235}U$	<sup>239</sup> Pu
$\sigma_{fission}$	3.1	2.2	2.0	530	580	750
σ <sub>capture</sub>	0.3	0.76	0.75	46	99	271
ν	2.51	2.44	2.92	2.50	2.44	2.90
$\eta = \nu \sigma_{f} / \sigma_{a}$	2.29	1.81	2.12	2.3	2.09	2.13
$\sigma_{\rm f}/\sigma_{\rm a}$	0.91	0.76	0.73	0.88	0.81	0.66

	$^{235}U$	$^{235}U$	<sup>239</sup> Pu
β <sub>delay</sub>	0.26	0.65	0.21
βeffective	0.3	0.7	0.23

Table 2: Comparison of the nuclear properties for different fissile isotopes.

In addition, the effective fraction of delayed neutrons ( $\beta_{eff}$ ) of <sup>233</sup>U is less than half of that of <sup>235</sup>U, leading to a smaller safety margin. While this factor is vital to the design of a critical assembly, it is completely unimportant to the design or operation of a driven sub-critical assembly. In a critical system, the effective neutron multiplication coefficient ( $k_{eff}$ ) is maintained equal to one by active control and feedback. The resulting safety of the system is then defined in terms of the probability for the system to become (or not to become) supercritical ( $k_{eff}$ ) as happened in Chernobyl in 1986. The probability of such an accident occurring may be very small, but is not zero. In a sub-critical system, the effective neutron multiplication coefficient is smaller than one by construction. Therefore, the resulting safety aspect is a deterministic one. The system is and remains sub-critical at all times and Chernobyl type accidents are simply impossible.

# Allowed Operational Safety Margin

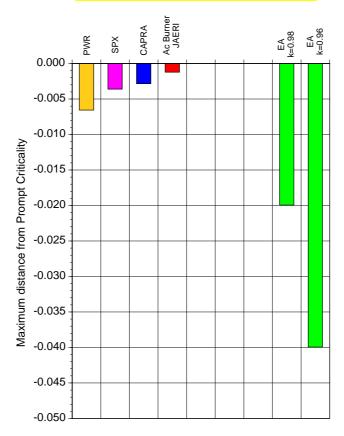


Figure 28: Comparison of the maximum allowable safety margins for minor actinide burning in critical reactors and in ADS.

ADS Design 125

Furthermore, in a critical reactor, whether its fuel is based on thorium or uranium, TRU enriched fuel leads to smaller ( $\beta_{eff}$ ) values, which affects the safety margin (Figure 28), while as already stated, ( $\beta_{eff}$ ) is unimportant for sub-critical assemblies.

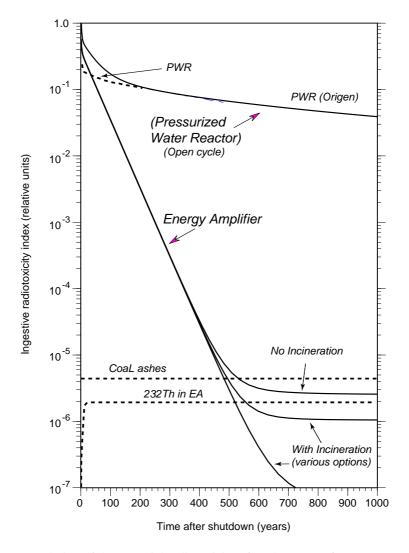


Figure 29: Evolution of the potential radiotoxicity of nuclear waste for PWR, EA and coal burning power station, showing that in the EA, the long-term radiotoxicity can be 4 orders of magnitude smaller than in a PWR in open cycle (adapted from Ref. [34]). The flattening of the curves above 600 years is due to LLFF. Note that the radiotoxicity of spent MOX fuel from a PWR would be about 10 times higher than that of ordinary PWR fuel.

### 5.6 Destruction of nuclear waste: Long-Lived Fission Fragments

In a system such as the EA, where TRU are destroyed, the long-term ( $\geq 500~years$ ) radiotoxicity of the waste becomes dominated by LLFF (Figure 29). This residual level of radiotoxicity could perhaps be tolerated, since it is lower than the level of radiotoxicity of coal ashes corresponding to the production of the same quantity of energy (Table 3). However, since the main LLFF ( $^{99}$ Tc and  $^{129}$ I) can be soluble in water, and therefore, have a non-zero probability over a time-scale of million of years of contaminating the biological chain with hard-to-predict long-term effects, it may be wise to destroy them also.

Radio- Isotope	Half-Life (years)	Mass (kg)	Activity @ 1000 yr (Ci)	Ingestive Toxicity $(Sv) \times 10^3$	Dilution Class A (m <sup>3</sup> )
<sup>129</sup> I	1.57 x 10 <sup>7</sup>	8.09	1.43	19.58	178.47
<sup>99</sup> Tc	2.11 x 10 <sup>5</sup>	16.61	284.29	27.67	947.65
<sup>126</sup> Sn	1.0 x 10 <sup>5</sup>	1.187	33.79	3.20	9.65
<sup>135</sup> Cs	2.3 x 10 <sup>6</sup>	34.12	39.32	9.87	39.32
<sup>93</sup> Zr	1.53 x 10 <sup>6</sup>	26.11	65.64	2.38	18.75
<sup>79</sup> Se	6.5 x 10 <sup>5</sup>	0.30	2.06	0.745	0.59

Table 3: Fission Fragments activity and toxicity after 1000 years of cool-down in a Secular Repository (Values are given for 1 GWe × year).

To provide for this option, Carlo Rubbia proposed to use Adiabatic Resonance Crossing (ARC) [35] (Figure 30). This enhances the neutron capture probability, turning, for instance, a  $2.1 \times 10^5$  year half-life  $^{99}$ Tc into  $^{100}$ Tc that decays quickly ( $t_{1/2} \sim 15.8 \text{ s}$ ) into stable  $^{100}$ Ru. The TARC experiment at CERN [10] showed that using the special (small elastic collision length  $\lambda \sim 3 \text{ cm}$  and small elastic  $\Delta E/E$ ) kinematics of neutrons in pure lead (the most transparent to neutrons of all heavy elements) maximizes the neutron capture probability, making optimum use of prominent resonances in the neutron capture cross-section. Note that  $^{129}$ I and  $^{99}$ Tc, which were studied in TARC represent 95% of the LLFF class A storage volume (see Ref. [31] page 10). The results from TARC imply that one could actually destroy twice as much  $^{99}$ Tc and  $^{129}$ I in the lead in the vicinity of the EA core as is produced over the same time period. The fact that this transmutation can be

carried out parasitically may be an additional incentive to eliminate LLFF, a process which, unlike the elimination of TRU producing energy, does not pay.

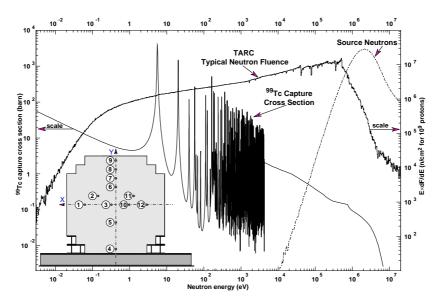


Figure 30: Illustration of the Adiabatic Resonance Crossing principle, showing how the presence of lead transforms the spallation neutron energy distribution into a flat flux distribution of slowing down neutrons, with iso-lethargic steps smaller than the width of cross-section resonances where they will be captured with high probability. A sketch of the 334 ton TARC lead volume is also shown.

## **6** Conclusions

Fundamental research is a strong driving force in innovation and that it can lead to potential solutions of some of the most difficult problems facing our society at the beginning of the third millennium. In particular, nuclear energy could make an important contribution to the solution of the energy problem and it would be a mistake to exclude it, a priori, from R&D.

Present accelerator technology can provide a suitable proton accelerator to drive new types of nuclear systems to destroy nuclear waste or to produce energy.

The Energy Amplifier, based on physics principles well verified by dedicated experiments at CERN, is the result of an optimization made possible by the use of an innovative simulation code validated in these experiments (FEAT and TARC).

An Energy Amplifier could destroy TRU through fission at about twice the rate at which they are produced in LWRs. LLFF such as <sup>129</sup>I and <sup>99</sup>Tc could be

transmuted into stable elements in a parasitic mode, around the EA core, making use of the ARC method.

This experimental programme has generated new applications in various fields: medical applications for which CERN now owns a patent [36], research with the approved CERN nTOF facility [37], and other surprising ideas such as a nuclear engine [38] for deep space exploration. All of these bring additional reward for those who have been involved in this project.

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