

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 subcritical fast neutron system driven by a proton accelerator. It is particularly attractive for destroying, through fission, transuranic elements produced by presently operating nuclear reactors. The EA could also efficiently and at minimal cost transform long-lived fission fragments using the concept of adiabatic resonance crossing (ARC), recently tested at CERN with the TARC experiment. The ARC concept can be extended to several other domains of application (production of radioactive isotopes for medicine and industry, neutron research applications, etc.).

1. Introduction

The research work presented here is an unusual contribution for a laboratory such as CERN, in principle devoted entirely to fundamental research. However, the energy amplifier (EA) [1] is an innovative approach to nuclear energy, and it should come as no surprise that this results from fundamental research, always an engine of innovation. Examples are legion and well known; one of the most recent, the World-Wide Web, was invented at CERN and not by the much more powerful and resourceful computer industry.

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 bound to 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 such a R&D. The present nuclear energy program is meeting growing public opposition in Europe and other parts of the world for three main reasons: (a) the association with military use and the fear of nuclear weapons 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), and (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 comes to the following: 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 measures were not sufficient.

The concept of the energy amplifier (EA) 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 this 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, of a size adapted to

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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 accumulate as a result of the fission of heavy nuclei and can only be destroyed by neutron capture; therefore, different methods will have to be used to eliminate them. As the long-term radiotoxicity of waste (Fig. 1) is clearly dominated by TRU, the EA has been designed to destroy them with the highest efficiency.

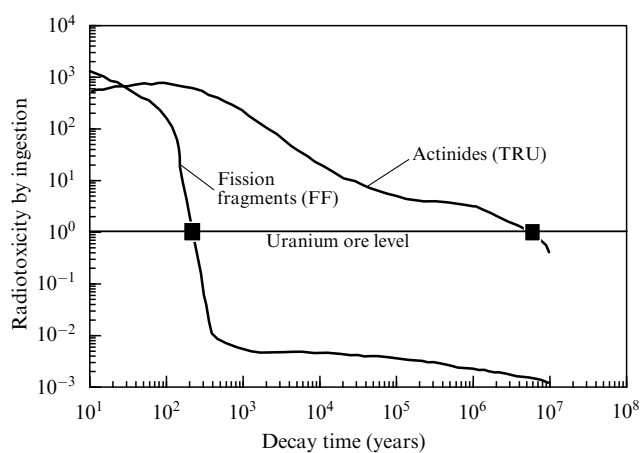


Figure 1. Time evolution of the potential radiotoxicity (relative to uranium ore) of the two main components of nuclear waste for PWR spent fuel, obtained with the ORIGEN2 code.

3. The Energy Amplifier

The energy amplifier is a subcritical, fast neutron system driven by a proton accelerator (Fig. 2). 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 10,000 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.

3.1 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 (see Fig. 3).

In addition, as the capture cross section of neutrons on FF is smaller for fast neutrons than for thermal neutrons (see Fig. 4), and since neutron capture on FF is the main limitation to long burn-ups, 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 burn-ups of 150 GW day/t (a larger burn-up of 200 GW day/t has already been achieved in the fast EBR2 system at Argonne National Laboratory, IL).

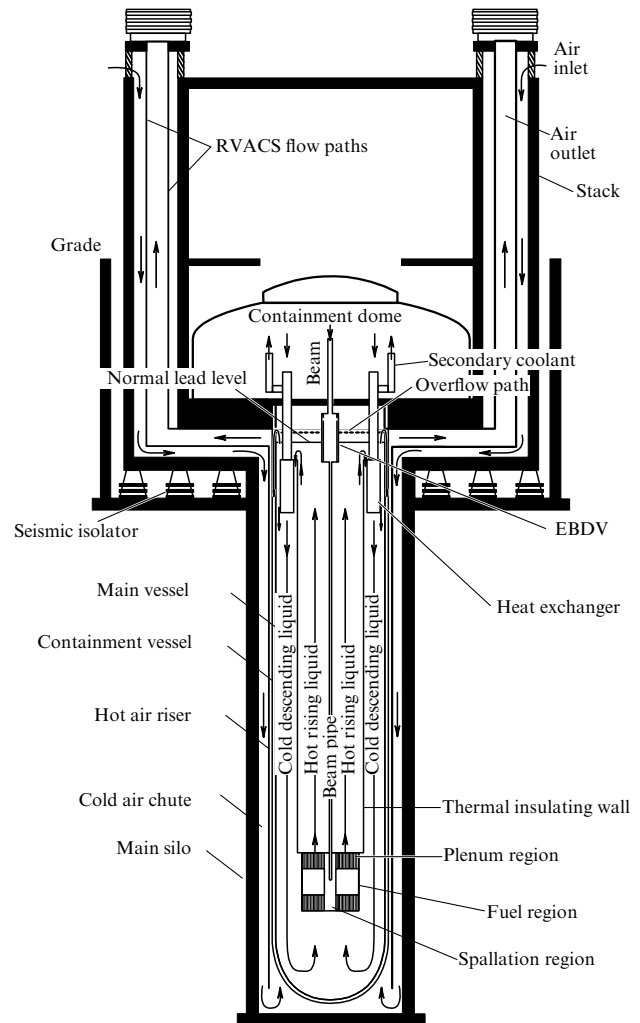


Figure 2. Schematic of the 1500 MW energy amplifier standard unit [1]. The main vessel is about 25 m high and 6 m in diameter. The proton beam is injected vertically through a vacuum pipe to produce spallation neutrons at the level of the core.

3.2 Subcriticality and the accelerator

The proposed system [1] has a neutron multiplication coefficient $k \sim 0.98$. 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. The traditional multiplication factor k_{eff} of the system itself (with the 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 [2] at CERN where it has been 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. This fortunate feature means that the accelerator can be of relatively modest size (see Fig. 5). Experts agree that current accelerator technology can provide the required beam power (10 to 20 mA at 1 GeV)

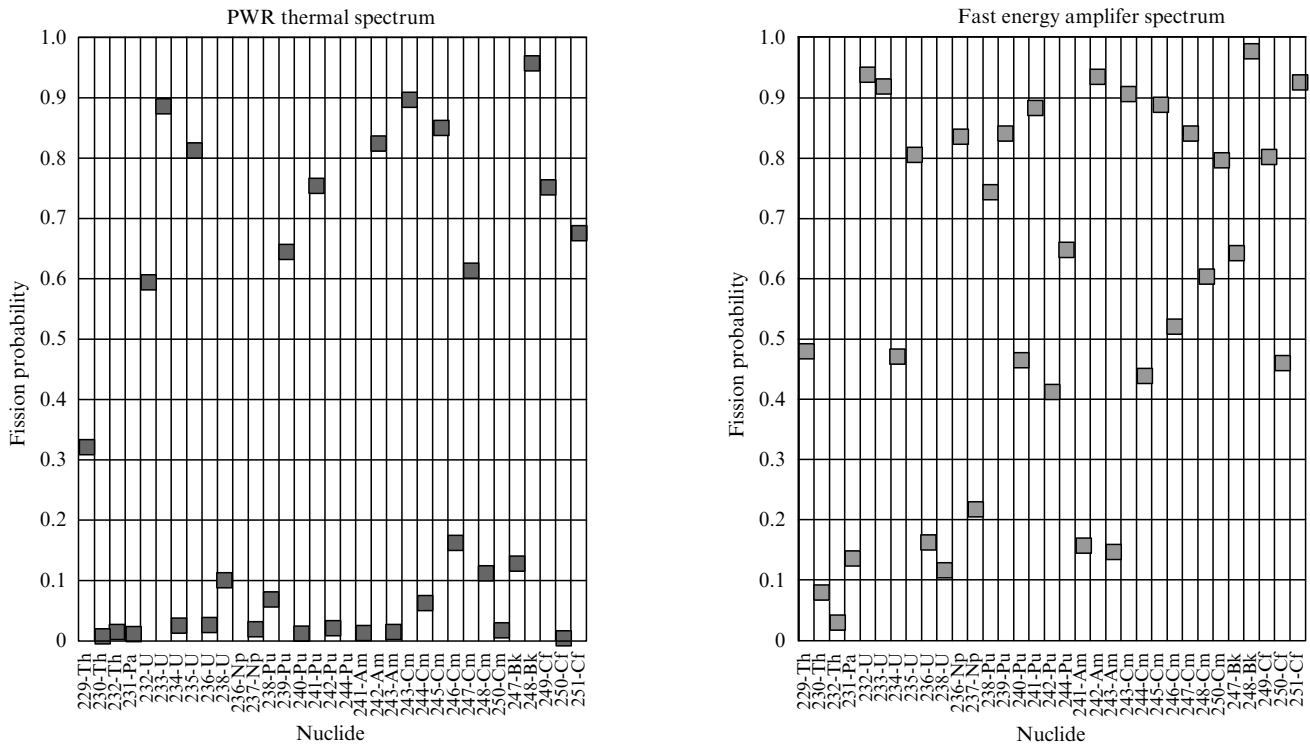


Figure 3. Comparison of fission and capture probabilities of actinides for thermal and fast neutron fluxes. In contrast to a thermal neutron flux, in a fast neutron flux all TRU can fission.

with either linac or cyclotron solutions [3]. Examples of suitable high-power accelerators which are planned or have been considered in various parts of the world already exist:

- the PSI (Switzerland) cyclotron now running at 1.4 mA, 590 MeV, and 0.826 MW [4];
- 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 [5];
- both the USA and Europe had projects to build linacs to produce tritium: (TRISPAL [6] at CEA (France): 600 MeV, 40 mA, 24 MW, and APT [7] 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 [8].

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

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 (Fig. 5) 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 favor 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 superconducting (SC) cavities, keeping the technology simple. In an SC linac, niobium-coated cavities, such as those developed at CERN, can be used down to $\beta = v/c$ of about 0.7. Below that, it is necessary to develop another cavity technology;

— In a warm linac, the efficiency is low and the small aperture is a problem for 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 $\geq 99.9\%$

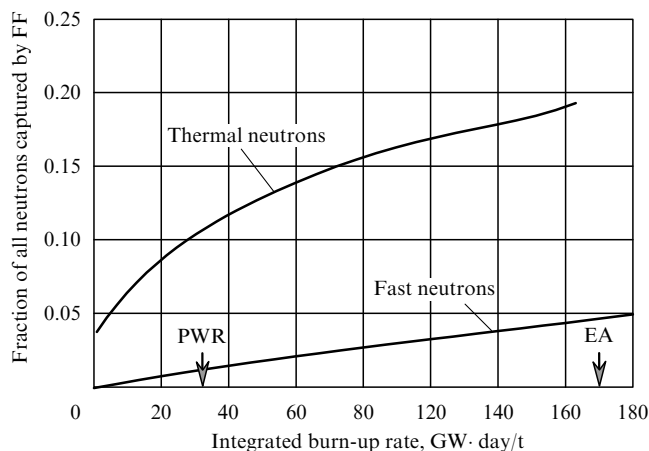


Figure 4. Fraction of neutron captures on fission fragments (FF) for thermal and fast neutron fluxes, as a function of burn-up. The maximum burn-up for a PWR, and the typical burn-up which one hopes to achieve with an EA are indicated (adapted from Ref. [1]).

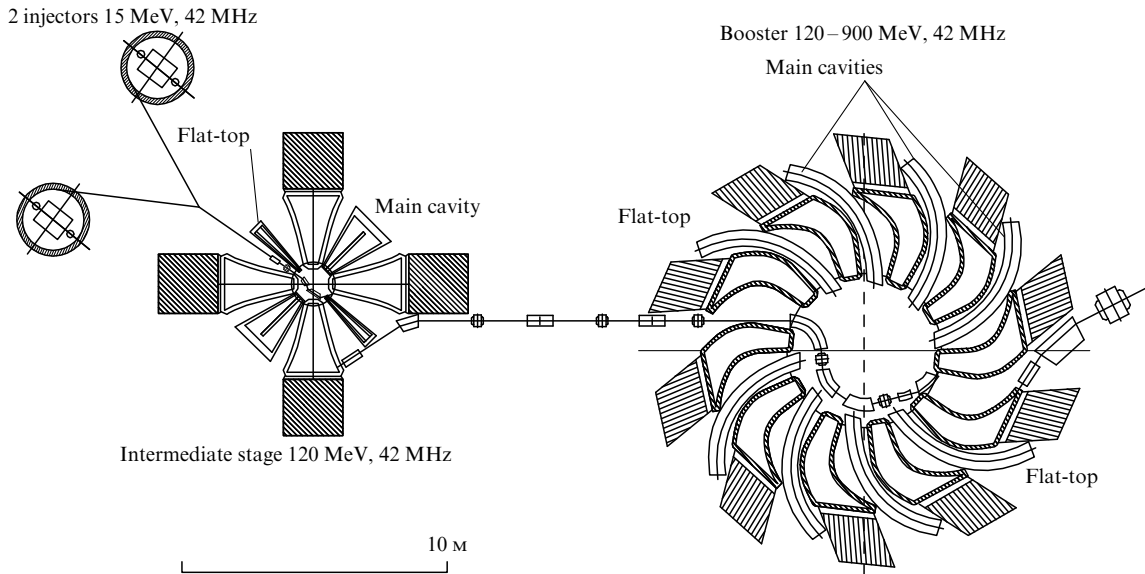


Figure 5. The full, high-intensity, cyclotron accelerator layout proposed to drive a $k = 0.98$ EA [1].

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).

An important achievement of the FEAT experiment was the validation of the innovative simulation of energy amplification in accelerator-driven subcritical 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 (see Fig. 6).

3.3 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 also has excellent thermodynamic properties and can participate

in cooling. The use of liquid targets is a tendency which presently makes itself evident in the design of spallation neutron facilities (for instance, ESS [9] and SNS [10] are developing liquid mercury targets, and SINQ [11] is planning an upgrade to a liquid lead–bismuth target). Tungsten, although acceptable from the point of view of spallation, is not favorable to neutron transport (owing to 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 (it is very brittle above 600 °C to 700 °C) or even explode if the proton source is pulsed.

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 over to pure lead would increase that maximum window temperature by about 200 °C, which requires developing new structural materials through technological R&D.

Because Pb–Bi targets produce significantly more radioactive elements (^{210}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] on pp. 77 to 82. It is anticipated that through proper R&D, materials will be developed which can stand the high lead temperature effects, including corrosion.

The target is presently an area where intense R&D is being carried out in Europe, within the 5th Framework Programme of the European Union. The Benchmark Working Group, a collaboration between 16 institutions (see, for instance, Ref. [12]), 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 vapor 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 [13] (see Fig. 7).

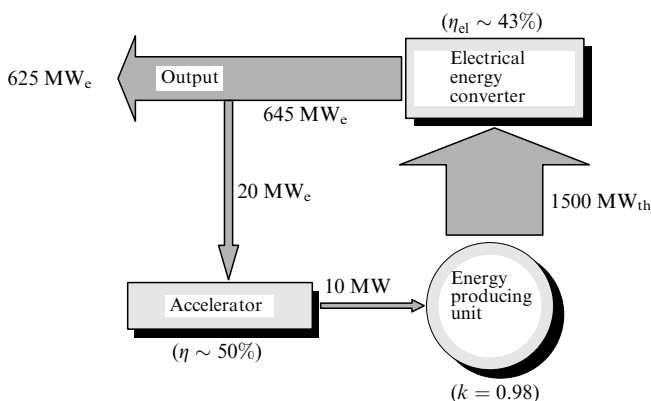


Figure 6. The energy production scheme in the standard EA system as proposed in Ref. [1].

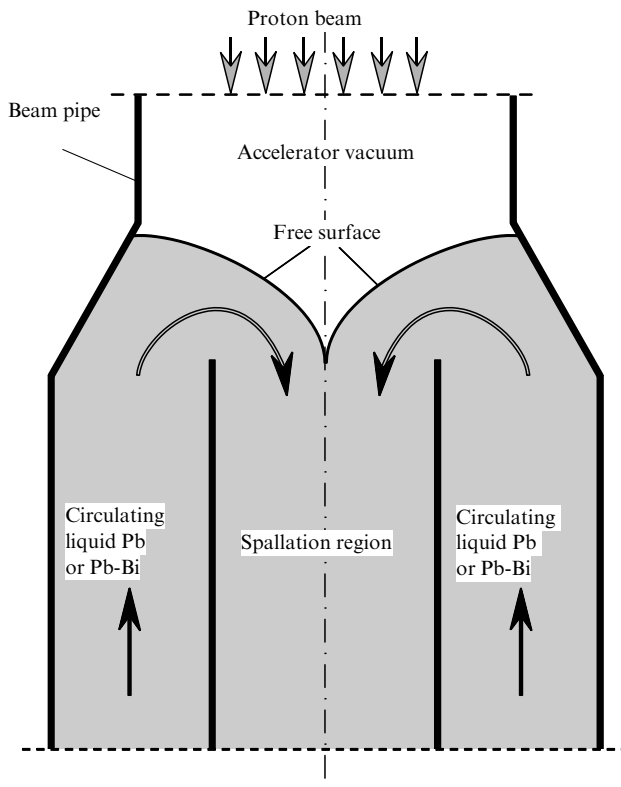


Figure 7. Sketch of a windowless interface between the accelerator and the spallation region. Because of the low vapor pressure of lead, the molten lead can be in direct contact with the beam vacuum. A cold trap type of device (not shown) can capture residual vapors.

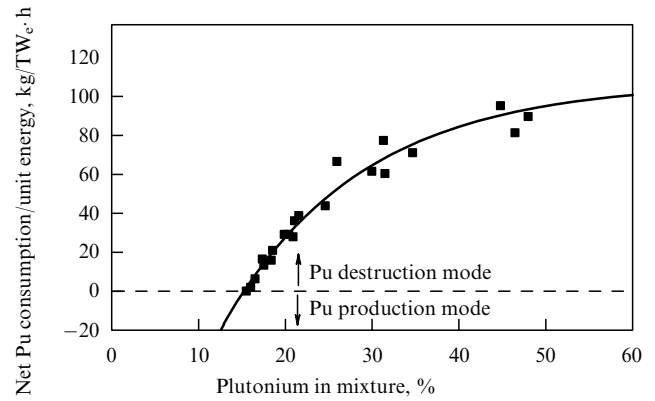


Figure 8. Net plutonium consumption per unit electric energy in a uranium–plutonium fast breeder (CAPRA [14]) as a function of plutonium concentration. Note that the unit is kg/TW_e·h electric and not thermal.

3.4 Destruction of nuclear waste: TRU

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

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 subcritical device with wide flexibility in the choice of fuel. For reasons which will become clear later, the preference is given to fuels based on thorium rather than plutonium. Pure thorium does not fission, but ²³³U bred from ²³²Th can produce energy

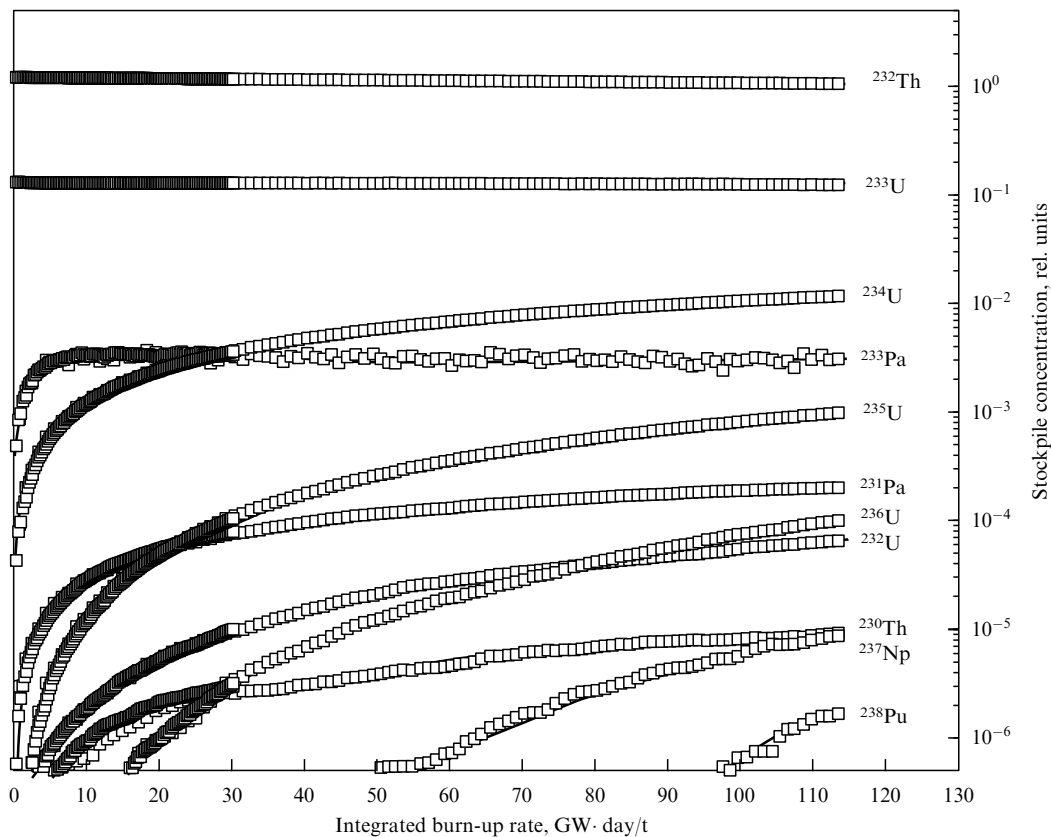


Figure 9. Evolution as a function of burn-up of the stockpile of the main elements present in EA fuel [1].

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: ^{233}U from a previous EA fuel load, ^{235}U extracted from natural uranium, military ^{239}Pu , or simply TRU, which are 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 ^{235}U in natural uranium from which PWR fuel is manufactured. Thorium lies about 5 neutron captures away from the TRU one wants to destroy, ensuring that it can more easily work in a mode where it destroys more TRU than it produces (lower equilibrium concentrations for TRU).

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 (see Fig. 8) necessitates 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^{-5} (see Fig. 9) naturally ensure a high burning rate for reasonable TRU concentrations.

A study [15] 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·year of thermal energy produced. In comparison, a PWR produces 123 kg of TRU per GW·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 [16] developed at the Argonne National Laboratory in the United States collects all TRU on a single electrode; this is acceptable since all of them fission in an EA-spectrum flux and they do not need to be separated from one another.

3.5 Why not a critical system using thorium?

Critical reactors using thorium fuel have worked in the past [17], motivated by the prospect of a high neutron yield per neutron absorbed, which ^{233}U offers over the whole neutron energy range, and only slightly surpassed by ^{239}Pu for fast neutrons. However, there is a price to pay for breeding ^{233}U . It is the production of ^{233}Pa , which has a large neutron capture cross-section and must be compensated by a higher enrichment in fissile material. Also, ^{233}U fissions produce more ^{135}Xe (direct yield of 1.4% for ^{233}U versus 0.3% for ^{235}U) and samarium precursors (^{147}Nd , ^{149}Pm) than ^{235}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.

In addition, the effective fraction of delayed neutrons (β_{eff}) for ^{233}U is less than half of that for ^{235}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 an accelerator-driven subcritical 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 of the system becoming (or not becoming) supercritical ($k_{\text{eff}} > 1$), as happened in Chernobyl in 1986. The probability of such an accident occurring may be very small, but is not zero. In a subcritical system, the effective neutron multiplication coefficient is smaller than one, its value being determined by construction of the system, not through controls. Therefore, the resulting safety aspect is a deterministic one. The system is and remains subcritical at all times and Chernobyl type accidents are simply impossible.

Furthermore, in a critical reactor, whether its fuel is based on thorium or uranium, TRU-enriched fuel leads to smaller β_{eff} values, which affects the safety margin, while as already stated, β_{eff} is unimportant for subcritical assemblies.

3.6 Destruction of nuclear waste: long-lived fission fragments (LLFF)

In a system such as the EA, where TRU are destroyed, the long-term (≥ 500 years) radiotoxicity of the waste becomes dominated by LLFF (see Fig. 10). 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. However, since the main LLFF (^{99}Tc and ^{129}I) can be soluble in water, and, therefore, have a nonzero probability over a time-scale

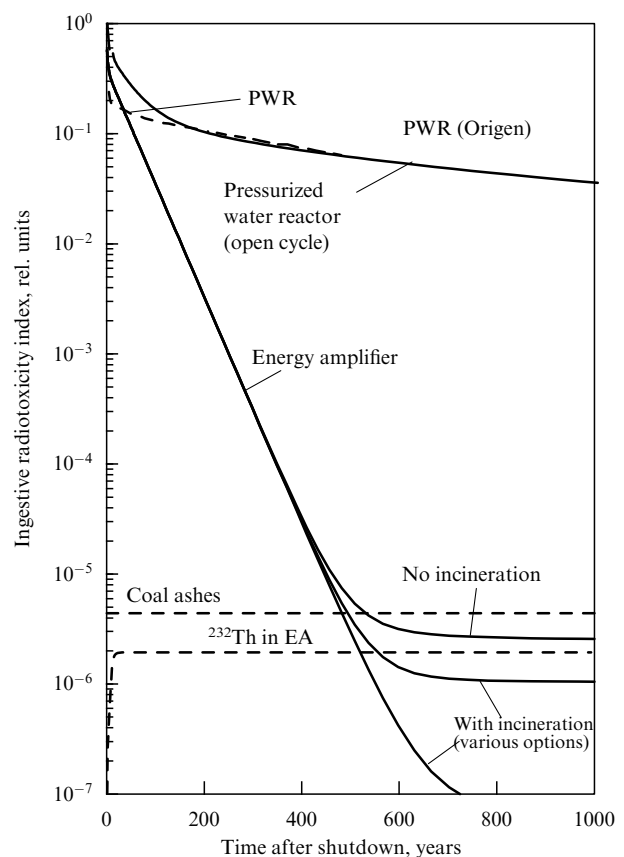


Figure 10. Evolution of the potential radiotoxicity of nuclear waste for PWR, EA, and coal-burning power station, showing that in the EA, the long-term radiotoxicity can be 4 orders of magnitude smaller than in a PWR in open cycle (adapted from Ref. [17]). The flattening of the curves above 600 years 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.

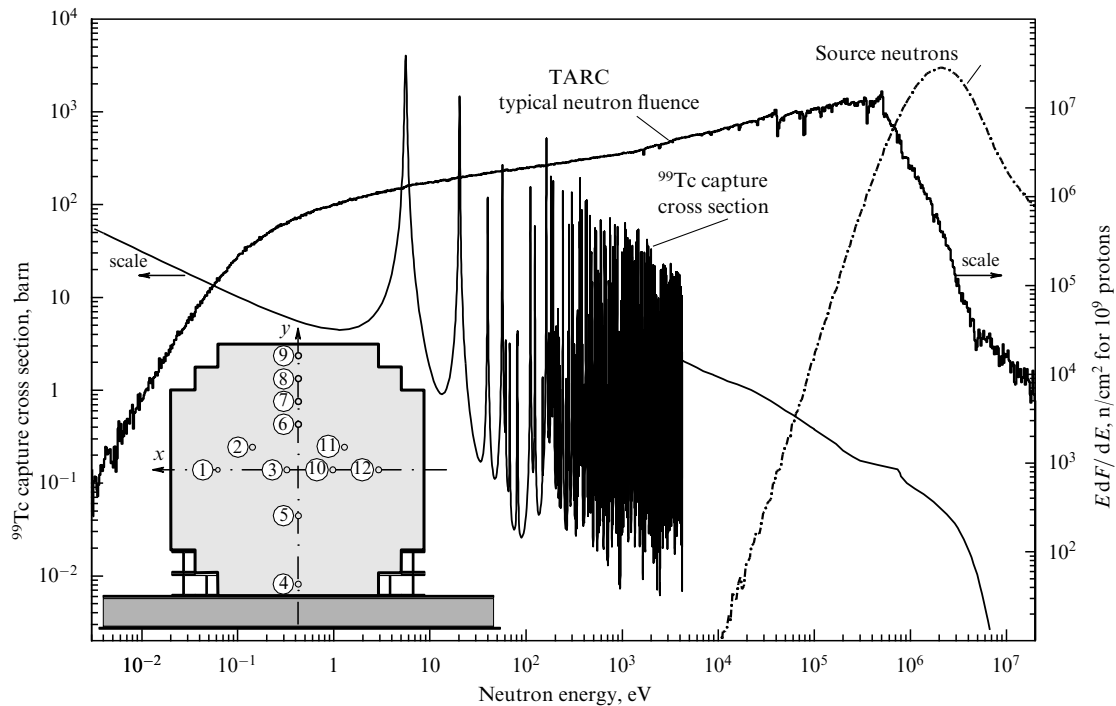


Figure 11. Illustration of the adiabatic resonance crossing principle, showing how the presence of lead transforms the spallation neutron energy distribution into a flat flux distribution for slowing down neutrons, with iso-lethargic steps smaller than the width of cross-section resonances where they will be captured with high probability. A sketch of the 334 t TARC lead volume is also shown.

of millions of years of contaminating the biological chain with hard-to-predict long-term effects, it may be wise to destroy them also.

To this end, Carlo Rubbia proposed using adiabatic resonance crossing (ARC) [19] (Fig. 11). This enhances the neutron capture probability, turning, for instance, a 2.1×10^5 year half-life ^{99}Tc into ^{100}Tc that decays quickly ($T_{1/2} \sim 15.8$ s) into stable ^{100}Ru . The TARC experiment at CERN [20] showed that using the special (small elastic collision length $\lambda \sim 3$ cm and low energy losses by elastic collisions) kinematics of neutrons in pure lead (the most transparent of all heavy elements to neutrons) 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. [17], 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 energy-producing elimination of TRU, does not pay.

3.7 Medical applications

A second important application domain of ARC is the production of radioisotopes for medical applications [18]. ARC, which is very efficient for destroying fission fragments, can also be used to induce any other type of nuclear transmutation (i.e., radioisotope production). An accelerator-driven radioisotope production system based on ARC would provide an attractive alternative to production with nuclear reactors. A relatively small system free of all the complications of running a critical nuclear reactor has many advantages, as it would:

- favor local radioisotope production thanks to the small size of the system (activator on the hospital site);
- favor the possibility of using shorter-lived isotopes, resulting in a much smaller dose to the patient [example: ^{128}I (25 min) instead of ^{131}I (8 d)];
- avoid long (costly) transportation allowing smaller doses at the production site;
- allow flexibility in the choice of neutron source according to need: from high-intensity accelerators (cyclotron) [e.g., industrial production of $^{99\text{m}}\text{Tc}$ ($T_{1/2} = 6$ h) from the decay of ^{99}Mo ($T_{1/2} = 65$ h)] to radioactive neutron sources [e.g., low-activity applications]. In TARC [19], we successfully tested the idea of employing natural molybdenum, which contains 24.13% of stable ^{98}Mo , to produce ^{99}Mo simply by neutron capture, instead of extracting ^{99}Mo from the spent fuel of a nuclear reactor.

These applications were considered sufficiently important that CERN has now obtained a patent [21] on medical radioisotope production based on ARC.

4. Conclusions

Fundamental research is a strong driving force in innovation and 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 PWR. LLFF such as ^{129}I and ^{99}Tc could be transmuted into stable elements in a parasitic mode, around the EA core, making use of the ARC method.

This experimental program has generated new applications in various fields: medical applications for which CERN now owns a patent, research with the approved CERN NTOF facility [22], and other surprising ideas such as a nuclear engine [23] for deep space exploration. All of these bring additional rewards for those who have been involved in this project.

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