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CONCEPTUAL DESIGN OF A FAST NEUTRON OPERATED HIGH POWER ENERGY AMPLIFIER

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Abstract

The basic concept and the main practical considerations of an Energy Amplifier (EA) have been exhaustively described in Ref. [1]. Here the realisation of the EA is further explored and schemes are described which offer a high gain, a large maximum power density and an extended burn-up, well in excess of 100 GW \times day/t corresponding to about five years at full power operation with no intervention on the fuel core. Most of these benefits stem from the use of fast neutrons, as already proposed in Ref. [2].

The EA operates indefinitely in a closed cycle, namely the discharge of a fuel load, with the exception of fission fragments, is re-injected in the sub-critical unit with the addition of natural Thorium to compensate for the burnt fuel. After many cycles an equilibrium is reached, in which the Actinide concentrations are the balance between burning and "incineration". The fuel is used much more efficiently, namely the power obtained from 780 kg of Thorium is roughly the same as the one from 200 tons of native Uranium and a PWR (33 GW \times day/t of burn-up). The probability of a criticality accident is suppressed since the device operates at all times far away from it. Spontaneous convective cooling by the surrounding air makes a "melt-down" leak impossible.

An EA module consists of a 1500 MW_{th} unit with its dedicated 1.0 GeV proton accelerator of 12.5 mA. A compact, highly reliable and modular Cyclotron has been designed. A plant may be made of several such modules. For instance a cluster of three such modular units will produce about 2,000 MWe of primary electrical power. A relevant feature of our design is that it is based on natural convection to remove the heat generated inside the core. The EA is a large, passive device in which a proton beam is dumped and the heat generated by nuclear cascades is extracted, without other major elements of variability. The delivered power is controlled exclusively by the current of the accelerator. The fuel needs no access during the whole burn-up and it may be kept sealed up as a non-proliferation safeguard measure. Contrary to Fusion, there are no major technological barriers.

After ≈ 700 years the radio-toxicity left is about $20,000 \times$ smaller than the one of an ordinary Pressurised Water Reactor (PWR) for the same energy. Geological storage (10^6 years) is virtually eliminated or at least strongly reduced [≤ 500 Ci/(GW_e \times y) after 1000 years]. It could be further reduced (< 35 Ci) "incinerating" some of the nuclides. Radioactivity dose to individuals truncated to 10,000 years and due to operation is about 1/330 of the one of PWR and about 1/33 of Coal burning.

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1.— Introduction.

The principle of operation of the <u>Energy</u> <u>A</u>mplifier (EA) has been described in detail in Refs. [1-3]. The present paper is aimed at the demonstration of the practical feasibility of an EA with power and power density which are comparable to the ones of the present generation of large <u>Pressurised Light Water Reactors</u> (PWR). This is only possible with fast neutrons [2].

Greenhouse induced Global Warming concerns related to a massive use of Fossil Fuels may lead to a new call for nuclear revival. But a much larger share of energy produced by conventional Nuclear methods (PWR) will sharpen concerns and enhance many of the problems which must be solved before extending its use. We believe that most of the criteria for a revival of nuclear power are very tough:

- (1) Extremely high level of *inherent safety;*
- (2)*Minimal production of long lived waste* and elimination of the need of the geologic repositories;
- (3) *High resistance to diversion,* since latent proliferation is a major concern.
- (4)*More efficient use* of a widely available natural fuel, without the need of isotopic separation.
- (5)*Lower cost of the heat* produced and *higher operating temperature* than conventional PWRs in order to permit competitive generation of substitutes to fossil fuels [4]. Substitution fuels are necessary to allow a widespread utilisation of the energy source and to permit retrofitting of existing facilities, now operating with CO₂ producing fuels.

Our design of an EA has these objectives as goals and it is intended as proof that they can be met fully. The primary fuel is natural Thorium which is completely burnt after a number of fuel cycles through the EA. Actinides present in the fuel discharge at the end of a fuel cycle are re-injected in the EA and become the "seeds" for the subsequent cycle. This ensures a very efficient use of the primary fuel element¹. This objective is identical to the one eventually met by Fast Breeders. Compared to the consumption of natural fuel material, the EA is about 250 times more efficient than the present PWRs based on an open fuel cycle.

Nuclear power has successfully developed the methods of retaining large amounts of radioactivity within the power plant and in isolation with the biosphere.

¹ The heat produced burning 70.3 kg of Thorium in the EA is equal to the one of 1 million barrels of oil

The limited amount of fuel material of the EA and the sealed, passive nature of the device further simplifies the realisation of such a concept. The fractions of radioactivity actually injected in the environment during (1) mining, (2) operation and (3) reprocessing and refuelling are considered first. Preventive measures to eliminate unwanted accidents and their possible consequences on the environment will be considered later on.

The radio-toxicity released by a Thorium driven EA is much smaller than the one of the PWR related throw-away cycle [1] [2]. In the phase of the fuel extraction and preparation, it is about $10^{-3} \div 10^{-4}$ for the same delivered energy, since a much smaller amount of Thorium is required (0.78 ton vs. 200 tons of Uranium for 3 GW_{th} \times year) in the first place and which is much less toxic to extract [5]. The toxicity released in form of waste at the back-end of the cycle for Actinides is reduced to the very tiny fraction lost during fuel re-cycling and reprocessing. Among fission fragments, excluding the short lived and stable elements, there are a few elements which are medium lived ($\tau_{1/2} \approx 30$ years, 90 Sr- 90 Y, 137 Cs, etc.) and some others (99 Tc, ¹³⁵Cs, ¹²⁹I, etc.) which are truly long lived. The policy we propose to follow is to store in man-watched, secular repositories for several centuries the medium lived in order to isolate them from the biosphere and to promote a vigorous research and development of methods of incinerating the bulk of the long lived FFs with the help of a fraction of the neutron flux of the EA or with dedicated burners [6]. Therefore, and contrary to the PWR related throw-away cycle, the need for a Geologic Repository is virtually eliminated.

UNSCEAR [7] has estimated collective radioactivity doses to the population associated to various forms of energy production. Coal burning emits radioactivity in fumes and dust, resulting in a typical, *collective radiation exposure* of 20 man Sv $(GW_e y)^{-1}$. The practice of using coal ashes for concrete production adds as much as 2.5×10^4 man Sv $(GW_e y)^{-1}$. In the case of the PWR throw-away cycle the estimated dose is 200 man Sv $(GW_e y)^{-1}$, with the main contribution coming from the mining and preparation of the fuel². Accidents which have plagued some of the present Nuclear Power stations and which are expected to be absent because of the new features of the EA, have added as much as 300 man Sv $(GW_e y)^{-1}$, bringing the toll of Nuclear Energy to about 500 man Sv $(GW_e y)^{-1}$. Translating the figures of Ref. [7] to the conditions of the EA, we arrive at much smaller collective doses, namely 2.75

²The main nuclide contributions in the nuclear fuel cycle are Radon from Mill Tailings (150 man Sv/GW/y) and reactor operation and reprocessing (50 man Sv/GW/y). The potential accumulation of collective radiation doses in the far future from the practice of disposing the long lived waste (geologic storage) is not included in the UNSCEAR estimates, since it is subject to major uncertainties.

man Sv (GW_e y)⁻¹ for the local and regional dose and $0.44 \div 1.42$ man Sv (GW_e y)⁻¹ for the global dose, depending on the type of mineral used. The total radioactivity absorbed by the population *is about one order of magnitude smaller than if the same energy is produced by burning Coal*, even if the ashes are correctly handled. In the case of the Coal option we must add the emissions of pollutants like dust, SO₂ etc. and their toll on the Greenhouse effect.

A novel element of our design is the presence of the proton beam. A recent experiment has specified the required characteristics of such an accelerator [3]. The accelerated particles are protons (there is little or no advantage in using more sophisticated projectiles) preferably of a minimum kinetic energy of the order of 1 GeV. The average accelerated current is in the range of 10 ÷ 15 mA, about one order of magnitude above the present performance³ of the PSI cyclotron [8]. This current is lower by one order of magnitude than the requirements of most of the accelerator-driven projects based on c-w LINAC [9]. In view of the present developments of high-intensity cyclotrons and the outstanding results obtained at PSI [8], we have chosen a three-stage cyclotron accelerator. In the design particular attention has been given to the need of a high reliability and simplicity of operation. The experience accumulated in the field at CERN, PSI and elsewhere indicates that this goal is perfectly achievable. The expected over-all efficiency, namely the beam power over the mains load is of the order of 40%. The penetration of the beam in the EA vessel is realised through an evacuated tube and a special Tungsten window, which is designed to sustain safely both radiation damage and the thermal stress due to the beam heating. As discussed in more detail later on, the passive safety features of the device can be easily extended to these new elements.

Since the accelerator is relatively small and simple to operate, if more current is needed, several of these units can be used in parallel, with a corresponding increase of the overall reliability of the complex. In this case, the beams are independently brought to interact in the target region of the EA.

For definiteness, in the present conceptual design of the EA we have chosen a nominal unit capacity of 1500 MW_{th}. This corresponds to about 675 MWatt of primary electrical power with "state of the art" turbines and an outlet temperature of the order of 550 \div 600 °C. The thermodynamical efficiency of \approx 45% is substantially higher than the one of a PWR and it is primarily due to the present higher temperature of operation. The general concept of the EA is shown in Figure 1.1.

³ An improvement programme is on its way to increase the average current to about 6 mA.

The nominal energetic gain⁴ of the EA is set to G = 120 corresponding to a multiplication coefficient k = 0.98. The nominal beam current for 1500 MW_{th} is then 12.5 mA × GeV⁵. In practice the proton accelerator must be able to produce eventually up to 20 mA ×GeV in order to cope with the inevitable variations of performance during the lifetime of the fuel. Such accelerator performance is essentially optimal for a chain of cascading cyclotrons. A significantly smaller current may not provide the required accelerator energetic efficiency; a higher current will require several machines in parallel. Hence, this size of the module is related, for a given gain, to the state of the art of the accelerator. The electric energy required to operate the accelerator is about 5% of the primary electric energy production. The choice of *k* is not critical. For instance an EA with k = 0.96 (G = 60) can produce the same thermal energy but with a fraction of re-circulated power about twice as large, namely 10% of the primary electricity, requiring two accelerators in parallel.

An energy generating module consists of a 1500 MW_{th} unit with its own dedicated 12.5 mA × GeV accelerator. An actual plant may be made of several such modules. For instance a cluster of three such modular units will produce about 2,000 MWatt of primary electrical power. Beams from the accelerators can be easily transported over the site and switched between units: a fourth, spare, accelerator should be added in order to ensure back-up reliability.

The modular approach has been preferred in several recent conceptual designs [10] of Sodium cooled fast reactors in the USA (ALMR, <u>A</u>merican <u>Liquid M</u>etal <u>R</u>eactor), Japan (MONJOU) and in Russia (BMN-170), for reasons of cost, speed of construction and licensing. Such modularity permits the use of the devices in relatively isolated areas. The power plant can be built in a well developed country and transported to the target area. Decommissioning of the device is also simplified. The European approach (EFR, <u>European Fast R</u>eactor) is more conservative and is based on a single, large volume pool for a nominal power in excess of 3,000 MW_{th}. Such an approach is possible also for the EA. In this case, because of the larger power, the beams from two accelerators will be simultaneously injected in the core of the EA. Both designs are robust, cost-effective and they incorporate many features which are the result of the extensive experience with smaller machines. They are designed for a number of different fuel configurations and they can easily

⁴The energetic gain G is defined as the thermal energy produced by the EA divided by the energy deposited by the proton beam.

⁵This notation is justified, since the energetic gain of the EA is almost independent of the proton kinetic energy, provided it is larger than about 1 GeV.

accommodate those appropriate to the EA. We have taken as "model" for our design many of the features of the ALMR. The ALMR was designed to provide high reliability for the key safety, including shutdown heat removal and containment. We intend to follow the same basic design, with, however, the added advantages of (1) sub-critical operation at all times (2) negative void coefficient of molten Lead (3) convection driven primary cooling system and (4) non reactive nature of Lead coolant when compared to Sodium.

The coolant medium is molten natural Lead operated in analogy with our (Sodium cooled) "models" at a maximum temperature of 600 ÷ 700 °C. In view of the high boiling temperature of Lead (1743 °C at n.p.t.) and the negative void coefficient of the EA, even higher temperatures may be considered, provided the fuel and the rest of the hardware are adequately designed. For instance direct Hydrogen generation via the sulphur-iodine method [4] requires an outlet temperature of the order of 800 °C. A higher operating temperature is also advantageous for electricity generation, since it may lead to an even better efficiency of conversion. Evidently, additional research and development work is required in order to safely adapt our present design to an increased operating temperature. In particular the cladding material of the fuel pins may require some changes, especially in view of the increased potential problems from corrosion and reduced structural strength. With these additions the present design should be capable of operating at temperatures well above the present figures.

A most relevant feature of our design is the possibility of using natural convection alone to remove all the heat produced inside the core. Convection cooling has been widely used in "swimming pool" reactors at small power levels. We shall show that an extension of this very safe method to the very large power of the EA is possible because of the unique properties of Lead, namely high density, large dilatation coefficient and large heat capacity. Convection is spontaneously and inevitably driven by (any) temperature difference. Elimination of all pumps in the primary loop is an important simplification and a contribution towards safety, since unlike pumps, convection cannot fail. In the convective mode, a very large mass of liquid Lead (\approx 10,000 tons), with an associated exceedingly large heat capacity⁶ moves very slowly (\leq 2.0 m/s inside the core, about 1/3 of such speed elsewhere) transferring the heat from the top of the core to the heat exchangers located some 20

⁶ The heat capacity of liquid Lead at constant pressure is about 0.14 Joule/gram/°C. For an effective mass of $\approx 10^4$ tons=10¹⁰ grams and a power of 1.5 GWatt (full EA power), the temperature rise is of the order of 1.0 °C/s. The mass flowing through the core for $\Delta T \approx 200$ °C is 53.6 tons/sec, corresponding to some 1.5 minutes of full power to heat up the half of the coolant in the "cold" loop, in case the heat exchangers were to fail completely.

metres above and returning at a lower temperature ($\Delta T \approx -200$ °C) from the heat exchangers to the bottom of the core.

The geometry of the EA main vessel is therefore relatively slim (6.0 m diameter) and very tall (30 m). The vessels, head enclosure and permanent internal structures are fabricated in a factory and shipped as an assembled unit to the site⁷. The relatively slender geometry enhances the uniformity of the flow of the liquid Lead and of the natural circulation for heat removal. The structure of the vessel must withstand the large weight of the liquid Lead. There are four 375 MW_{th} heat exchangers to transfer the heat from the primary Lead to the intermediate heat transport system. They are located above the core in an annular region between the support cylinder and the walls of the vessel.

The vessel is housed below floor level in an extraordinarily robust cylindrical silo geometry lined with thick concrete which acts also as ultimate container for the liquid Lead in case of the highly hypothetical rupture of the main vessel. In the space between the main vessel and the concrete wall the <u>Reactor Vessel Air Cooling System</u> (RVACS) is inserted. This system [11], largely inspired from the ALMR design, is completely passive and based on convection and radiation heat transfer. The whole vessel is supported at the top by anti-seismic absorbers. Even in the case of an intense earthquake the large mass of the EA will remain essentially still and the movement taken up by the absorbers.

The fuel is made of mixed oxides, for which considerable experience exists. More advanced designs have suggested the use of metallic fuels or of carbides [12]. These fuels are obviously possible also for an EA. We remark that the use of Zirconium alloys is not recommended since irradiation leads to transmutations into the isotope 93 Zr, which has a long half-life and which is impossible to incinerate without separating it isotopically from the bulk of the Zirconium metal. The choice of the chemical composition of the fuel is strongly related to the one of the fuel reprocessing method. A relative novelty of our machine when compared to ordinary PWRs is the large concentration of ThO₂ in the fuel and the corresponding production of a small but relevant amount of Protactinium. A liquid separation method called THOREX has been developed and tested on small irradiated ThO₂ fuel samples [13]. The extrapolation from the widely used PUREX process to THOREX is rather straightforward and this is why we have chosen it, at least at this stage. Methods based on pyro-electric techniques [14], which imply preference to metallic

⁷ The shipping weight is about 1500 tons. Removable internal equipment is shipped separately and installed through the top head.

fuels, are most interesting, but they require substantial research and development work. Since the destination of the Actinides is now well defined i.e. to be finally burnt in the EA, the leakage of Actinides in the Fission Fragment stream must be more carefully controlled, since they are the only Actinides in the "Waste". We have assumed that a "leaked" fraction of 10⁴ is possible for Uranium. The recycled fuel has a significant radio-activity. We have checked that the dose at contact is similar to the one of MOX fuels made of Uranium and Plutonium, already used in the Nuclear Industry.

The average power density in the fuel has been conservatively set to be $\rho = 55$ *Watt/gr-oxide*, namely about 1/2 the customary level of LMFBR⁸ (ALMR, MONJOU, and EFR). The nominal power of 1500 MW_{th} requires then 27.3 tons of mixed fuel oxide. The fuel dwelling time is set to be 5 years equivalent at full power. The average fuel burn-up is then 100 GWatt day/ton-oxide. Since the fissile fuel is internally regenerated inside the bulk of the Thorium fuel, the properties of the fuel are far more constant than say in the case of a PWR. As shown later on, one can compensate to a first order the captures due to fission fragments, operating initially with a breeding ratio below equilibrium. All along the burn-up, the growth of the fissile fuel concentration counterbalances the poisoning due to fission fragments. *Therefore neither re-fuelling nor fuel shuffling appear necessary for the specified duration of the burn-up*.

No intervention is therefore foreseen on the fuel during the five years of operation, at the end of which it is fully replaced and reprocessed. Likewise in the "all-convective" approach there are no moving parts which require maintenance or surveillance. In short the *EA is a large, passive device in which a proton beam is dumped and the generated heat is extracted, without other major elements of variability.*

Safety and nuclear proliferation are universal concerns. In the case of conventional Nuclear Power, accidents have considerably increased the radioactivity exposure of individuals and the population [7]. The total nuclear power generated, 2000 GW \times year, is estimated to have committed an effective dose of 400,000 man Sv from normal operation. Accidents at Windscale, TMI and Chernobyl have added 2000, 40 and 600,000 man Sv respectively. These types of accidents are no longer possible with the EA concept: Chernobyl is a criticality accident, impossible in a sub-critical device and TMI, a melt-down accident, is made impossible by the "intrinsic" safety of the EA.

⁸ This choice is motivated by the relative novelty of the "all-convective" approach and the relative scarce experience with ThO_2 , when compared with UO_2 .

A thermal run-off is the precursory sign of a number of potentially serious accidents. The present conceptual design is based on a swimming pool geometry where the heat generated by the nuclear cascade is extracted from the core by convection cooling, completely passive and occurring inevitably because of temperature differences. Thermal run-off is prevented, since a significant temperature rise due for instance to an insufficiency of the secondary cooling loop and of the ordinary controls will inevitably produce a corresponding dilatation of the liquid Lead. Because of the slim geometry of the vessel, the level of the swimming pool will rise by a significant amount ($\approx 27 \text{ cm}/100 \text{ °C}$), filling (through a siphon) additional volumes with molten Lead, namely :

- (1)The <u>Emergency Beam Dump Volume (EBDV)</u>, a liquid Lead "beam stopper" sufficiently massive as to completely absorb the beam some 20 metres away from the core and hence bring the EA safely to a stop. In the unlikely event that the beam window would accidentally break, molten Lead will also rise, so as to fill completely the pipe and the EBDV, thus removing the incoming proton beam from the core.
- (2)A narrow gap normally containing thermally insulating Helium gas, located between the coolant and the outer wall of the vessel, which in this way becomes thermally connected to the coolant main convection loop. The outer wall of the EA will heat up and bleed the decay heat passively through natural convection and radiation to the environment (RVACS) [11]. This heat removal relies exclusively on natural convection heat transfer and natural draught on the air side.
- (3)A scram device based on B_4C absorbers which are pushed into the core by the liquid Lead descending narrow tubes. These absorbers anchor the device firmly away from criticality.

These passive safety features are provided as a backup in case of failure of the active systems, namely of the main feed-back loop which adjusts the current in order to maintain constant the temperature at the exit of the primary cooling loop. Multiply backed-up but simple systems based on current transformers and physical limitations in the accelerator (available RF power in the cavities, space charge forces, etc.) sharply limit the maximum current increase that the accelerator can deliver. Were these methods all to fail, the corresponding increase of temperature will dilate significantly the Lead, activating the ultimate shut-off of the proton beam from the accelerator, the emergency cooling and the scram devices, before any limit is exceeded in the EA.

Normally the EA is well away from criticality at all times, there are no control bars (except the scram devices) and the power produced is directly controlled by the injected beam current. However in some unforeseen circumstances the EA may become critical. In itself, this is not an unacceptable though exceptional operation mode, provided the amount of power produced does not exceed the ratings of the EA. Indeed even a quite large reactivity insertion is strongly moderated by the large negative temperature coefficient (Doppler) of the fuel. Since the operating temperature of the fuel is relatively low, even a rapid increase of the instantaneous power will increase the temperature of the fuel within limits, large enough, however, to introduce a substantial reduction of k as to exit from criticality. The safety of multiplying systems depends to a large extent on fast transients. A kinetic model dealing with fast transients due to accidental reactivity insertions and unexpected changes of the intensity of the external proton beam shows that the EA responds much more benignly to a sudden reactivity insertion than a critical Reactor. Indeed, no power excursions leading to damaging power levels are observed for positive reactivity additions which are of the order of the sub-criticality gap. Even if the spallation source is still active (the accelerator is not shut-off), the power changes induced are passively controlled by means of the increase of the natural convection alone (massive coolant response) thus excluding any meltdown of the sub-critical core.

Any very intense neutron source ($\geq 10^{13}$ n/s) could in principle be used to produce bomb grade Plutonium by extensive irradiation of some easily available depleted Uranium. This is true both for fission and fusion energy generating devices. We propose to prevent this possibility by "sealing off" the main vessel of the EA to all except a specialised team, for instance authorised by IAEA. This is realistic for a number of reasons. The energetic gain of the EA is almost constant over the lifetime of the fuel, though it changes significantly after a power level variation. Convection cooling is completely passive and occurring inevitably because of temperature differences. There are no active elements (pumps, valves etc.) which may fail or need direct access to the interior of the main vessel. In addition the fuel requires no significant change in conditions over its long lifetime of five years, since the fissile material is continuously generated from the bulk of Thorium. The only two maintenance interventions to be performed are the periodic replacement of the beam window about once a year and the possible replacement of some failing fuel elements, performed remotely with the pantograph. Both activities can be carried out without extracting the fuel from the vessel. We can therefore envisage conditions in which the EA is a sort of "off limits black-box" accessed very rarely and only by a specialised team, for instance authorised by IAEA. The ordinary user (and owner) of

the EA will have no access to the high neutron flux region and to the irradiated fuel, a necessary step towards any diversion which may lead to proliferation or misuse of the device.

Proliferating uses of the fuel are further prevented by the fact that the fissile Uranium mixture in the core is heavily contaminated by strong γ -emitter ²⁰⁸Tl which is part of the decay chain of ²³²U and by the fact that the EA produces a negligible amount of Plutonium. As shown later on, a rudimentary bomb built starting with EA fuel, in absence of isotopic separation, will be most impractical and essentially impossible to use or to hide.

The EA can operate with a variety of different fuels. Several options will be discussed in detail in the subsequent sections. A specialised filling can transform Plutonium waste into useful ²³³U, for instance, in order to accumulate the stockpile required at the start-up of the EA. More generally one could envisage a combined strategy with ordinary PWRs. Presently operating PWRs represent an investment in excess of 1.0 Tera dollars. It is important to make every possible effort in order to minimise their impact on the environment and to increase their public acceptability. A specially designed EA could be used to (1) transform Plutonium waste into useful ²³³U and (2) reduce the stockpile of "dirty" Plutonium waste. The EA will be initially loaded with a mixture of Actinide waste and native Thorium, in the approximate ratio 0.16 to 0.84 by weight. Other Actinides, like Americium, Neptunium and so on can also be added. The mixture is sub-critical and the EA can be operated with *k* = 0.96-0.98.

During operation, the unwanted actinides are burnt, while ²³³U is progressively produced. The freshly bred ²³³U compensates the drop of criticality due to the diminishing and deteriorating Actinide mixture and the one due to the build-up of Fission Fragments. A balanced operation over a very long burn-up of up to 200 GW day/t is thus possible without loss of criticality, corresponding to 5-10 years of operation without external intervention. The fuel of the EA is then reprocessed, the ²³³U is extracted for further use. FFs are disposed with the standard procedure of the EA. The remainders of Plutonium⁹ and the like, could either be sent to the Geological Repository to which they were destined or further burnt in the EA, topped with fresh Thorium. This combination of a PWR and an EA has several advantages:

⁹ The discharge after $\approx 150 \text{ GWatt} \times \text{day/t}$ contains about 50% of the initial Pu, but is highly depleted of 239 Pu (1/5) and 241 Pu(1/4), while other Pu isotopes are essentially unchanged. Am and Cm isotopes stockpiles are essentially unchanged. Note that the Plutonium is "denatured" of the highly fissile isotopes, making it worthless for military diversions.

- 1) It eliminates permanently some of the Actinide waste of the PWR reducing the amount to be stored in a Geological Repository.
- 2) It produces additional power through the EA, thus increasing by about 50% the energetic yield of the installation.
- 3) The amount of fissile Uranium, which is by weight about 80% of the incinerated Plutonium is a valuable asset. It can be used either to start a new, Thorium operated, EA or it can be mixed with depleted Uranium to produce more fuel for PWRs. As is well known, 233 U is an almost perfect substitute for 235 U in a ratio very close to 1. The yearly Plutonium and higher Actinides discharge of a typical $\approx 1 \text{ GW}_{e}$ PWR operated 80% of the time is of the order of 300 kg, thus producing via the EA 240 kg of 233 U, which in turn can be used to manufacture ≈ 8 tons of fresh fuel from depleted U with 0.3% 235 U and 3.0% 233 U. This is $\approx 1/3$ of the supply of enriched Uranium fuel for the operation of the PWR.

We have also considered as an alternative a fast neutron driven EA operated on Plutonium only, namely without Thorium. Similar schemes, though mostly operated with thermal neutrons are under consideration at Los Alamos [15], JAERI [16] and elsewhere [17]. Such potential devices require frequent refills and manipulations of the fuel, since the reactivity of the Plutonium is quickly deteriorated by the burning and choked by the emergence of a large relative concentration of FF's. At the limit one is lead to the "chemistry on line" proposed by the Los Alamos Group [15]. Adding a large amount of fertile Thorium greatly alleviates such problems and the device can burn Plutonium and the like for very many years without intervention or manipulation of the fuel, since the bred ²³³U is an effective substitute to Plutonium to maintain a viable and constant criticality. In addition FFs are diffused in a much larger fuel mass. Finally the ²³³U recovered at the end of the cycle constitutes a valuable product.

In principle our method of a Th-Pu mixture could be extended to the operation of a Fast Breeder used as incinerator [18], however, probably at a much higher cost and complexity due to the higher degree of safety involved.

We have indicated Thorium as main fuel for the EA since the radio-toxicity accumulated is much smaller than Uranium and it offers an easier operation of the EA in a closed cycle. But there are also reasons of availability. Thorium is relatively abundant on earth crust, about 12 g/ton, three times the value of Uranium [19]. It ranks 35-th by abundance, just after Lead [20]. It is well spread over the surface of the planet. In spite of its negligible demand ($\approx 400 \text{ t/y}$) the known reserves in the

WOCA¹⁰ countries are estimated [21] to about 4×10^{6} tons (Table 1.1). Adding a guessed estimate from the USSR, China and so on, we reach the estimate of perhaps 6×10^{6} tons, which can produce 15,000 TW × year of energy, if burnt in EAs, namely about a factor 100 larger than the known reserves of Oil or Gas and a factor 10 larger than Coal. This corresponds to 12.5 centuries at the present world's total power consumption (10 TW).

There are reasons to assume that this figure is largely underestimated. Firstly the demand is now very low and there has been very little incentive to date to search for Thorium "per se". Additional resources of any mineral have always been found if and when demand spurs a more active perspection. The presently exploited Thorium ores are richer, by a factor $10 \div 100$, than the ones which are exploitable at a price acceptable by market conditions applicable to the case of Uranium.

In view of the small contribution of the primary Thorium to the energy cost, one may try to estimate how the recoverable resources would grow if exploitation is extended to ores which have a content for instance an order of magnitude smaller, i.e. similar to the best Uranium ores. Such analysis has been performed for Uranium [22], assuming that the distribution in the crust follows a "log-normal" (Figure 1.2) distribution. Other metals for which a better mining history is known, show a similar trend, though the slope parameter may be different in each case (Figure 1.3). In the case of Thorium, in absence of better information, we may assume the same slope as in the case of Uranium. Then, a tenfold decrease in the concentration of the economically "recoverable" ores¹¹ would boost reserves of Thorium by a factor of 300, still a small fraction (3×10^{-5}) of what lies in the Earth crust. Reserves of Thorium energy would then be stretched to 4.5 million TW × year, *corresponding to* ≈ 2200 centuries at twice the present world consumption level which can be considered truly infinite on the time scale of human civilisation.¹².

Several other projects have sought the realisation of a "clean" Nuclear Energy. The project CAPRA [23] focuses on the incineration of Plutonium in a Fast Breeder. On a longer time scale, Fusion holds the promise of a "cleaner" energy. Amongst the various projects, Inertial Fusion offers the largest flexibility in design of the combustion chamber and hence the best potentials of reduction of the activation

¹⁰This stands for World Outside Centrally Planned Activities.

¹¹We remark that even this 10-fold decrease would make these minerals somewhat more concentrated than the 2000 ppm "high content ores" used today for Uranium.

¹²In order to estimate the magnitude of the error in such a "prediction", we note that the somehow extreme cases of Tungsten and of Copper have boost factors of 500,000 and 40 respectively. But even the lower limit of Copper predicts \geq 300 centuries at twice the today's world consumption.

effects due to neutrons [24]. But neither inertial nor magnetic fusion have so far achieved ignition¹³. We have compared the activity of the remnants (Ci) of the EA with the one of the CAPRA project and of two of the Inertial Fusion concepts, namely LIBRA [25] and KOYO [26] in which the greatest care has been exercised to reduce activation. In order to make the comparison meaningful we have to take into account that the published values of activation for fusion are given in Ci after shut-down and 40 years of operation. Therefore the activities quoted for the fission case (CAPRA, EA) have been normalised to the same scenario, namely counting the total activity of remnants (sum of all fuel cycles, in the case of EA excluding recycled fuel) after 40 years of continued, uninterrupted operation. Activities have been normalised 1 GW of electric power produced (Figure 1.4).

After the cool-down period in the secular repository (≈ 1000 years) the activity of the remnants (40 years of operation) stabilises at levels which are : 1.7×10^7 Ci for CAPRA, 2.35×10^4 Ci for LIBRA, 900 Ci for KOYO and 1.3×10^4 Ci for the EA without incineration. With incineration we reach the level of 950 Ci, out of which about one half is due to ¹⁴C. The activation for unit delivered power of the EA without incineration is comparable to the one of LIBRA concept whilst with incineration we reach a level which is close to the one of KOYO concept based on second generation design of the combustion chamber. The expected doses after 1000 years of cool-down from Magnetically Confined Fusion are typically three order of magnitude larger than the quoted values for Inertial confinement due to substantial differences in the neutron spectra. This improvement is mainly due to the moderation of neutrons in the blanket consisting of LiPb liquid circulating through SiC tubes, before they hit the first wall [24]. *Therefore we conclude that the EA concept can reach a level of "cleanliness" which is well in the range of the best Fusion conceptual designs.*

From the point of view of cleanliness, as well as for the other major goals — namely non-criticality, non-proliferation and inexhaustible fuel resources — the EA matches fully the expectations of Fusion. But like CAPRA — which however is about 1000 times less effective in eliminating radioactive remnants — the EA has no major technological barriers, while in the case of Fusion, major problems have to be solved.

¹³ The project ITER is aimed at demonstrating Ignition in magnetically confined fusion, presumably circa 2005. The new large megajoule range optical LASERs in development at Livermore and in France have the potential for ignition with inertial fusion.



	Reasonably	Additional	Total
	Assured	Resources	
Europe			
Finland		60	60
Greenland	54	32	86
Norway	132	132	264
Turkev	380	500	880
Europe Total	566	724	1290
America			
Argentina	1		1
Brazil	606	700	1306
Canada	45	128	173
Uruguay	1	2	3
USA	137	295	432
America total	790	1125	1915
Africa			
Egypt	15	280	295
Kenya	no estimates	no estimates	8
Liberia	1		1
Madagascar	2	20	22
Malawi		9	9
Nigeria	no estimates	no estimates	29
South Africa	18	no estimates	115
Africa total	36	309	479
Asia			
India	319		319
Iran		30	30
Korea	6	no estimates	22
Malaysia	18		18
Sri Lanka	no estimates	no estimates	4
Thailand	no estimates	no estimates	10
Asia total	343	30	403
Australia	19		19
Total WOCA	1754	2188	4106

Table 1.1 - Thorium resources (in units of 1000 tons) in WOCA (World Outside Centrally Planned Activities) [21]

This compilation does not take into account USSR, China and Eastern Europe. Out of 23 listed countries, six (Brazil, USA, India, Egypt, Turkey and Norway) accumulate 80% of resources. Brazil has the largest share followed by Turkey and the United States.



- Figure 5.11a Pressure drop inserted as a function of the radial position. As a reference the total pressure drop due to the Lead column is also displayed.
- Figure 5.11b Lead velocity distribution as a function of the radial position.
- Figure 5.12 Velocity map of Lead in the column above the Core.
- Figure 5.13 LMFBR power excursion benchmark (as defined in a comparative NEACRP exercise) assuming a rod ejection accident.
- Figure 5.14 Comparison of power excursions in a critical reactor (lead cooled) with the Fast Energy Amplifier for an accidental reactivity insertion of 170 \$/s for 15 ms.
- Figure 5.15 Comparison of power excursions in a critical reactor (lead cooled) with the Fast Energy Amplifier for an accidental reactivity insertion of 255 \$/s for 15 ms.
- Figure 5.16 Power excursion and reactivity behaviour during a beam run-off in the Fast Energy Amplifier.
- Figure 5.17 Effect of ²³³Pa on the decay heat of the Fast Energy Amplifier.
- Figure 5.18 Time Evolution of the γ -activity of the fuel after discharge of the EA. The number of γ -rays is normalized according to their energy in MeV. The curve for the PWR has been calculated for the same energy delivered and a burn-up of 33 GW \times day/t.



Figure 5.1







Figure 5.3











Figure 5.7







Figure 5.10





Figure 5.11b



Figure 5.12



Figure 5.13



Figure 5.14



Figure 5.15



Figure 5.16



Figure 5.17



6. — Closing the Fuel Cycle.

6.1 - General Considerations. There are significant, conceptual differences between what one means by "reprocessing" in the case of a PWR and an EA. In the case of a PWR, the primary purpose of reprocessing — if one excludes recovery of Plutonium for military applications — is the one of preparing for a more orderly, definitive repository of the radio-toxic products, separating for instance Actinides from FFs. Many conceptual designs have been proposed for the purpose of further healing the strong radio-toxicity of such individual products with nuclear transformations with the help of neutrons from Accelerators and Reactors. We shall mention as our reference case the project CAPRA [23] in which one intends to reduce the radio-toxicity of the Plutonium from spent fuels by about a factor 30 with the help of Fast-Breeders similar to SuperPhénix. In addition to producing a large amount of electric energy, one such device could process Plutonium and eventually Americium produced by about five ordinary PWRs.

In the case of the EA, at "replacement" time the fuel itself (Actinides) is still perfectly sound and it could continue to burn much further if it were not for the neutron absorption due to the accumulated FFs. Hence after a "reprocessing", which is in fact basically a "FF separation and disposal", the fuel can and should be used again. This is a fundamental difference with a PWR, where spent fuel is hardly more than waste material and for which reprocessing is arguable. In the case of an EA, fuel reprocessing could be better described as <u>fuel regeneration</u>. The purpose of such a procedure is

- (1) to remove the poisoning FFs;
- (2) to add the fraction of the Thorium fuel which has been burnt;
- (3) to re-establish mechanical solidity to the fuel and the cladding which has been affected by the strong neutron flux.

In nuclear power generation, radioactive materials must be isolated at all times from the environment with an appropriate, multiple containment. The residual radio-toxicity is defined as the toxicity of products extracted from such a closed environment. Since the bulk of the Actinides are recycled inside the core for further use, the relevant toxicity is basically the one which is spilled out during the fuel regeneration process and the one of the elements which are deliberately removed, like for instance the one of the FFs which are not incinerated and of the sleeves which contain the fuel which are not reused. This is in contrast with an ordinary PWR — at

least if no incineration is performed — in which the totality of the radio-toxicity of the spent fuel constitutes "Waste" and it must be isolated from the environment by a Geologic Repository over millions of years.

6.2 - Strategy for the Spent Fuel. The main requirement of the reprocessing of the fuel from the EA is the one of generating a new fuel free of FFs. Therefore reprocessing is inevitable in our conception of the EA. In practice one must separate the Fuel into two different stock piles, the first destined to the next fuel load and the remainder which is usually called the high activity stream (HLW). The bulk of the Actinides are to be recycled into new fuel and they belong to the former stockpile. There is no need to worry about their long lasting consequences, since they will be burnt in the successive, cycles. The latter stockpile will contain all fission fragments and activity in the cladding plus the tiny fraction f of Actinides which is not separated by the reprocessing. They represent a considerable radio-toxicity, which will be handled either with natural decay or with active incineration of some specific radio-nuclides. Figure. 6.1 gives the ingestive radio-toxicity [31] of such a high activity stream assuming $f = 1.0 \times 10^{-4}$ (the choice of such a value will be clearer later on). The total radio-toxicity of a PWR initially loaded with 3.3% enriched Uranium and without reprocessing is also shown for comparison. Data are given for the fuel discharge after the first fill and for asymptotic fuel composition. The two distributions are very similar, since the fuel remaining radio-toxicity at long times is dominated by the ²³³U contamination which is the same for all fillings. After a large drop over the first ≈ 500 years due to the decay of medium lifetime FFs (90 Sr- 90 Y, ¹³⁷Cs), the ingestive radio-toxicity stabilises to a roughly constant level, dominated by the truly long lived FFs (129I, 99Tc, 126Sn 135Cs, 93Zr and 79Se) and to a lower extent by the residual fraction f of Actinides. After such a cooling-off time the residual radio-toxicity is comparable to the one of the 232 Th in the EA and about 5 \times 10^{-5} times smaller than the one of a throw-away PWR of equivalent yield. The α activity is very modest since it is dominated by the leaked fraction *f* of Actinides.

Inspection of Figure 6.1 suggests that the HLW should be stored for about 500 \div 700 years in what we call the "Secular Repository". Beyond such period, the residual radio-toxicity is considerably reduced as shown in Figure 6.2. The specific FFs contributing to radio-toxicity after 1000 years are listed in Table 6.1. It is possible to consider at this point the surviving radiation as Class A (10 CFR 61) for surface storage material even if the waste material will remain buried and provided it is diluted in $\ge 1000 \text{ m}^3/(\text{GW}_e \times \text{year})$.

It is possible to further reduce the activity of the residual waste by extracting some or all the sensitive elements of Table 6.1 and "incinerating" them with neutrons in the EA. A more detailed paper on incineration is in preparation [78] and an experiment is in preparation at CERN [6], since most of the relevant cross sections are poorly known. Here we shall limit our considerations to the ones on general strategy. Three possible further steps are possible:

- 1) Technetium and Iodine are chemically extracted and incinerated. The first is a pure ⁹⁹Tc isotope and the second besides ¹²⁹I contains about 33% of stable isotopes which are kept in the incineration stream. The total mass to be incinerated is about 19 kg/(GW_e × year), which is modest. The ingestive radio-toxicity of the remainder after 1000 years is reduced from 63.4 kSv to 16.2 kSv and the Class A dilution volume from 1194 m³/(GW_e × year) to 68 m³/(GW_e × year).
- 2) Procedure as point 1) but also Caesium is chemically extracted. The amount of Caesium is much larger, $\approx 100 \text{ kg/(GW}_e \times \text{year})$. In addition isotopic separation is necessary in order to separate the 34 kg/(GW_e × year) of ¹³⁵Cs from the very radio-toxic ($3.92 \times 10^6 \text{ Sv}$) but shorter lived ¹³⁷Cs. This may be difficult, although a feasibility study has been carried out [79]. After incineration of ¹³⁵Cs, the ingestive radio-toxicity after 1000 years of the remainder is reduced to 6.3 kSv and the Class A dilution volume to 29 m³/(GW_e × year).
- 3) Procedure as point 2) but also Zirconium and Tin are chemically extracted. Both elements require isotopic separation. One of the other isotopes of Tin is radioactive and slightly toxic. In this way the only known long lived isotope left in the discharge is ⁷⁹Se (0.3 kg) which represents 0.745 kSv and the ridiculously small Class A dilution volume of 0.6 m³/(GW_e × year).

These procedures (Figure 6.3) will ensure that the radio-toxicity of the FFs in the "Secular Repository" is exhausted in less than 1000 years, which is a sufficiently short time to be absolutely confident that current technologies of vitrification and of containment can make the storage totally safe.

In addition to the FFs, in the High Level Stream there will be leaks of Actinides due to the imperfections of the reprocessing. These radio-nuclides are more worrisome since some of them are important α -emitters. The radio-toxicity and the α -activity in Ci for leaked fractions $f = 10^{-4}$ and $f = 2 \times 10^{-6}$ are displayed in Figure 6.4 and in Figure 6.5 respectively. The radio-toxicity has two maxima or "bumps", the first roughly for time span of the secular repository and a second for very long times, namely $10^5 \div 10^6$ years. The second maximum is due to 233 U and its descendants.

The first bump in the toxicity in the early fillings is due to 232 U and it grows substantially in the later fillings and in the asymptotic fuel composition because of the increased presence of 238 Pu and its descendants. The α -activity is instead always determined by the 232 U and its descendants at short times and by 233 U and its descendants at long times. The total α -activity of Actinides is about 10⁵ Ci, for a fuel mass of the order of 22 tons, which corresponds to an average activity of about 5 mCi/g. Note that the activity of Thorium which is the largest mass is very small and that if Uranium's are separated out they will have a specific activity which is about ten times larger than the bulk of the spent fuel.

6.3 - *Fuel reprocessing methods.* In our case production of the lighter Neptunium and Plutonium isotopes is very low and higher actinides are nearly absent. However the (n,2n) reactions, more probable at high energies, increase the amount of highly toxic ²³¹Pa and ²³²U.

The EA requires the recovery of the Uranium (²³³U). However, it offers the opportunity of destroying the other Actinides by concentrating them, after each discharge, in a few dedicated fuel bars (targets) inserted somewhere in the bundles of ordinary fuel, where an incineration lifetime of years is at hand. The amount of leaking Actinides in the High activity Waste stream destined to the Secular repository must be a small fraction $f \le 10^{-4}$ of the produced amount. If incineration of the long lived FFs is performed to alleviate the radio-toxicity of the stored products after 500 years, an even higher performance in separating power is advisable, $f \le 2.0 \times 10^{-6}$. The efforts in order to attain such a figure is justified by the considerable benefit attained by the practical elimination of the "Geologic times Repository". We remark that such an incentive has been so far absent.

Two methods have been considered and appear suitable to our application: (1) aqueous methods, presently in use and (2) the newly developed pyro-electric method. We shall review both of them in succession.

Aqueous reprocessing methods have proven to be efficient, particularly for the separation of U and Th (99.5% and higher). The best known example is the THOREX process, based on solvent extraction through the use of tributyl phosphate (TBP), which extracts and separates the Thorium and Uranium. Other Actinides can also be extracted although their concentrations are so low that the extraction efficiency will be lower.

Figure 6.6 describes schematically the overall fuel cycle. The fuel rods should be stored for cooling at least for one year, to allow the ²³³Pa to decay to ²³³U. Fuel rods are then sheared and chopped. The gaseous fission products will be accumulated, with in particular attention for the ⁸⁵Kr and ¹⁴CO₂ which are destined to the secular repository. Dissolution should be made with a mixture of nitric acid (HNO₃) and hydrofluoric acid (HF) since ThO₂ is a very refractory ceramic material. The HF concentration should not be higher than 0.1 M and the addition of aluminium nitrate Al(NO₃)₃ as reagent could be needed in order to avoid corrosion of the stainless steel dissolver. Before carrying out the solvent extraction process from the obtained liquids they should be cleared of the remaining solids. The main components of the liquid will then be Thorium, Uranium, Fission Products, Protactinium and other trans-uranic Actinides.

The classic process to carry out the separation of Th and U from fission fragments is the acid THOREX. It uses TBP 30% v/v diluted with an organic solvent like dodecane. The partition of U from Th is done by washing the organic phase with diluted nitric acid. The U stream will also contain the very small amount of Pu and some contamination of Th and FF. The contamination of the Thorium stream will be mainly FF. The high active liquid waste stream will mainly contain FF, trans-uranic Actinides (²³¹Pa, ²³⁷Np) and some residual contamination of Th and U. Further cycles for purification of Uranium and Thorium should be applied using TBP as extractant.

There is little information on the recovery of Pa and it will possibly require some additional studies. Tests carried out at Oak Ridge have shown [80] that Pa could be absorbed from solutions with high content of nitric acid by using various absorbents like unfired Vycor glass, silica gel or Zirconium phosphate. Its extraction should be done from the high level waste stream. Relative to the other Actinides its extraction will be less efficient since their concentration in the Highly Radioactive liquid Waste stream, although it can and should be increased, will nevertheless be very low.

The performance quoted in Figure 6.6 is above the current values according to standard experience on the THOREX process [13], [81], but appropriate tuning of the chemical parameters should allow higher efficiencies. The minimisation and ultimate disposal of High-Level radioactive Waste (HLW) generated from the reprocessing of spent fuel (THOREX) is an important part of the global nuclear fuel recycling strategy proposed in the framework of the Energy Amplifier Concept, as an alternative to classical disposal methods. The goal is twofold, (i) to recover from the

insoluble residue useful metals such us Ru, Rh and Pd; (ii) and to separate Actinides⁴⁵ and some of the LLFPs (Long-Lived Fission Products) for their further use (incineration) or disposal. We believe this can best be achieved with the method developed in the context of the IFR (Integral Fast Reactor) programme [82], where it is proposed to separate actinides⁴⁶ and FPs from HLW by dry process with pyrochemical (or pyro-metallurgical) methods (Figure 6.7). However, the only process that has reached an industrial scale is, at least for the moment, the PUREX process (aqueous method) which has already been described in the previous paragraphs. All the other methods are still in the technical or laboratory development phase.

Figure 6.8 shows the flow diagram of the dry process for partitioning of Actinides [83]. This process consists of (i) denitration to obtain oxides, (ii) chlorination to oxide to chlorides, (iii) reductive extraction to reduce Actinides from molten chlorides in liquid cadmium by using lithium as reductant, and (iv) electrorefining to increase the purity of Actinides recovered. Both denitration and chlorination steps are pre-treatment processes prior to the application of the pyrometallurgical process.

The principle of the reductive extraction with the subsequent step of electrorefining is schematically drawn in Figure 6.9. The electro-refiner is a steel vessel that is maintained at 775 K (500 °C). Liquid LiCl-KCl electrolyte in the electro-refiner contains about 2 mol% of the Actinide chlorides. The Actinide solution (in liquid cadmium) is inserted into the electrolyte and connected to the positive pole of a dc power source (anode). The negative pole of the power source is connected to a cathode immersed in the same electrolyte. The cathodes are simple steel rods. About 80% of the Actinide metals is electro-transported from the anode to the cathode rods, where it deposits as nearly pure metal along with a relatively small amount of rare earth fission products⁴⁷. All the products are retorted to remove salt (and Cadmium from the Cadmium electrode). Ingots from the retort are blended to appropriate composition, and recast into special fuel pins. The fission products, with the exception of Tritium, Krypton and Xenon, accumulate in the electro-refiner during processing, and some noble metal fission products are removed with the anode after each batch of fuel has been processed. The three gases are released into the process

⁴⁵In the F-EA, the Actinide residue consists mainly of Thorium, Protactinium, Uranium and a very small amount of TRUs, whereas in a PWR it is mostly TRUs.

⁴⁶ We expect this method can be extended to the extraction of Thorium and Protactinium.

⁴⁷ In reprocessing F-EA fuel, the complete removal of fission products may not be necessary since their effect on the neutron economy is much less in a fast neutron spectrum than it is in a thermal spectrum.

cell which has an argon atmosphere. They are recovered at high concentrations by the cell gas purification system.

Several dozen batches of fuel are processed in a "campaign". At the end of a campaign, the salt in the electro-refiner is treated by a series of steps to remove active metal fission products, particulate noble metals, and any oxide or carbide impurities for incorporation in high-level waste forms. The salt and its associated Actinide chlorides are returned to the electro-refiner. The Actinide inventory in the electrorefiner amounts to about 20% of the Actinide elements fed; this must be recovered to achieve more than 99.9% overall Actinide recovery. A non-metal and a metal waste form will accommodate all of the high-level wastes. The non-metal waste form will contain Samarium, Europium and Yttrium; the halogens and chalogens; the alkali, and alkaline earth fission products; and a small amount of excess salt generated in the process. The Actinide content of that waste form will be exceptionally low (less than 1 part in 10⁶ of the Actinides in the fuel that is processed). The only significant long-lived activity in this waste will be Se-79, I-129 and Cs-135: the total alpha activity should be less than 10 nCi. g⁻¹. Metal wastes from the electro-refiner - noble metals, cladding hulls and salt filter elements - will be combined with any process scrap such as broken electrodes and the rare earths from the salt purification process in the metal waste form. The metal waste form will have a very low Actinide content, because of the effective Actinide recovery in the pyro-metallurgical process, but its Actinide level will not be quite as low as that of the non-metal waste form. This whole process can be made continuous, and thus can take place in a matter of only a few hours.

Pyro-processing offers a simple, compact means for closure of the fuel cycle, with anticipated high decontamination factor (> 99.9%), minimal production of highlevel radioactive waste, and significant reductions in fuel cycle costs. In addition, mainly from the weapons proliferation viewpoint, it offers an advantage over the PUREX and/or TRUEX methods, in that there is only partial removal of the fission products. Even though the process is based on the use of a metallic fuel alloy with nominal composition U-20Pu-10Zr, we believe it can be readily adapted to the EA fuel cycle without much efforts.

The final content of the HLW stream coming from the EA fuel reprocessing is mainly FFs, with only traces of Actinides. The volume generated is about 5 m³ per ton of fuel. The following step is to concentrate the aqueous raffinate and to transfer it to an intermediate storage of the reprocessing plant. The volume of the concentrate will be about 1 m³/t. of fuel and the usual intermediate storage are tanks of suitable

stainless steel such as to minimise the acid waste corrosion. To prevent the highly active liquid from boiling a redundant cooling system is required. Then, the concentrate is cooled for a period of about 10 years in order to reduce the heat generation by more than an order of magnitude before proceeding to waste solidification. Among the fission fragments, excluding the short lived and stable elements, there are a few elements which are medium lived (30 years, ⁹⁰Sr, ⁹⁰Y, ¹³⁷Cs, etc.) and some others (⁹⁹Tc, ¹³⁵Cs, ¹²⁹I, etc.) which are long lived (Table 6.1). Since Actinides are essentially absent from the HLW concentrates the policy we proposed to follow is to store in man-watched, secular repositories for several centuries the medium lived, in order to isolate them from the biosphere and to promote a vigorous research and development of methods for incinerating the bulk of the long lived FFs. The EA is an efficient tool to incinerate these wastes at the price of fraction of the neutron flux [6], but alternatively dedicated burners can be used.

In parallel with the R&D on incinerators, development on solvent extraction methods of long lived FF, which in some cases may additionally require isotopic separation, should be promoted, the goal being to virtually eliminate the need for Geological Repositories.

After the concentrates will be cooled down for the 10 years period and the longlived FF extraction applied for later incineration the wastes will be solidified by using well known techniques. For instance by calcination and vitrification. The first step allows to get waste oxides and in the second step glasses are obtained by melting the waste oxides together with additives such as SiO₂, B₂O₃, Al₂O₃, P₂O₅, Na₂O, and CaO. Borosilicate glass is the most studied solidification product but others like phosphate glass, glass ceramic, etc. are also used. When the solidification process is finished the wastes are ready for disposal in the appropriate secular repositories.

6.4 - Spallation induced Radio-nuclides. In addition to the radioactive waste produced in the Fuel and in a minor extent in the Breeder, substantial amounts of radio-nuclides are produced by the spallation target. As pointed out they divide roughly into two batches, those which remain inside the molten Lead and those which are either gases or volatile and which can be found in the neutral filling gas of the main vessel. These last compounds are collected from the gas and stored in an appropriate way in order to avoid leaks in the biosphere (paragraph 5.8). The relative ingestive radio-toxicity of the various components of the Spallation target are given in Figure 6.10. Following Table 5.9 spallation products at 700 °C can be broadly divided into three different categories namely (1) gases or vapours in which

the contribution of ¹⁹⁴Hg (751.9 y, 123 g/(GW_e × year)) is largely dominant in size and duration; (2) volatiles which, after a few years, are essentially dominated by to ²⁰⁴Tl (5.466 y, 114 g/(GW_e × year)), (3) inter-metallic combinations (alloys) with the molten Lead which at short times, shows a leading contribution from ⁹⁰Sr and, at longer times by ²⁰²Pb (7.59 × 10⁴ y, 614 g/(GW_e × year)). The radio-toxicity of the spallation products is by no mean negligible: at early times it is about 10⁻³ of the total radio-toxicity produced. At the end of the Secular repository time for FFs, the effects of ¹⁹⁴Hg exceed all other contributions until about 2,000 years. There is no major difficulty in extending safely and economically the storage of about 2.3 kg/(GW_e × year) of Mercury collected as vapours from the top main Vessel up to about 2000 years. Note that at least in the present design, the molten Lead of the Target region is directly mixed with the big volume (≈ 1000 m³) which constitute the main coolant. Therefore at least the elements which remain inside the liquid are largely dispersed. They will follow the fate of the Lead at the time of final decommissioning of the installation.

We finally remark the existence of another lead isotope, 205 Pb (2.21 × 10⁷y) which is abundantly produced by neutron capture of 204 Pb, namely 3.54 kg/(GW_e × year) in the target region and 23.15 kg/(GW_e × year) in total, and fortunately it is also rather inoffensive, since it is very long lived and it decays by K-conversion with an energy release of 51 keV mostly in the form of neutrinos.

6.5 - Radio-toxicity emitted in the Environment. Nuclear power production is based on the concept that pollutants and toxic materials are retained within the plant and in total isolation from the biosphere. The limited mass of such products makes it possible to achieve such a goal. Mining process however cannot retain all products and a significant amount of radiation is emitted in the biosphere during preparation of the fuel. Likewise in the reprocessing of the spent fuel some radioactive elements are currently re-emitted in the biosphere. Finally the ultimate storage of such materials (geologic repository) have raised some question on the ability of isolating them from the biosphere for times which largely exceed what can be considered an experience based retention. The EA concept strongly reduces such environmental impacts, when compared to the present reactor technology. We examine these points in turn.

(1)- *Mining.* Thorium is largely present in the Earth's crust, but in small concentrations. In addition several minerals exist, which have an excellent concentration of Thorium and which can be exploited economically. The

primary choice is the monazite, which is a phosphate of Cerium and other lantanides, containing a variable amount of Thorium and Uranium in a solid mixture. Usually the Thorium concentration is of the order of 10% but some mineral may reach as much as 20% by weight. Uranium minerals are usually much less rich, its concentration being in the best cases of the order of 0.2%. Incidentally one can remark that the solubility of Thorium is 1000 times smaller than the one of Uranium. Taking into account that Thorium burnt in the EA has an energetic yield which is 250 times larger than one of natural Uranium destined to PWRs, we conclude that the relative mining effort is reduced by a factor of the order $250 \times 50 = 12500$ times for a given produced energy. Starting with mineral containing 10% of Thorium by weight we need to dig only 70 tons of mineral to produce GW_e × year. For comparison and for the same energy produced the standard PWR methodology would require 0.875 10⁶ ton of mineral. In the case of Coal, the mass of fuel (TEC) is 4.24 10⁶ ton.

A pure Thorium mineral out of which the totality of Thorium is extracted will produce tailings with a negligible radio-toxicity after some sixty years, since all descendants of 232 Th have short decay lifetimes. Their evolution is governed by the 5.7 year half-life of 228 Ra. Furthermore there will be no risk associated to Radon, since 220 Rn has a half-life of 55.6 seconds and it decays before escaping the minerals. As pointed out by Schapira [5] the situation in reality is somewhat more complex, mainly because the monazite, which is the primary source of Thorium is generally mixed with some Uranium contamination. Such a contamination is strongly source dependent, as shown in Table 6.2, taken from Ref. [5]. Assuming somewhat pessimistically that the Uranium content is about 10% of the one of Thorium and that the long lived toxicity and Radon contamination are primarily due to Uranium, we conclude that the radio-toxicity produced at the mine is in the case of an EA about 250 / (10% = 2500 times smaller than the one of today's PWR for a given energy produced.

The UNSCEAR report [7] has estimated that the level of exposure of individuals to mining for today's PWRs amounts to about 1.5 man Sv $(GW y)^{-1}$ as local and regional component and to 150 man Sv $(GW y)^{-1}$ as global component. We remark that according to the same report the production of electricity from Coal is estimated to result in a global collective dose of 20 man Sv $(GW y)^{-1}$. The practice of using coal ashes for production of concrete will add as much as 20,000 man Sv $(GW y)^{-1}$. Values relative to Thorium and its use in the EA for some possible mineral sources are listed in

Table 6.2. We conclude that the typical radiation exposure to public with the EA due to mineral mining for the same energy produced are much smaller than today's PWRs and also Coal burning, even if solid ashes are correctly handled.

The same report estimates the collective dose due to initial Uranium enrichment and fuel fabrication to as little as 0.003 man Sv (GW y)⁻¹. In the case of the EA it is expected to be at least 1/4 of such a number, since the burn-up is four times longer and there is no isotopic separation. The collective doses are negligible in both cases.

(2) -EA Operation. During the EA operation the fuel and the spallation target volumes are kept strictly sealed. Indeed also for proliferation protective measures it is recommended that such volume be opened only in occurrence with the re-fuelling, namely once every about five years and only by a specialised team. While the fuel is safely sealed, the Lead coolant produces a significant amount of radioactive products, some of which remain in the liquid phase, but others are either gaseous or volatile and are found in the neutral gas (Helium) with which the main Vessel is filled. These volatile compounds are summarised in Table 6.3, extracted from Table 5.9. Some of these are noble gases and Tritium which remain gaseous at room temperature. Other, mostly Mercury and Thallium can be condensed and preserved in the solid state. Some other elements will be collected by the Lead purifier. In view of its small amount involved we believe that the gaseous elements can be released in the atmosphere. The collective effective dose per unit energy release is given by the UNSCEAR report [7] and summarised in Table 6.4. It is assumed that gases are released every 6 months, without cool-down period. A short cool-down will dramatically reduce the effects of ¹²⁷Xe (52.63 d) and it is recommended. The total local and regional doses are 0.42 man Sv (GW y)⁻¹. The global doses, integrated over 10,000 years, following the convention of Ref. [7] are of 0.18 man Sv $(GW y)^{-1}$. Both values are dominated by the effects of Tritium.

The rest of the solid high activity waste from the spallation products (dominated by Mercury and Thallium) has a substantial ingestive radiotoxicity (Figure 6.10) and it should be carefully accumulated and destined to the repository.

(3)-*Fuel reprocessing* has to deal with the very large radioactivity of the spent fuel. Since the techniques are not different that those generally in use, we can

make direct use of the estimated collective doses of Ref. [7] (Table 6.5), taking into account the differences in stockpile of the radio-nuclides produced (see Table 5.8). It is however assumed that both ¹⁴C and ⁸⁵Kr are extracted during reprocessing and sent to the repository for cool-down. Separation of ⁸⁵Kr can be performed cryogenically according to a well documented procedure [84]. Also ¹⁴C once reduced in the form of CO₂ can be extracted on the same time by the same method.

The total doses to members of the public are summarised in Table 6.6. Total global dose truncated at 10,000 years is 0.6 man Sv (GW y)⁻¹, namely for the same energy produced about 0.003 of the one of an ordinary Reactors [7] — without counting occurred criticality and melt-down accidental releases, avoided by the EA, (\approx 300 man Sv (GW y)⁻¹ — and about 0.03 of the alternative of Coal burning, even if solid ashes are correctly handled.

6.6 - Conclusions. Realistic schemes are possible in which the spent Fuel from the EA is "regenerated" for further uses. Separation of the fuel materials into two streams is performed, the Actinide stream destined to the fuel fabrication and the FFs stream which is destined to the Secular repository. After 500 years the radio-toxicity for unit energy produced of the EA is about 20,000 smaller than the one of a PWR with a "throw-away" cycle. Incineration with the help of neutrons of some of the critical, long lived radio-nuclides can strongly reduce the radio-toxicity of the waste beyond 500 years. If sufficiently diluted it could be also let "die away" without incineration since it can be made to satisfy the requirements for Class A repository. Note also that at that time the residual ingestive radio-toxicity is comparable with the one of the Thorium metal burnt in the EA.

An essential element in the clean disposal of the spent fuel is the small leakage of Actinides (mainly Uranium) into the FFs stream. A level of 100 ppm. or better is required. We believe that it is within the state of the art, eventually with a few improvements.

An important source of radio-toxicity are the spallation products due to the proton beam interacting with the molten Lead target. A specific element of concern is ¹⁹⁴Hg which is the main surviving source of toxicity of the EA in the period of time between 500 and 2000 years. It can either be preserved far from the biosphere that long or, alternatively, incinerated, following the fate of the Actinides inside the EA. Unfortunately the relevant cross sections are only poorly known but they should be measured soon [6].

An experimental test of the feasibility of incineration with neutrons in a Lead diffuser [6] is in preparation at CERN. Would it be successful it could offer the right technique in order to eliminate also the modest amount of long lived radio-toxic elements produced.

Likewise important is the total radioactivity doses to members of the public due to operation. Total global dose of the EA truncated at 10,000 years is estimated to be 0.6 man Sv (GW y)⁻¹, namely about 1/330 of the one of an ordinary Reactors for the same energy produced (200 man Sv (GW y)⁻¹)— without counting occurred criticality and melt-down accidental releases, avoided by the EA (\approx 300 man Sv (GW y)⁻¹)— and about 1/33 of the alternative of burning Coal (\approx 20 man Sv (GW y)⁻¹), even if solid ashes are correctly handled.

Table 6.1 - Fission fragments' activity after 1000 years of cool-down in the Secular Repository. Values are given for 1 GW $_e\times$ year.

Radio-	1/e Life	Mass	Other Isotopes	Activity @ 1000 y	Ingestive Toxicity	Dilution Class A
Isotope	years	(kg)	(kg)	(Ci)	(Sv) $\times 10^3$	(m ³)
129I	2.27E+7	8.09	3.48	1.43	19.58	178.47
⁹⁹ Tc	3.05E+5	16.61	—	284.29	27.67	947.65
126Sn	1.44E+5	1.187	1.783	33.79	3.20	9.65
135Cs	3.32E+6	34.12	66.77	39.32	9.87	39.32
⁹³ Zr	2.21E+6	26.11	99.11	65.64	2.38	18.75
⁷⁹ Se	9.40E+5	0.30	3.02	2.06	0.745	0.59

Table 6.2 - Uranium and Thorium content in percent [5] and levels of population exposure for typical Ores [7].

			Ratio	Local	Global
Source	UO_2	ThO ₂	U/Th	Sv (GW y)-1	Sv (GW y) ⁻¹
Italy	15.64	11.34	1.38	$8.28 imes10^{-3}$	0.828
Sri Lanka	0.10	14.32	0.007	4.20 ×10 ⁻⁵	$4.2 imes 10^{-3}$
California	6.95	4.22	1.64	9.84×10^{-3}	0.984
India	0.29	9.80	0.029	$1.74 imes10^{-4}$	0.0174

	Gas at Room Temperature				
	Mass	1/e	Boils at		
	(g)	Lifetime	°C		
³ H	1.435	17.83 y	-252		
³⁹ Ar	0.336	389.0 y	-186		
⁴² Ar	0.336	47.57 y	-186		
⁸¹ Kr	5.777	0.331E+6 y	-153		
⁸⁵ Kr	4.326	15.55 y	-153		
¹²⁷ Xe	0.37 (675) ⁴⁸	52.63 d	-108		

Table 6.3 - Radio-nuclides emitted in the neutral gas inside the Vessel by the Spallation target and the molten Lead coolant (≈ 700 °C).

S	Solid at Room Temperature					
	Mass	1/e	Boils at			
	(g)	Lifetime	°C			
³⁵ S	0.009	126.5 d	445			
⁶⁵ Zn	0.004	353.2 d	907			
⁷⁰ Zn	2.424	0.723E+15y	907			
73As	0.329	116.1 d	615			
			615			
⁸³ Rb	0.036	124.6 d	688			
⁸⁶ Rb	0.181	26.94 d	688			
¹⁰⁹ Cd	1.627	1.833 y	767			
125 _I	0.014	85.90 d	184			
¹²⁴ Sb	0.043	87.05 d	1585			
¹²⁵ Sb	0.404	3.988 y	1585			
¹³¹ Cs	0.003	14.01 d	671			
¹³⁴ Cs	0.282	2.982 y	671			
¹⁹⁴ Hg	415.9	751.9 y	357			
²⁰³ Hg	6.252	67.40 d	357			
202Tl	15.25	17.68 d	1473			
²⁰⁴ Tl	386	5.466 y	1473			
²¹⁰ Po	0.995	200.1 d	254			

⁴⁸ Total integrated production, without decay over 5 years

	Normalised	Collective dose per unit		Normalised collective		
	release (Tbq)	release (man	release (man SvTbq ⁻¹)[7]		Dose (man Sv (GW y)-1)	
		Local & Regional ⁴⁹	Global ⁵⁰	Local & Regional	Global	
³ H	521	0.0027	0.0012	0.418	0.185	
14C	_	0.40	85		_	
³⁹ Ar	0.430	7.4 10 ⁻⁶	5.0 10 ⁻⁴	9.4 10 ⁻⁷	6.38 10 ⁻⁵	
⁴² Ar	3.268	7.4 10 ⁻⁶	6.1 10 ⁻⁵	7.2 10 ⁻⁶	5.91 10 ⁻⁵	
⁸¹ Kr	0.004	7.4 10 ⁻⁶	1.8 10 ⁻²	9.2 10 ⁻⁹	2.24 10 ⁻⁵	
⁸⁵ Kr	63.6	7.4 10 ⁻⁶	2.0 10 ⁻⁵	1.4 10-4	3.77 10 ⁻⁴	
¹²⁷ Xe	3718 . ⁵¹	7.4 10-6	1.05 10 ⁻⁷	8.2 10 ⁻³	1.16 10 ⁻⁴	
Totals	4307			0.42	0.186	

Table 6.4 - Normalised, collective effective dose from locally, regionally and globally dispersed radio-nuclei during operation over a period of 10,000 years.

Table 6.5 - Normalised released dose of airborne and liquid effluents of radionuclides during reprocessing of Fuel. Values have been normalised to current practices [7].

	Process	EA/	Normalised collective		Comments
	(kg)	PWR	Dose (man S	Sv (GW y) ⁻¹)	Comments
			Airborne Effluents	Liquid Effluents	
³ H	—	1.0	0.11	0.0012	assumed same as PWR
¹⁴ C	0.0145	9.2	(7.45)		Retained
⁸⁵ Kr	21.64	10.16	(0.924)		Retained
129I	27.28	1.722	0.430		standard practices
131I	0.2924	0.458	1.37 10 ⁻⁴	_	"
137Cs	118.5	1.109	0.0188	1.22	"
⁹⁰ Sr	74.76	1.578		0.205	"
106Ru	1.147	0.074		0.207	" "
Totals			0.60	1.63	

 $^{^{49}}$ For noble gases, values are taken to be the same as $^{85}\mathrm{Kr.}$

 $^{^{50}}$ For noble gases, values are taken to be the same ones as 85 Kr, for decay over 10,000 years. 51 Periodic (every 6 months) release, without cool-down.

Source	Local and regional Doses (man Sv (GW y) ⁻ 1)	Global Doses (man Sv (GW y) ⁻¹)
Mining ⁵² , Milling, Fuel fabrication	4.2 $10^{-5} \div 9.8 \ 10^{-3}$	0.0042 ÷ 0.984
EA operation	0.42	0.188
Reprocessing (Atmospheric)	0.60	0.1
Reprocessing (Aquatic)	1.63	0.1
Miscellanea ⁵³	0.1	0.05
<i>Totals(variation over mining range)</i>	2.75 ÷ 2.76	0.44 ÷ 1.42

Table 6.6 - Summary of normalised, collective doses to members of the public from radio-nuclides released from the EA.

 $^{^{52}}$ The dose range depends on the Uranium content in the Thorium mineral. We have taken extreme values of Table 6.1.

⁵³ This includes mainly Transportation, Fuel fabrication, Solid Waste disposal. Figures are taken from Ref. [7].

Figure Captions.

- Figure 6.1 Evolution of the ingestive radio-toxicity of High Level Waste(HLW) during Secular Repository period.
- Figure 6.2 Evolution of the ingestive radio-toxicity of HLW beyond the "Secular Repository" period.
- Figure 6.3 Evolution of the ingestive radio-toxicity of the FFs for different incineration procedures.
- Figure 6.4 Radio-toxicity of the residual Actinide waste stream for different leak fractions.
- Figure 6.5 α activity of the residual Actinide waste stream for different leak fractions.
- Figure 6.6 Flow diagram of the partitioning process of spent fuel.
- Figure 6.7 High-Level Waste (HLW) reprocessing scheme.
- Figure 6.8 Flow diagram of the pyro-metallurgical process for partitioning of the residual Actinides from HLW.
- Figure 6.9 Schematic illustration of the pyro-metallurgical partitioning process.
- Figure 6.10 Relative ingestive radio-toxicity of the spallation target products.



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