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FAST NEUTRON INCINERATION IN THE ENERGY AMPLIFIER AS ALTERNATIVE TO GEOLOGIC STORAGE: THE CASE OF SPAIN

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Abstract

In previous reports [1][2] we have presented the conceptual design of a fast neutron driven sub-critical device (Energy Amplifier) designed both for *energy amplification* (production) and for the *incineration* of unwanted "waste" from Nuclear Light Water Reactors (LWR). The latter scheme is here applied to the specific case of Spain, where 9 large LWR's are presently in operation.

It is shown that a cluster of 5 EA's is a very effective and realistic solution to the elimination (in 37 years) of the present and foreseen (till 2029) LWR-Waste stockpiles of Spain, but with major improvements over Geologic Storage, since: (1) only a Low Level Waste (LLW) surface repository of reasonable size is ultimately required; (2) the large amount of energy stored in the trans-Uranics is recovered, amounting — for each of the 37 years of incineration — to a saving of about 8% of the present primary energy demand of Spain (100 MTep/y); (3) the slightly enriched (1.1%) Uranium, unburned by LWR's, can be recovered for further use; (4)Trans-Uranic waste is transformed into fissile ^{233}U, which can be used to make +20% of LWR fuel without isotopic separation and (5) the capital cost of the complete facility, including reprocessing is convincingly lower than the one of the Geologic Storage.

The volume of the ultimately Class A LLW can be further reduced by about an order of magnitude transmuting parasitically in the EA's into stable nuclear species also the two most offending long-lived isotopes, ^{99}Tc and ^{128}I. The total LLW volume ultimately required (60'000 m^3) will then be roughly $\approx$ 1% of the present LLW storage in the USA and comparable to the present capacity of the now operational "El Cabril" in Spain (50'000 m^3).

We conclude that EA-driven incineration — when compared to direct Geological Disposal — is environmentally more acceptable and economically more advantageous. Finally, no major technical barriers hinder its realisation.

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1.— INTRODUCTION

The Energy Amplifier (EA), a fast neutron driven not-critical device [1] is the result of a mature cross-fertilisation between accelerator technologies and the energy production technologies with reactions on nuclei. Particle beams accelerators have been universally used to induce nuclear reactions for investigation purposes: why not use them to produce practical amounts of energy (high temperature heat) ?

Nuclear reactions occurring in the EA can also be used to "incinerate" unwanted radioactive waste [2] from Light Water Reactors (LWR), in particular

(1) trans-uranic elements (TRU, primarily Np, Pu, Am, Cm etc.) which are destroyed by fission, generating a large amount of energy or
(2) transmute long-lived LWR's radioactive products such as Technetium (Tc^{99}) and Iodine (I^{129}) into stable, harmless isotopes.

TRU elements in metallic form are suitable fuel for the EA. The choice of the metal rather than oxides as customary in the Nuclear industry is not mandatory, but it offers significant advantages, since it is closely matched to the reprocessing technology chosen and it has been thoroughly tested in the US in the IFR programme. The EA is very effective in burning the TRU's because its neutron spectrum is very energetic, as a result of the metal fuel and metal coolant (molten Lead) environment. TRU's are instead troublesome and cannot be fully eliminated in a thermal neutron environment. Previous studies [3], for instance the one at Los Alamos by Bowman et al. [4], were generally based on thermal neutrons.

The present generation of LWR's produce large amounts of radioactive fuel waste, which are currently destined to very long ($\approx 10^6$ years) underground storage, the so-called "Geological Repository". Recycling of this spent fuel and complete burning of the transuranic elements in the EA would instead:

(1) avoid the expense and the concerns of a deep underground, long term disposal and
(2) provide large amounts of supplementary energy for future needs.

The general concept of the incineration plant will be briefly described, having in mind the specific case of Spain [5]. However similar considerations apply to spent fuels accumulated in several other countries. The spent fuel —

presently in the form of oxide fuel in Zircaloy sub-assemblies, stored in swimming pools near power stations — is processed first with a simple pyro-chemical process which can extract the TRU's in a form suitable as feed to the EA fuel cycle. This process [6] has been under study for over 30 years, mainly in Argonne (US). Its main promises are:

(1) ≥ 99.9 % recovery of the bulk of TRU's, which are all simultaneously extracted;
(2) minimum generation of extra waste ;
(3) simplicity of the process and promising economics;
(4) small scale plant which could be located in the vicinity of the LWR or of the EA. Plutonium shipping over long distances is eliminated;
(5) strong resistance to military diversion of TRU elements which are always extracted simultaneously and inherently contaminated with fission products (noble metals).

As a result, three streams of materials develop:

(1) pure Uranium (95% of mass, with ≈ 1.1 % ^{235}U), which can be re-used, for instance as LWR fuel;
(2) Fission Fragments (FF) (≈ 4 % of mass), whose initial activity is dominated by Strontium ($\tau_{1/2}$ = 29.1 years) and Cesium ($\tau_{1/2}$ = 30.2 years), which are kept in a local Cool-down Facility until they have decayed sufficiently to become low level waste (LLW) for disposal on surface or at shallow depth;
(3) the TRU[1]'s, which become the fuel for the (closed) cycle of the EA (≈ 1% of mass), i.e. they are mixed with Thorium and indefinitely recycled within the EA until they are completely eliminated. They carry the overwhelming radiotoxicity of the waste at long times (>99.995% at 1000 years).

The surviving, residual ingestive radiotoxicity at ≥ 1000 years is reduced by a factor ≈ 1/20'000 with respect to the open cycle followed by a Geological Repository. Another factor 10 of reduction (final reduction factor ≈ 1/200'000) is achieved by transmuting parasitically in the EA also ^{99}Tc and ^{129}I.

The key idea put forward in Ref. [2] has been the one of using in the EA a Thorium-TRU mixture which is much more effective in eliminating TRU's at acceptable concentrations than the conventional mixture of Uranium-

[1] More precisely these are all the Actinides different than Uranium.

Plutonium for instance in the CAPRA scheme [7]. The device operates as a TRU burner and as a ^{233}U breeder from the added Thorium. The latter can be later mixed with ordinary or alternatively depleted Uranium and it constitutes an excellent fuel for the LWR's or it can be stored for further use in an EA operated without incineration, namely as pure energy producer [1]. The EA not-critical mode is preferred over the conventional Fast Breeder (critical) Reactor, because of the risks associated to the reduced criticality window due to the presence of the TRU's (approximately ± 0.1 % in Δk) of a Fast Reactor and its more positive void coefficient related to the presence of Plutonium. Finally, Lead replaces the customary Sodium[1] coolant. As we shall see, the presence of Lead permits, unlike Sodium, also an effective transmutation of some particularly harmful FF's like ^{99}Tc and ^{129}I into stable elements.

The corresponding streams from the EA fuel reprocessing are much less in volume (≈10%) and recycled fuel can be processed through the same facility. Note that its spent fuel is already metallic and no Oxygen reduction process is required in front of the reprocessing chain. The component (3), which now includes the Thorium matrix, becomes new fuel for the EA, with the addition of fresh TRU's to be incinerated from the LWR's and of some extra Thorium to compensate for the burnt fraction; the component (2) is sent to the cool-down facility and later to the LLW disposal; the Uranium produced by the Thorium, mostly ^{233}U is recovered from stream (1) and burnt later on either in the LWR's or in the EA. Hence the only waste is stream (2).

With this procedure, the need of geological repository is virtually eliminated and a large amount of supplementary energy is generated, approximately 40% of the LWR's energy originally produced in association with the waste[2]. The surviving radio-toxicity for potential ingestion by humans of the whole waste stream from a LWR after a few hundred years is reduced to the one of the radioactivity contained in Coal Ashes for the same energy produced, for which protection measures are not customary.

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[1] One of the reasons for choosing Sodium has been also the fact that the corresponding breeding of Plutonium is enhanced by the resulting neutron spectrum, which is precisely the opposite of what we are seeking.

[2] In the case of Spain the electric power installed as LWR's is 7.1 GWatt (electric). As shown later on, incineration of the waste requires 5 EA's, each producing 0.625 GWatt (electric), corresponding to a total of 0.625 x 5 = 3.125 GWatt or + 15% increment in the total generated electric power. The present increase of electricity demand in Spain is +3 %/year; the extra produced energy will easily find a market !

More specifically, it satisfies to all norms of US Nuclear Regulatory Commission [8], low level waste (LLW) for disposal on surface or shallow depth. This classification system includes Classes A, B and C with a growing level of activity and it is designed to minimise potential exposures in both the short and long term. Essentially all medical wastes are Class A. Industrial wastes are largely Class A, but they contain some Class B and C. The LWR residual waste after incineration and after an appropriate cool-down at the plant is finally to be disposed as Classes C, B or A depending on the length of the cool-down, evolving naturally into a final Class A. The volume required to ensure safe disposal is reasonably small and it is further reduced since the two most offending long-lived isotopes, ^{99}Tc and ^{129}I are transmuted parasitically in the EA's into stable nuclear species. Referring to the case of Spain and a projected operation of the LWR's to their natural end (2029), the total LLW volume ultimately required (60'000 m^3) is roughly $\approx$ 1% of the present LLW storage in the USA and comparable to the present capacity of the now operational low and medium waste storage "El Cabril" in Spain (50'000 m^3).

Our proposed procedure is definitely attractive for many reasons, amongst which:

(1) from the *economical point of view*, since the projected capital cost of the incineration facility is conservatively less than the one of a geologic repository for a given amount of fuel and this initial investment pays also for a major fraction of the cost of the additional energy produced, which becomes very profitable.

(2) from the *environmental point of view*, since it strongly reduces the burden of radioactive waste to future generations (f.i. the half-life of Plutonium which is naturally 25'000 years, is reduced in the EA to few years).

(3) from the *safety point of view*, since the device is not "critical", namely it keeps the waste at all times in conditions ($k \leq 0.95$) which are essentially the same as the ones in the projected geologic repository. The risks of an accidental "melt-down" are also removed by the "swimming pool" arrangement with natural convection.

The basic underlying physics issues of the energy production and incineration have been completely tested in two experiments [15] [16] carried out at CERN. We are now ready for a demonstration plant of reasonable size (100 $\rightarrow$ 250 MWatt). The final 1500 MWatt incineration unit will closely resemble the demonstration unit, except of course the much larger power.

We believe that there are no major technological barriers to the realisation of the EA and that the schedule could be short enough to offer a realistic alternative to the difficult problem of the radioactive waste from LWR's in Spain and elsewhere. In particular the innovative accelerator which is needed to initiate the incinerating nuclear cascades is a direct extrapolation of the modern accelerator technologies developed at CERN and elsewhere. In particular the presently operating LEP200 superconducting accelerating cavities could be modified to operate with protons rather than electrons and easily adapted to the requirements of the EA [9].

We believe that the demonstration phases could be completed say, by 2010÷2015, permitting soon after that date the deployment of the 5 EA units for Spain. The incineration process could be therefore completed by 2045÷2050 [17], well matched to the perspective end of the present Spanish LWR's which is 2029. We remark incidentally that the EA's produce little waste, since they auto-incinerate their own TRU's. Therefore they are an excellent candidate to substitute the present LWR's, though continuing with nuclear power after the natural closure of the present phase, including incineration, is not only a technical but a vastly political decision, for the moment, at least premature. In the case of the EA's there exist of course also other considerations which speak in its favour when compared to LWR's, namely intrinsic, deterministic safety, smaller and efficient use of an abundantly available natural fuel and no appreciable proliferation risks. The EA's are in direct competition with Magnetic Fusion, since both have a closely similar environmental impact [18], but the EA at much lower cost, faster time schedule and simpler technology.

If more than the minimum number of EA's needed to complete incineration were eventually deemed necessary, the LWR's waste could still be used as a "starter" and eventually eliminated. But then only the minimum amount of waste should be used, proceeding as soon as possible to a pure energy production phase with Thorium and ^{233}U [1]. A larger Breeder could be used, increasing the transformation rate of waste into ^{233}U. We have estimated that the Spanish waste should then suffice to start about 20 EA's, capable of producing 12.5 GWatt of electricity, commensurate to the full extrapolated need of nuclear power of the country by that distant future time. We remark that the EA's can also be started with other methods, like for instance standard enriched Uranium.

2.— THE LWR'S WASTE IN SPAIN.

2.1.- Waste stockpiles. There are today in Spain nine LWR Reactors with an installed nominal electric power capacity of 7.1 GWatt. With the assumption that the useful lifetime of the nuclear plants will be of 40 years, it is estimated [5] that the accumulation of irradiated fuel elements will be (2029) of the order of 11502 elements of the type PWR and 8364 of type BWR. As a simplificative assumption one can assume that one fuel element of the type PWR is equivalent to three elements of the type BWR. Therefore the final stockpile will be equivalent to about 14400 elements of irradiated fuel of type PWR. At the end of 1995 the number of equivalent fuel elements were 3780. The expected increment of the present stockpile at the projected end of the commercial life of the Reactors, is therefore a multiplicative factor 3.81. The main parameters of the PWR elements are listed in Table 1.

Table 1. *Main parameters of a single PWR Fuel Element from Spain [5].*

Number of fuel rods	$17 \times 17 = 289$	
Dimensions	$0.21 \times 0.21 \times 4.06$	m^3
Total weight	668.6	kg
- Uranium	461.41	kg
- Inconel 718	14.875	kg
- 304SS steel	14.875	kg
- Zircaloy	115.17	kg
- Oxygen	62.07	kg
Initial ^{235}U enrichment (%)	4.10	
Average burn-up	40.0	$GW \times d/t$

The effects of burning on the isotopic composition has been calculated with the help of ORIGEN [10]. After 15 years of cool-down, the approximate composition of the Uranium Oxide fuel is listed in Table 2. For the isotopic composition we refer to Table 10.

The present policy for the disposal of all this waste — ultimately 9628 tons of spent fuel — after encapsulation in units of approximately 15 tons each, once loaded with the spent fuel is the one of a Geological Storage. The estimated number of units is about 3600. It is fair to say that the underground disposal of such a large amount of radioactive material is at the development

stage and that considerable uncertainties still persist on the ultimate cost and complexity of the method, not without mentioning the strong local opposition in the general public. The effects on the neighbouring rock of the heat produced by the radioactive materials (in the cave T > 100 °C) also need to be better understood and mastered. The containment of radioactive capsules can be guaranteed to remain intact inside the repository only for a limited period of time, of the order of 1000 years. Beyond this limit, significant amounts of radio nuclides are expected to leak into the environment, in particular 100% of the long lived ^{129}I and a major fraction of the ^{99}Tc [11].

Table 2. *Total Stockpiles, now (1995) and projected (2029).*

Element	Fraction (%)	Stockpile 1995 (ton)	Stockpile end (ton)
Uranium	94.771	1652.918	6296.832
Neptunium	0.059	1.032	3.931
Plutonium	0.951	16.594	63.216
Americium[1]	0.092	1.610	6.134
Curium	0.00217	0.038	0.144
Fission Fragments	4.124	71.933	274.032

Note that this last concern will not be any less for the LLW disposal proposed as an alternative. Hence we propose to transmute also these two isotopes into stable elements as part of the incineration programme. The remaining very long lived elements ^{93}Zr, ^{126}Sn, and ^{79}Se (see Table 5) are of smaller stockpile and present less concern, since, they can be more easily retained. The main remaining concern is ^{135}Cs [11], which has a relatively small, constant total radiotoxicity of 2.41×10^5 Sv for quantities of Figure 1. In comparison the total ingestive radiotoxicity of the TRU's for the throw-away open cycle is about 10^{10} Sv after 10^4 years, 3×10^8 Sv after 10^5 years and 3×10^7 Sv after 10^6 years. Transmutation of ^{135}Cs into stable ^{136}Ba would be possible, but only after the strongly radioactive ^{137}Cs contaminant has decayed, namely several hundred years from now. We believe that ^{135}Cs is hardly a problem, anyway covered by the world-wide applied specifications for the LLW [8] and that we intend to follow.

[1] Cool-down dependent; 15 years of cool-down assumed.

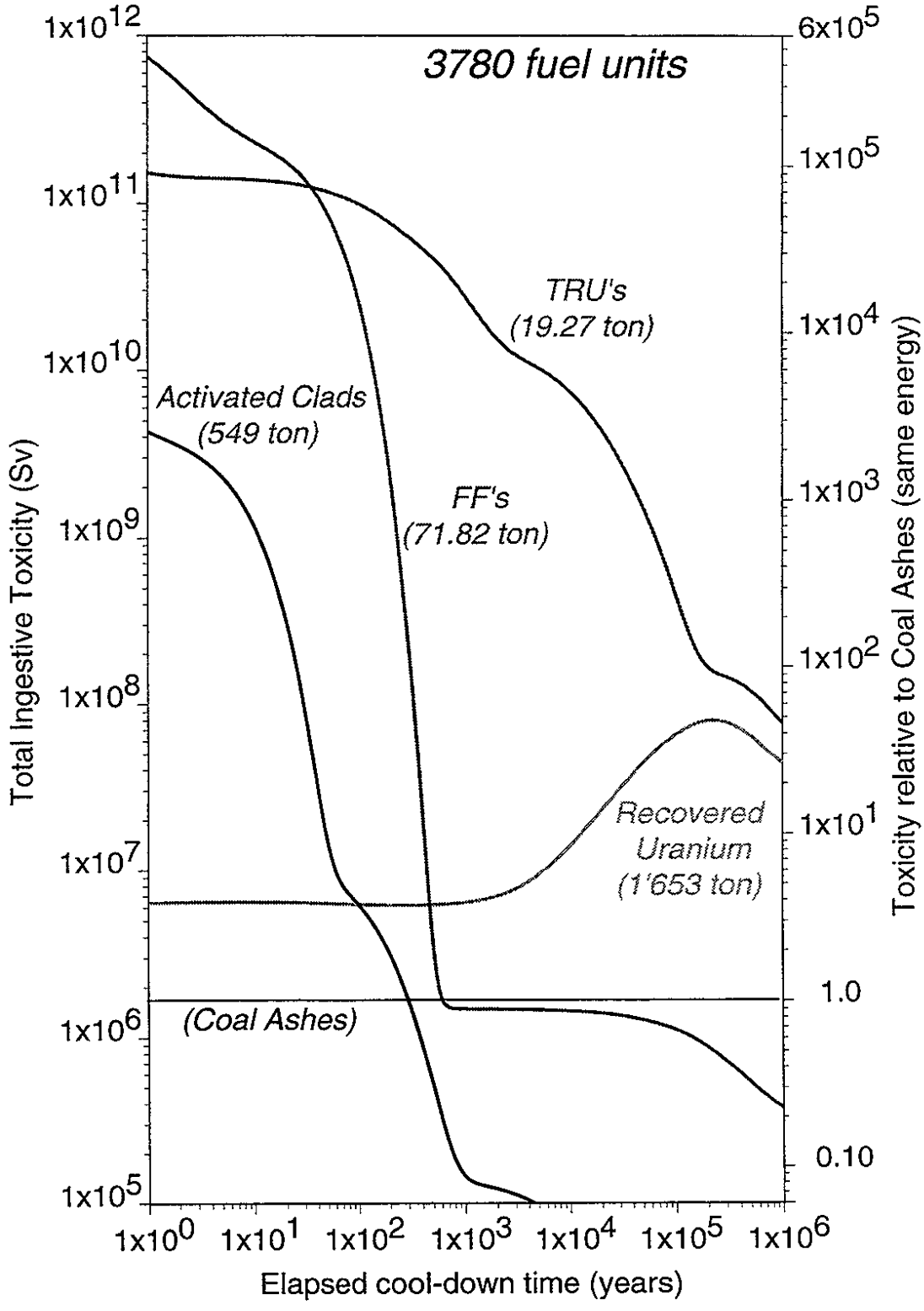

Figure 1. Radiotoxicity in absolute units and referred to Coal Ashes for the same delivered energy of the presently accumulated LWR Waste in Spain. For the perspective end of the life of the Reactors, toxicities and stockpiles must be multiplied by a factor 3.81. TRU's, which are ≈ 1% of the Waste, dominate the toxicity at long times. About one order of magnitude reduction in the long times toxicity of FF's is achieved by transmutation.

The criticality coefficient of the sub-critical, fissile material storage is set to be $k \leq 0.95$ and it must be preserved under the most exceptional evolution of the environment during hundred of thousands of years. As we shall see the same condition $k \leq 0.95$[1] can be preserved at all times during the incineration process in the EA. Therefore the criticality associated risks of the incineration process are at all times comparable to those deemed safe for the Geologic Storage, except that in our case they have to be maintained for a comparatively short time and are under direct and continuous instrumental watch.

The ingestive radiotoxicity [19] of the already accumulated waste in Spain, is shown in Figure 1 as a function of the elapsed time. In order to extrapolate these figures to the perspective end of the exploitation of the existing Reactors (2029), they must be multiplied by a factor 3.81.

The main feature of relevance evidenced by Figure 1 and which justifies our proposal of incineration is the fact that *the TRU's, which represent only 1% of the total waste mass, dominate the radiotoxicity* to times which are extremely long (i.e. about 20'000[2] times the Fission Fragments (FF) after 1000 years, and 100 times FF's after 200,000 years). Incineration of such relatively small amount of TRU's materials (19.27 tons so far) — which, as we shall see, are readily separable from the rest by electro-refining — makes a lot of sense, also in view of the enormous potential energy which they contain. Instead the Activated Clad and the FF's which are the main "waste" in the case of incineration, reach a level of acceptable radiotoxicity for surface disposal with the well proven and existing technology of the LLW after a few hundred years — period over which it is technically possible to ensure safe containment. Instead, in the case of the very long storage of the TRU's, any prediction is for the moment, to say the least, speculative. Incineration of the TRU's is therefore extremely beneficial in reducing the environmental concerns associated to the LWR's waste.

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[1] The actual choice of the operating k for the EA depends on the fraction of energy recirculated through the Accelerator. The thermal gain (produced power/beam power) is roughly $2.5/(1 - k)$. For $k = 0.95$, $G = 50$. Assuming a thermodynamical efficiency of 0.43 [1] and an accelerator efficiency of 0.5, the fraction of electric power needed to run the accelerator is then $1/(50 \times 0.43 \times 0.5) = 9.3$ %. In the paper we use $k = 0.97$, that we believe safe enough.

[2] As already pointed out another order of magnitude reduction is achieved by transmutation of specific fission fragments.

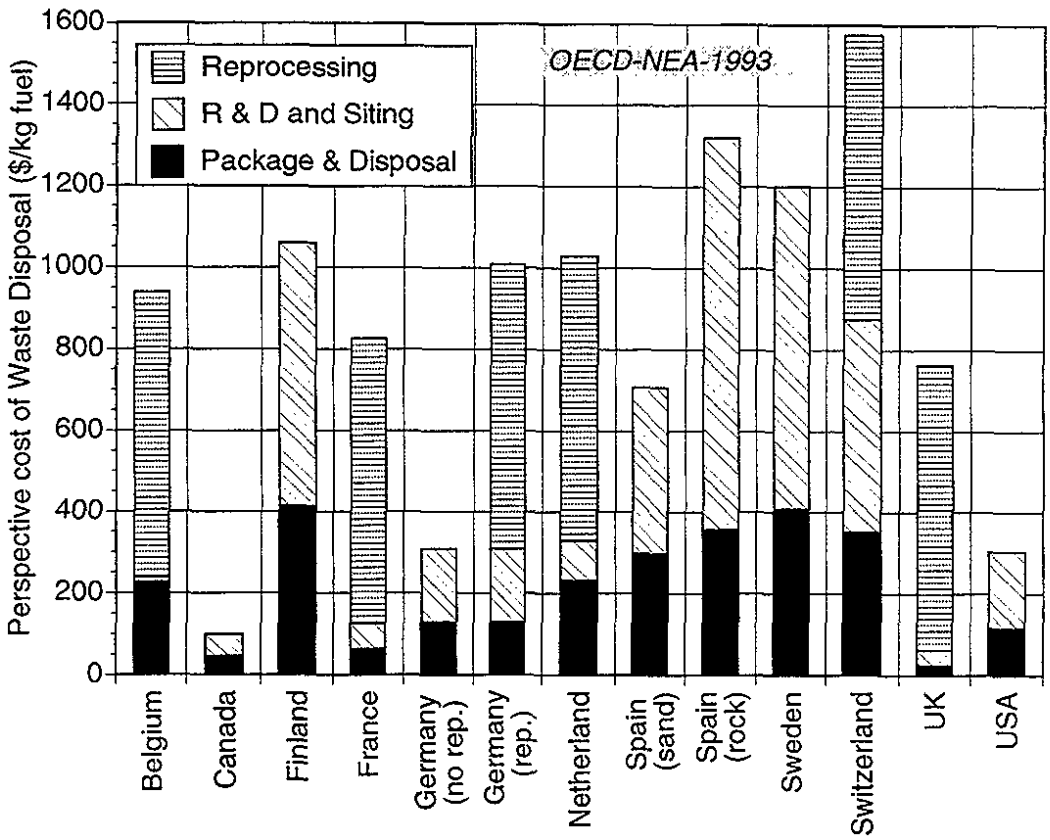

Figure 2. Estimated costs of geologic waste disposal for unit fuel mass. The actual package and Disposal and the Preparation and R & D costs are indicated. Reprocessing is added, whenever appropriate (700 $/kg). From Ref. [12].

2.2.- *Financial benefits of TRU incineration.* Elimination of the waste is primarily a political problem, strongly felt by a wide public. Deep underground storage over geological times is technically possible, but it is not without acceptability problems. Notwithstanding, we shall address the financial aspects of Actinide elimination with the help of the EA. The estimated costs of geologic storage are shown in Figure 2, taken from a OECD Report [12]. Underground storage may be performed either with or without reprocessing. Reprocessing is presumed to offer added insurance to containment, since it allows vitrification and optimal packing according to the nature of each element, but at extra cost. We remark the large amount of R & D and Siting costs before the actual Package and Disposal and the added costs due to Reprocessing. In our view these figures are in many instances underestimates, since for instance a full bench-mark study of all the consequences of deep storage of sub-critical mixtures has not been performed and the public opposition is still hard to quantize in terms of added costs.

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

In order to get a price estimate for a corresponding Incineration Facility we have used as reference two preliminary estimates of the cost of a single 1500 MWatt (thermal) EA, including infrastructures and electricity production:

(1) an analytic cost analysis [13] based on the actual conceptual project [1] of 941 M$ for one EA plant, leading to a total investment of 4.7 B$ for the 5 units required to incinerate the full Spanish waste in 37 years.

(2) an extrapolated cost estimate of the STC Committee [14], based on the cost of French LWR's (10 B FF, i.e. 1.8 B$[2]) for a thermal power[3] of 3800 MWatt. In this estimate it is suggested a + 20% precautionary cost increase for the EA, leading to the figure of $7500/3800 \times 1.2 \times 1.8 = 4.26$ B$ for the 5 EA's.

Substantial cost reductions are promised by pyro-processing, when compared to aqueous methods (PUREX). Notwithstanding we shall take the nominal cost of a 1500 t/year aqueous plant to be 5 B$ and scale it down linearly to the required total reprocessing rate of 300 t/y, leading to a ≈ 1 B$ capital investment. We add also 0.25 B$ for the Cool-down facility and 0.45 B$ for the LLW disposal[4]. The total investment required is then 5.95 - 6.4 B$, probably overestimated. This has to be compared with the 7.7 B$ indicated in the estimates for the Geological Repository for the full Spanish waste.

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[1] It should be pointed out that cost estimates vary widely from country to country. See for instance OECD report [12]. For instance in the case of Switzerland, where only 5 reactors are in operation, the much higher figure of 14 B SWF (11 B$) has been given, for a complete storage by year 2061.

[2] We assume: 5.5 FF = 1 US $.

[3] Thermal power is relevant, since one talks about incineration.

[4] The cost of the Spanish LLW repository "El Cabril" has been evaluated in 0.4 B$ for a storage capacity of 50'000 m^3. Of the cost, about 30% are the initial investment and the rest is waste management, disposal and maintenance.

Therefore the capital investment for the alternative of an incineration facility is likely to be significantly cheaper and it could then be financed entirely out of the saved costs of the Repository.

The fission elimination process applied to the full TRU stockpile is largely eso-energetic. The actual incineration process of the TRU's involves in addition a significant amount of Thorium and its transformation into fissile ^{233}U, which in part is burnt and in part is recovered at the end of the cycle

Table 3. *Energetic (thermal) yield of TRU incineration with EA.*

Source	So far (1995)	At end life (2029)	
Stockpile TRU	19.27	73.42	ton
Energy from TRU's	1.565×10^9	5.963×10^9	GJoules
Energy from ^{232}Th->^{233}U[1]	1.017×10^9	3.876×10^9	GJoules
Energy from ^{233}U (LWR)[2]	5.478×10^8	2.087×10^9	GJoules
Total Recovered energy	3.130×10^9	1.193×10^{10}	GJoules
Equiv. Barrels of Oil (BOE)	5.482×10^8	2.089×10^9	
Commercial Value[3] @20$/BOE	10.96×10^9	41.78×10^9	US-$

and could be later burnt in the EA itself or used to make an additional fuel for a LWR. These two additional, Thorium related processes roughly double the energy generated in the incineration process with respect to the straight burning of the TRU's. Relevant figures are listed in Table 3.

The total energy recovered by the incineration process for the full perspective waste from Spain is then about 1.193×10^{10} GJoule, equivalent to about 2.85×10^8 Tep (Ton equivalent Oil). If incineration is carried out over 37 years, during that time there will be an effective saving of 7.7 Million Tep/year, which is roughly 8 % of the present primary energy requirements of Spain[4], with presumably a corresponding reduction of the energy imports.

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[1] Energy produced in EA because of breeding and fissions of ^{233}U.

[2] Energy recovered by ^{233}U production as fuel for LWR's.

[3] The corresponding value of the energy is about 3.5 $/GJoule.

[4] The primary energy requirement of Spain is estimated to be of the order of 100 MTep (Million tons equivalent oil) > the approximate fractional contributions for different energy

This is considerable, for instance equal to the present natural gas consumption of Spain.

An order of magnitude estimate of the perspective profit during the operation of the Incineration Facility is the following:

(1) In order to estimate the value of the produced energy we make the reasonable assumption that the 5 EA's will produce electricity, resulting in a power of $0.625 \times 5 = 3.125$ GWatt. Assuming 90% operational efficiency, the corresponding yearly energy production is $8760 \times 0.9 \times 3.125 \times 10^6 = 2.46 \times 10^{10}$ kWh. At the standard price of 5 ¢/kWh gives a gross earning of 1.23 B$/year, or a total of 49.92 B$ over the 40 years of operation of the facility[1]. Since the operating costs do not include interests for initial capital costs (the facility has been paid out of the Geologic Repository savings) and fuel, the main surviving elements of cost are the Operation and Maintenance (O & M) costs. In our estimates [13] we have arrived at the figure of 0.50÷ 0.75 ¢/kWh. For a French LWR the figure 1.03 ± 0.04 ¢/kWh is given. An average over several countries for LWR's gives the figure of 1.18 ± 0.04 ¢/kWh. Our cost estimates are slightly lower than the ones for LWR's since we take into account the longer burn-up and less frequent interventions of the EA. The O & M costs of the pyro-processing Plant are hard to estimate. We take from the aqueous case the current operating figure of 0.1 B$/year[2], corresponding to 0.4 ¢/kWh. In any case, the profit in the energy produced is very substantial, since these surviving O & M costs represent only a fraction of the commercial value of the energy produced.

(2) The recovered Uranium metal (6296.8 ton) is already enriched to 1.1% and has a commercial value of 175 $/kg, taking into account the 0.47

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sources are : 55% oil, 19% coal, 15% nuclear, 8% gas, 2% hydraulic, the remaining 1 % other sources.

[1] Though the incineration time of the Spanish waste is expected to take about 37 years, the power plant can continue to operate at comparable costs, fed with natural Thorium, which has a negligible price in the present scale and ^{233}U breeding [1]. The projected lifetime of the EA plant has been estimated to exceed 60 years [1].

[2] In Ref. [11] the costs of a large (2700 tons/year of spent fuel) Reprocessing plant are as follows: 7.320 B$ of capital cost and 374 M$/year of operation. In our case we have 9628/37 = 260 tons/year from the LWR plus about 10% of that for the EA. Our figures are therefore about 10% of those given in Ref. [11]. Notwithstanding we have taken 0.1 B$/year for O & M costs of pyro-reprocessing.

SWU (enrichment units) (51.7 \$/kg) and the saved natural uranium[1] (120 \$/kg). The total recovered value is therefore $6296.8 \times 10^3 \times 175 =$ 1.1 B\$.

(3) The ^{233}U produced by the EA (875 kg/y for 5 EA's) can be used to make enriched fuel for LWR's with a mix of ^{235}U (1.1%) and ^{233}U (2.8%), which has very similar properties as the 4.1% enriched fuel used in the Spanish LWR's. The resulting diluted Uranium mass is 31.25 tons, with an estimated commercial value of 1124 \$/kg (6.09 SWU) amounting to 35.12 M\$/year or 1.3 B\$ for the full stockpile of waste from Spain.

In conclusion incineration of the LWR's waste from Spain makes a lot of sense both environmentally and financially. It would be a real pity to bury all that energy in a deep underground hole !

[1] Unit cost of natural Uranium : 50\$/kg.

3.—TRANSFORMATION OF THE SPENT FUEL

3.1.- Strategy Issues. The main justifications for Incineration rather than Deep Storage have been given in the previous paragraph. The next question is on how to proceed with a practical scheme. In our proposal, while the TRU's are incinerated as efficiently as possible with the help of the EA, the FF's are considered as "waste". Two main properties characterise the environmental impact of such a FF-dominated waste, i.e. radiotoxicity and heat, both gradually falling functions of the elapsed time from the moment of the discharge from the LWR. In both cases, as seen from Figure 1, behaviour is settling to a constant, low level after a few hundred years. Therefore we have chosen a length of display of 1000 years and a linear scale, which is more sensible than the enormous logarithmic, time span of Figure 1. The amounts of FF's chosen are the same as in Figure 1. They correspond to the full processing capacity of a single EA over 40 years and roughly to the amount of waste accumulated to date in Spain.

The (ingestive) radiotoxicity of the FF's of the present LWR discharge from Spain (as of end 1995) is shown in Figure 3. We have separated the stream in the three main components of different general physical properties: (1) Gases and Volatiles, dominated at short times by Strontium and Cesium activities, (2) Noble Metals and (3) Rare Earths. Actually in the chosen reprocessing procedure, Gases will escape naturally at the beginning and Volatiles and Rare Earths will come out in one common stream. They may need to be separated if some transmutation has to be performed to reduce the long lived radionuclides in the waste disposal. The main conclusion is that during the active life of the Plant (≈ 100 years), most components become asymptotic, with the exception of the Strontium and Cesium which dominate the toxicity at short and medium times.

The produced decay heat is shown in Figure 4, again as a function of the time from discharge from the LWR. Again the behaviour in the few hundred years scale is dominated by the activity of Strontium and Cesium. The amount of heat produced, initially of the order of 10 MWatt, will then decay to less than a kWatt after about 400 years.

The whole procedure from the LWR discharge to the definitive disposal of the FF's waste will realistically take a considerable length of time. This

elapsed time is not only necessary but also beneficial, since the radiotoxicity and decay heat will be correspondingly reduced. The main steps are:

(1) after discharge it is customary to keep the spent fuel in the swimming pool near the LWR for a relatively long time. Actually at present this provisional storage, perhaps in the lack of a clear final decision on what follows, is building-up to a considerable amount. Even in the most optimistic estimates on the possibility of our scheme, the fuel will have to wait for as long as 20 years before being reprocessed.

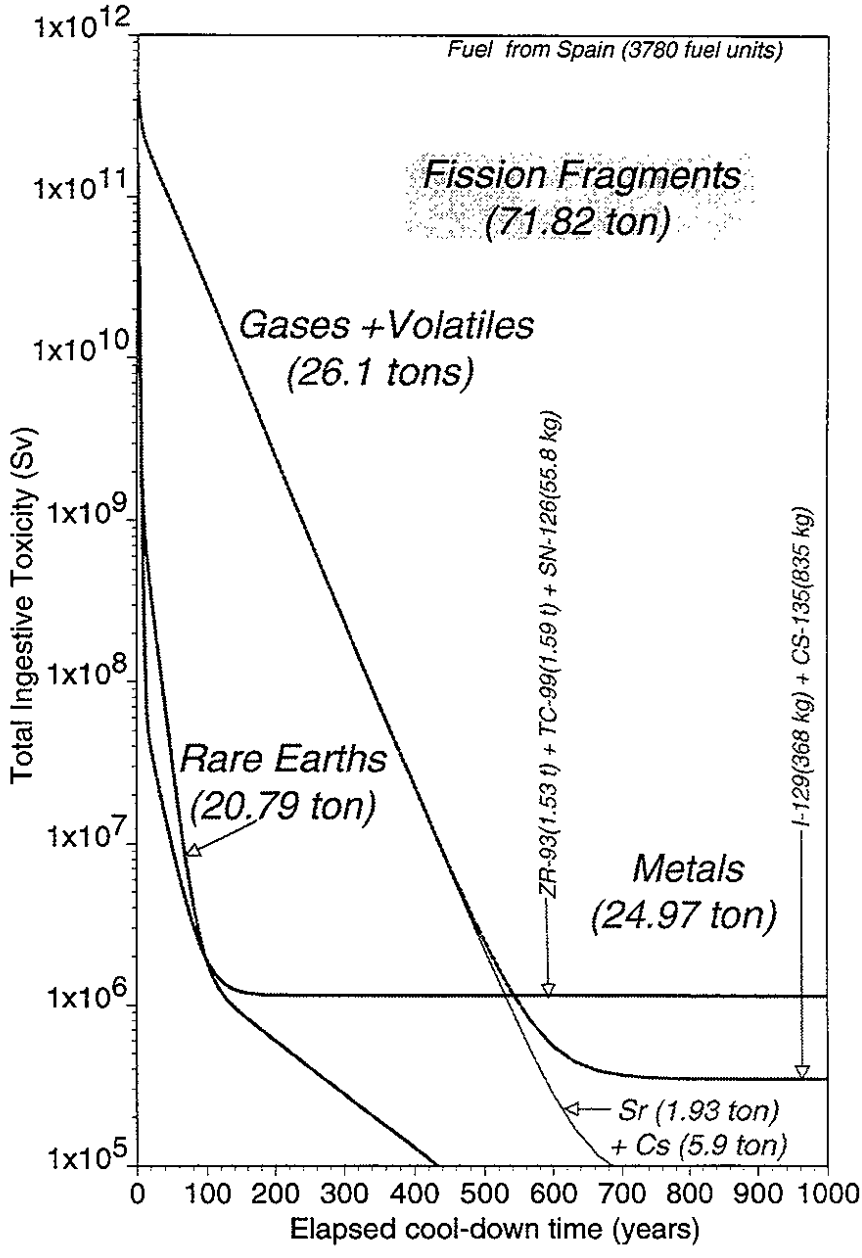

Figure 3. Ingestive radiotoxicity of the Fission Fragments of present Waste from Spain. For projected radiotoxicity at the end of the LWR's commercial life, multiply values by a factor 3.81.

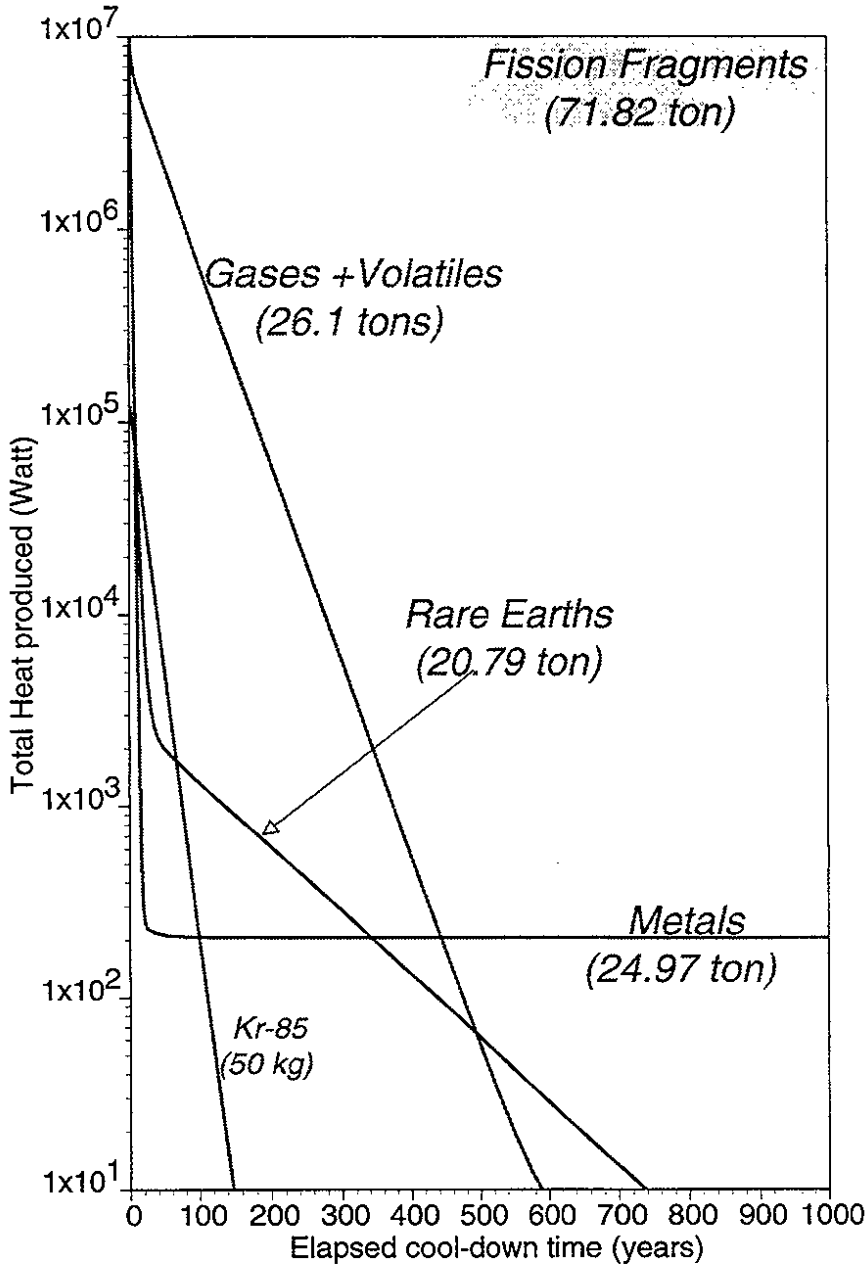

Figure 4. *Same as Figure 3, except for produced decay heat.*

In the latter part of the lifetime of the LWR's once the incineration procedure has been started, this time could be actually reduced. We have assumed for our first order considerations an "average" dwelling time of the spent fuel in the pools in vicinity of the LWR's of 15 years.

(2) After this nominal holding time, the LWR spent fuel will be processed at a constant rate by the EA's. As described later on, the

reprocessing rate for a single EA is about 1 ton of fuel every week. For the minimal number (5) of EA's needed to eliminate the Spanish waste, we have assumed a duration of the whole operation roughly equal to the active life of the LWR's, namely 40 years, corresponding to a cluster of 5 EA's. Actually it is expected that, at least on a purely economical point of view, the activity of the EA site will continue beyond that, since the estimated life of the EA is largely in excess of 60 years. It would make financially good sense to continue running the EA without LWR's spent fuel injections, namely with natural Thorium fuel, of which the small fresh amount of less than 1 ton/year is needed. In this way the TRU's recirculated in the EA could be thoroughly liberated of the residual, exponentially decreasing, LWR waste. Adding decommissioning[1] etc. it is reasonable to assume that the EA site will remain active for about 100 years.

(3) After that time, it would make sense to keep radioactive materials in a dedicated Cool-down facility (Secular Repository), for instance on the site, prior to definitive disposal. This procedure is essentially relevant for Strontium and Cesium and eventually Krypton. Other materials, like Noble Metals etc. could be sent to the final disposal much earlier, essentially at the end of the reprocessing phase or at any rate before the end of activity of the EA. The activity and heat produced in such Cool-down facility at any time will be substantially less than the values of Figures 3 and 4, since the FF's are produced and processed at uniform rates (see Paragraph 3.3). The peak power, occurring at the end of the active incineration period will be of the order of 2.8 MWatt for each EA, decaying after that roughly by a factor 10 every 100 years.

(4) At this point and in the absence of an appreciable quantity of α-emitters, it would make sense to handle all the residuals as a low level waste (LLW) with a permanent disposal on surface or shallow depth[2]. As already pointed out a considerable reduction in the diluting volume can be achieved extracting some of the most

[1]The decommissioning of the EA itself is outside the purpose of this paper and will be discussed later on in another Report.

[2]More specifically, we shall follow all norms of US Nuclear Regulatory Commission [8].

troublesome elements (^{99}Tc amongst the metals and ^{129}I amongst the volatiles) and to introduce them as well in the EA for incineration. This possibility will be discussed in more detail later on (Paragraph 5.3). As we shall see (Paragraph 3.3) most elements, with the exception of Strontium and Cesium satisfy from the beginning easily to all conditions relevant to Class A. Immediate storage of Cesium and Strontium as Class A would require an excessive volume and it is preferable, in view of their relative short lifetime, either (i) to keep them in the Cool-Down facility (see point (3) above) until they reach Class A or (ii) to dispose them with an initial Class C condition, thus saving a few hundred years in the Cool-down facility needed to reach a Class A storage. The main operational difference between a Class C and a Class A repository is that no surveillance is required for the latter in order to prevent intrusion.

To conclude, we shall follow an illustrative scenario in which we assume:

(1) 15 years dwell time of the LWR waste at the Reactor site;
(2) 40 years to reprocess all the waste at a constant rate followed by another 40 years of operation of the EA as an energy producer;
(3) elimination (90 % efficiency) of the most troublesome long lived radio nuclides ^{99}Tc and ^{129}I, by parasitic transmutation in the EA;
(4) continued (modest) surveillance of a (initially few MWatt) Cool-down facility for few hundred years;
(5) ultimate storage of all residuals as low level waste (LLW) for disposal on surface or shallow depth, ultimately as Class A, for which no surveillance for accidental intrusions is required.

3.2.- Pyro-processing of LWR spent Fuel. In order to proceed to the incineration of the TRU's the initial LWR waste must be separated into several streams, namely (1) the TRU's which are destined to the EA, (2) the unburned metallic Uranium which has to be purified to a level such as to be reused again, (3) the Fission fragments (FF) which are to be destined to cool-down and to selective incineration and (4) the activated materials, mostly the fuel Zircaloy clad which are to be left to cool down. At present, most of the fuel reprocessing is performed in centralised facilities, like La Hague, Sellafield

etc. and it is based on aqueous separation methods (PUREX etc.). In this paper an alternative, simpler pyro-processing method has been selected in order to visualise our procedure, since it holds significant promises over the conventional methods:

(1) The separation method is able to extract simultaneously and efficiently all TRU's elements.
(2) It is simple and compact enough to be performed directly at the incineration plant, as one step of the integral transformation process.
(3) It involves small amounts of fuel at each cycle, thus removing the risks of accidental criticality accidents.
(4) It produces no appreciable radioactive chemical waste and otherwise no emission of radioactive materials to be discharged in the environment, thus ensuring the complete closure of the fuel cycle
(5) It holds promises of a cheaper economics.
(6) It permits an integrated fuel reprocessing with the EA operation.

With these guidelines in mind, the LWR waste transformation prior to incineration in the EA should follow the flow-chart of Figure 5, which will be briefly discussed. It is assumed that there is a local plant for each EA unit of the nominal thermal power of 1500 MWatt [1], corresponding to an incinerating capacity for TRU's of 400 kg/year (see Table 11). As a result of this incineration the EA will produce a net of 625 MWatt of electric power, once the needs of the accelerator and of the plant have been subtracted. The yearly processing rate of a single EA will then be of 78.446 PWR Fuel Elements from Spain, corresponding to 52.4 tons of spent fuel. The complete incineration of the 3780 LWR elements presently in Spain will therefore need 48.18 (EA-units) × year and the ultimately expected stockpile 183.59 (EA-units) × year, namely EA 5 units during 36.7 years.

The first step is the separation of the Fuel Oxide from the rest of the activated material, which will be performed according to classic methods[1]. Fuel pins are removed from the subassembly and transferred to an Argon cell for the tube stripping operation. The actual purification process may begin after the fuel has been separated from its can and chipped in pieces of small size.

[1] ANL is investigating electrolytic decladding as well, using Zircaloy hulls as anode.

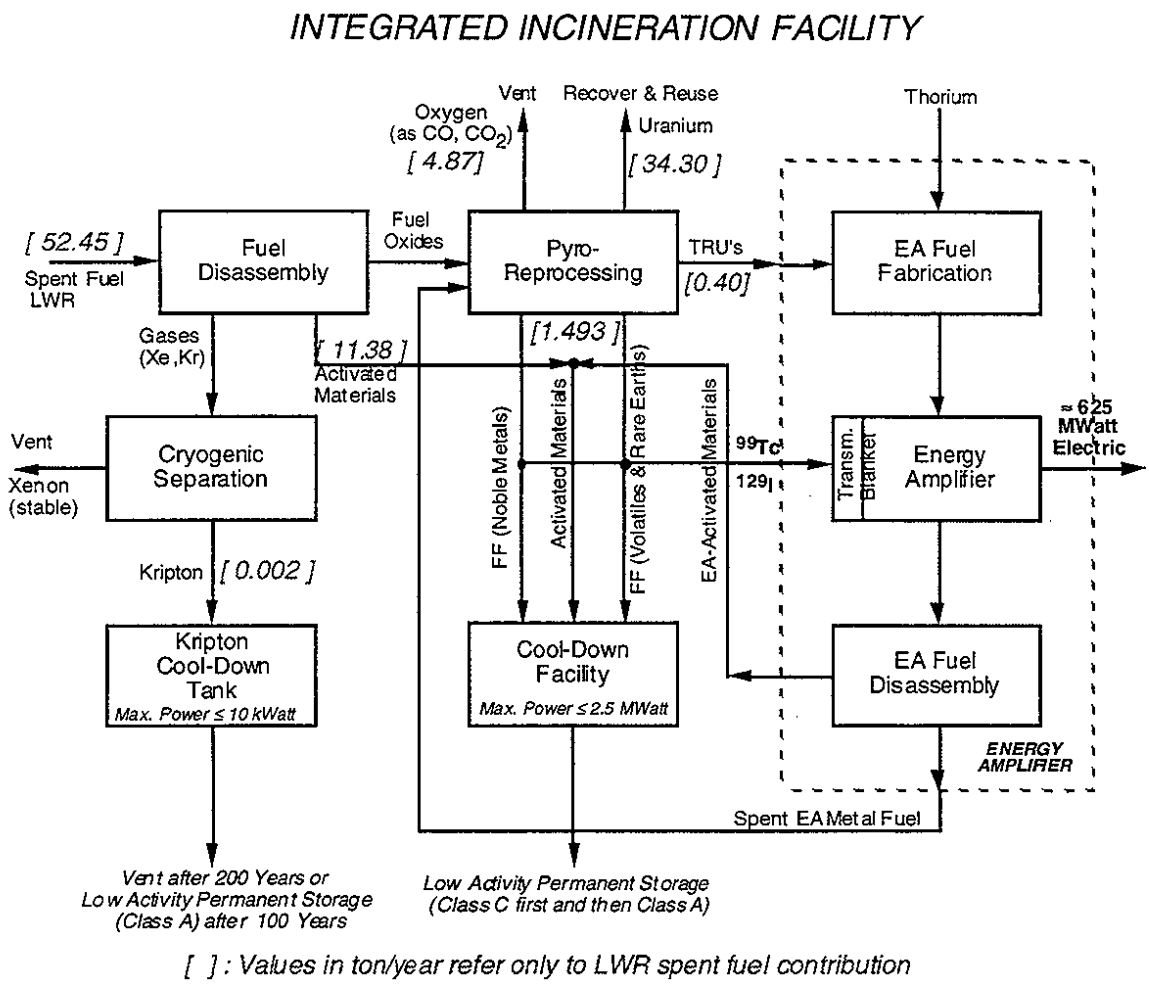

Figure 5. Flow diagram of the LWR waste fuel preparation for the EA incineration. The Pyro-processing Facility is also used for the recycled fuel from the EA. Quantities within square brackets represent the yearly material flow in tons.

During this process a major fraction of the FF gases are liberated, mostly Xenon and Krypton. While Xenon has no appreciable activity, one of the Krypton isotopes, ^{85}Kr is active with a half-life of 10.7 years. Its activity is substantial, amounting to about 1.5×10^5 Cie/year of incineration (15 years of LWR fuel cool-down).

The accumulated radioactivity after 40 years of incineration operation (see Figure 8) is 2.21×10^6 Cie, rising less than linearly due to the intervening decays. The current practice consists in venting all this gas in the atmosphere. It is instead suggested to cryogenically separate the Noble Gases and to store the Krypton in sealed containers until it has decayed, following a well documented procedure [20]. The yield of Krypton (all isotopes) is about 2 kg/year, corresponding to a modest volume at n.p.t. After 100 years from the

start of the EA, activity has decayed to about 44'000 Cie and after 200 years the residual activity is only 80 Cie, at which point it can be vented away safely. If instead we wish to store the gas as Class A after 100 years, it must be packaged in (small) bottles, each containing no more than 100 Cie, leading to a total of 440 bottles, which is reasonable.

The total mass of fuel subassemblies and of pin cladding to be destined to the cool-down repository is 11.38 ton/year, leading to some 39.2 ton/year ($\approx$ 108 kg/day) of fuel oxides which have to be pyro-processed.

The pyro-processing chosen as an exemplificative example is the so-called Lithium Process proposed by the Argonne National Laboratory [6]. A laboratory experiment at the level of 20 kg/batch has been performed and an engineering pilot plant with 200 kg/batch is under construction. The material balance flow sheet of this process, taken from Ref. [6] is given in Figure 6. In view of the fast processing offered by the method, a plant of the size of this first engineering prototype is largely sufficient for a 1500 MWatt EA unit. As the outcome of such a process the oxygen of the fuel (4.87 ton/year), once extracted with the help of Lithium, is vented in the atmosphere as a CO or CO_2 gas formed at the electrodes of the electro-purification of the salt. Evidently the corresponding amount of Carbon, in the form of Graphite must be supplied. For the rest of the materials, at the end of the process, the following streams develop:

(1) The pure Uranium stream, which after Electro-refining at 500 °C and Uranium Consolidation at 1200 °C is essentially pure metal. The isotopic composition of this metal will be : 98.34% of ^{238}U, 1.1% of ^{235}U, 0.544% of ^{236}U and 0.021% of ^{234}U. The essential new isotope with respect to Natural Uranium is ^{236}U, which is however very long-lived (2.34×10^7 years) and therefore constitutes no problem to the further utilisation of the metal, for instance enrichment.

(2) The transuranic elements (TRU) which are also an essentially pure stream, with the exception of a significant amount of Uranium, which is about 3×10^{-3} of the initially processed Uranium. As a result about 100 kg/year of Uranium, mostly ^{238}U will end-up in the 400 kg/year of TRU's yearly. This addition will not appreciably affect the operation of the EA, since it is diluted in the $\approx$ 10 ton mass of the fuel and since the behaviour of the fertile Uranium is easily trimmed with the Thorium concentration. However this Uranium will mix with the ^{233}U bred during the cycle, which is about 170 kg/year. As a result

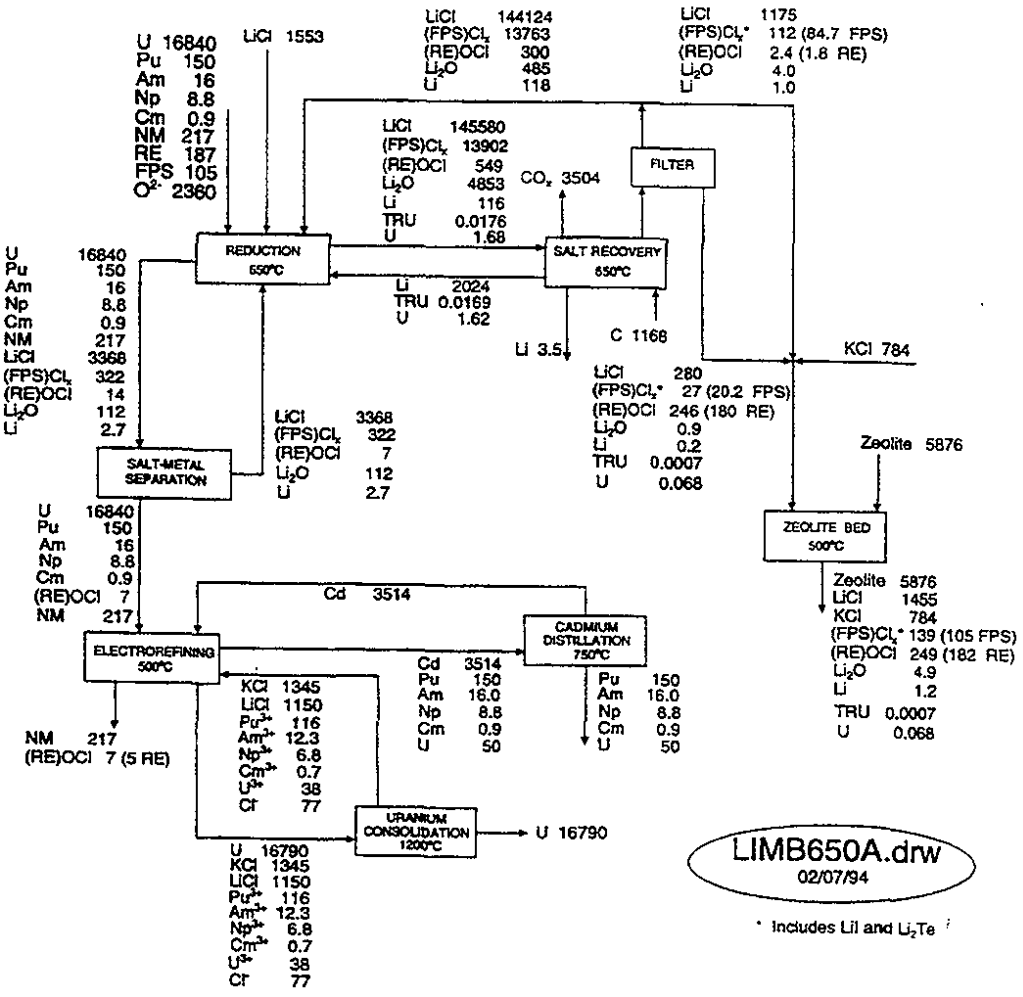

Figure 6. Lithium process material balance flow sheet (basis: 20 kg original UO_2 LWR fuel; all masses are in grams; LiCl reductant salt at 650 °C). From Ref. [6].

the produced Uranium will be "denatured" to an isotopic mixture with 63% of fissile ^{233}U, which excludes military diversions[1].

(3) Fission Fragments, which are produced in two streams, one as result of the electro-refining, which will contain all Noble Metals[2] in metallic form and another as the result of the salt recovery process which will contain all the rest, in the form of chlorides or oxi-chlorides. The essentially pure noble metals could be for instance

[1] Note that the likely fate of the bred Uranium is new fuel for LWR and therefore it will be anyway largely diluted in ^{238}U.

[2] Some confirmatory work may be needed in the case of Technetium.

blended in a Copper matrix and sent to the Cool-down Facility. As one can see from Figures 3 and 4, the amount of heat produced and the radiotoxicity are not very large and decay relatively quickly. The rest of the FF's are dominated at short times by the Strontium and Cesium heat and radiotoxicity. The stream from the salt recovery also contains tiny amounts of Uranium and of TRU's. Taking at face value the flow chart of Figure 5 these amounts are extremely small, namely 138 gr/year of Uranium and 1.4 gr/year of TRU's. However one should wait for the results of the larger scale facility in order to see if this remarkable performance can be maintained.

It would be obviously attractive to make use of the same reprocessing facility for the recirculated fuel from the EA as well. The fuel volume is much smaller (≈ 5 ton/year) but the TRU concentration is much higher (10 ÷ 15 %). Smaller unit loads should be therefore used. The fuel is already in metallic form and therefore the reduction process is not necessary. The Uranium stream will contain the bred ^{233}U isotope (≈ 170 kg/year), as already pointed out mixed with some ^{238}U and intended to be used as fissile material in LWR's in substitution for enriched Uranium. It should be therefore appropriately diluted before being transformed for instance in Oxide fuel. Fission fragments (≈ 1.5 ton/y) though slightly different in relative concentrations [1] and activated materials, mostly stainless steel can without problem be sent to the Cool-down Facility and or to the LLW disposal.

3.3.- The Cool-down Facility. The recovered FF's and activated materials must be stored somewhere in order to let radio-toxicity and heat decay to a level acceptable for the final LLW disposal. We have assumed that such a storage is to be located at least initially on site. The cool-down facility will be progressively filled with radionuclides during the active period of the plant. After that, the natural decay processes will gradually reduce the activity.

We show in Figure 7 the total produced heat in the case of a single 1500 MWatt EA steadily operated during 40 years, corresponding to 2098 tons of processed PWR fuel. By then, the total FF accumulated mass is 59.71 tons. A delay of 15 years has been assumed between the LWR discharge and the reprocessing and subsequent storage in the Cool-down facility. The

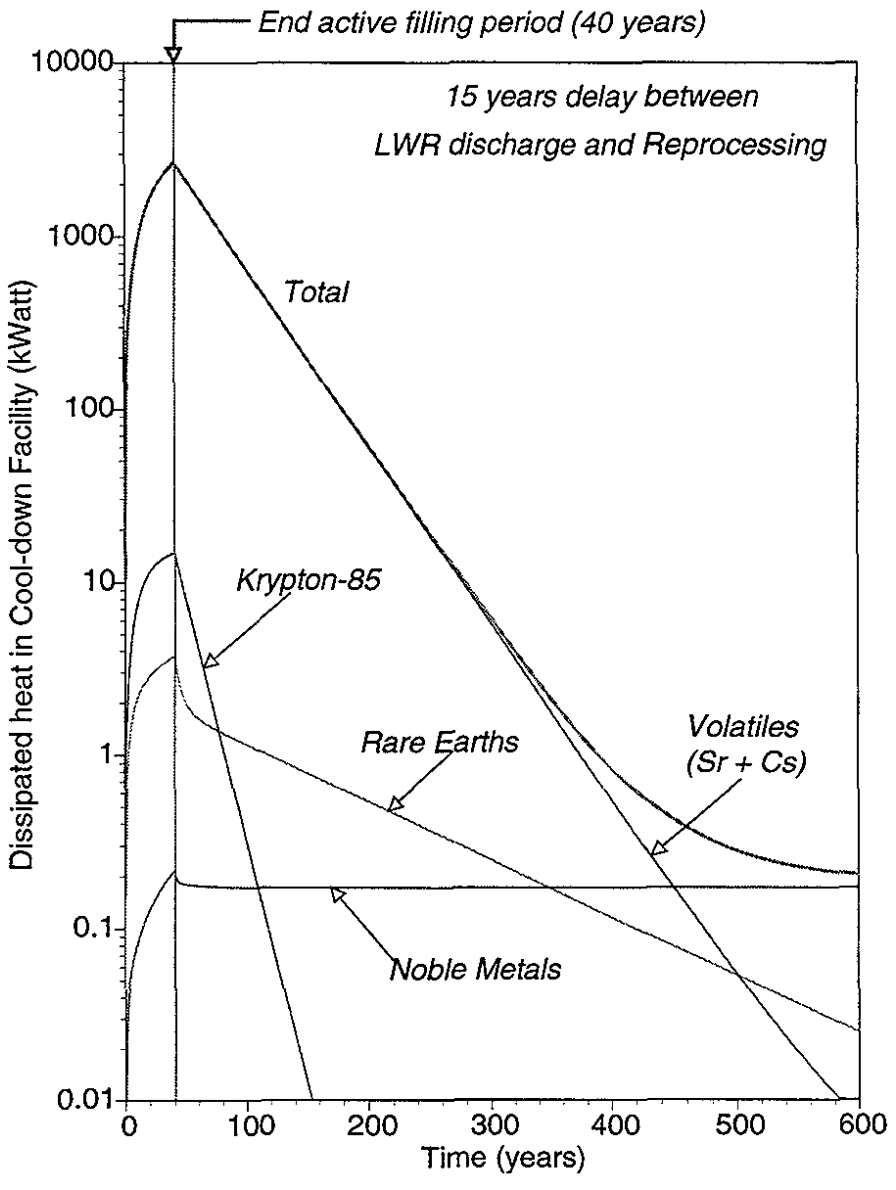

Figure 7. Decay heat produced in the 59.71 tons of FF's fragments as a function of time. The active period of the plant is set to 40 years, during which 2098 tons of spent fuel from LWR's are processed.

maximum power to be dissipated due to all FF's at the end of the active period is about 2.76 MWatt, corresponding to 46.2 Watt/kg.

We have also subdivided the heat contributions of (1) ^{85}Kr gas, amounting to a maximum peak power of a mere 14.7 kWatt; (2) the dominant Strontium and Cesium contribution amongst the volatile materials; (3) the small contribution due to the Rare Earths, 3.7 kWatt max. and finally (4) the Noble Metals which have an almost time independent level of ≈ 170 Watt dominated by the contributions of ^{99}Tc (72.2 Watt), ^{93}Zr (14.43 Watt) and ^{126}Sn (83.18 Watt). These elements are very long lived and dominate the residual small amount of heat at times longer than say 600 years. It is

proposed to separate out the ^{99}Tc for transmutation to stable Ruthenium and to retain the other elements of the Noble metals in a Copper matrix. The activity is sufficiently low to send without additional delay these elements to the LLW disposal (see next Paragraph).

After the incinerating period of the plant is over (40 years), the time decay of the produced heat density, 100 years after start-up, is 10 Watt/kg, falling about a factor 10 after each 100 years (1.02 Watt/kg after 200 years, 0.102 Watt/kg after 300 years and 0.013 Watt/kg after 400 years). The duration of active storage in the Cool-down plant is set by the nature of the following disposal and it is illustrated in the next Paragraph.

The modest amount of power to be dissipated suggests the use of a simple convection driven air-cooled arrangement. The containment of the materials must of course be perfect over the several hundred years of the storage. An interesting solution has been proposed by the ANL group which consists in absorbing the FF's, with the exception of the Noble Metals, in a Zeolite matrix, with a total mass which is approximately ten times the mass of the FF's. The total mass of the Zeolite is therefore of the order of 600 tons and the maximum decay heat to be dissipated in the Cool-Down Facility of the order of 4.6 Watt/kg.

3.4.- The final storage. According to the US Nuclear Regulatory Commission [8], low level waste (LLW) for disposal on surface or shallow depth is defined on the basis of concentration of certain long- and short-lived radionuclides. This classification system include Classes A, B and C with a growing level of activity[1] and it is designed to minimise potential exposures in both the short and long term. Essentially all medical waste are Class A. Industrial wastes are largely Class A, but they contain some Class B and C. The Separation between classes as specified in 10 CFR Part 61.55 is as follows:

(1) Class A requires no segregation of the waste for Disposal. Stability conditions are specified in 10 CFR Part 61.56.

[1] An additional level GTCC (greater than class C) is used mostly for temporary storage. The concentration of actinides and of I-129 determines the lower activity boundary. There is no limit on concentrations of nuclides with half-lives < 5 years and on Co-60.

(2) Class B must meet more rigorous requirements on waste form to ensure stability after disposal (10 CFR Part 61.56).

(3) Class C not only must meet more rigorous requirements on stability after disposal (see Class B) *but also requires additional measures to protect against inadvertent intrusion.*

Conditions on radionuclides to meet such specifications are listed in Table 4.

Table 4. *Concentration limits set by 10 CFR Part 61 on radionuclides for LLW storage[8].*

Radionuclide	Max. Concentration (Cie/m^3)		
	Class A	Class B	Class C
Long lived elements			
C-14	0.80		8.0
C-14 in activated material	8.0		80.0
Ni-59 in activated material	22.0		220.0
Nb-94 in activated material	0.02		0.20
Tc-99	0.3		3.0
I-129	0.008		0.08
α-emitting transuranic with $\tau_{1/2} > 5$ y	10.[1]		100.[1]
Pu-241	350.[1]		3500.[1]
Cm-242	2000.[1]		20000.[1]
Medium-lived elements			
H-3	40.0	no limit	no limit
Co-60	700.0	no limit	no limit
Ni-63	3.5	70.	700.
Ni-63 in activated material	35.0	700.	7000.
Sr-90	0.04	150.	7000.
Cs-137	1.0	44.	4600.
Short (<5 years) lived elements			
Total of all	700	no limit	no limit

[1] Units are nCi/g.

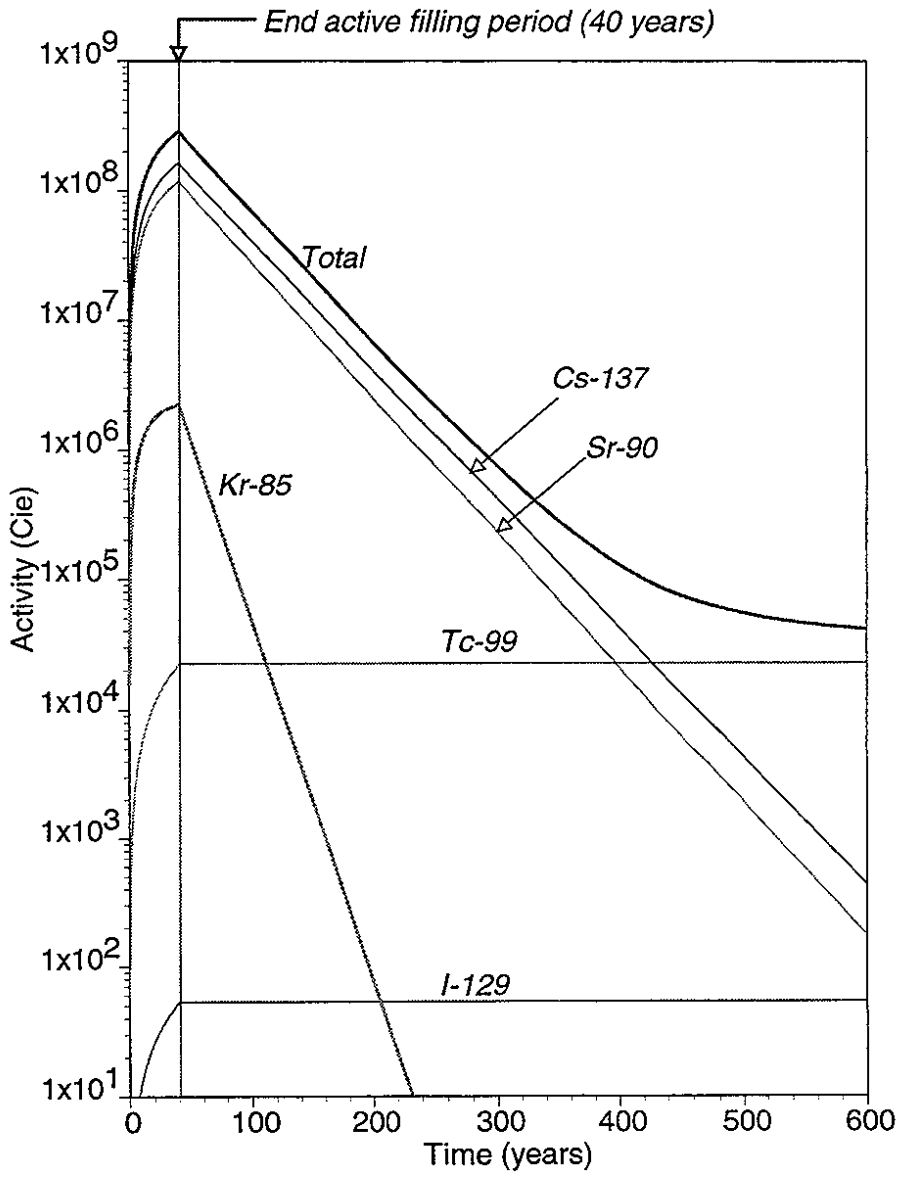

Figure 8. *Total radioactivity in Curie of the 59.71 tons of FF's fragments as a function of time. The active period of the plant is set to 40 years.*

The activity in Curies (Cie) of the FF waste from the spent LWR fuel (2098 tons) over the 40 years of the plant operation is shown in Figure 8. As already pointed out it is dominated at short times by the activities of Strontium and Cesium and at long times by ^{99}Tc and ^{129}I.

We consider first the storage volume requirements at long times. They must be necessarily Class A[1], since surveillance against accidental intrusion on the site cannot be guaranteed for hundred of thousand of years, required

[1] Note that Class B is not defined for the long lived isotopes.

for the very long lived elements to decay. We are helped in that by the relatively small amounts of few long-lived radioisotopes, listed in Table 5. The total dilution volume[1] (85234 m^3) is dominated by the ^{99}Tc (74818 m^3) and ^{129}I (6732 m^3), the rest (3684 m^3) amounting to a mere 4.32 % of the total volume.

Table 5. Class A requirements for LLW storage of long lived radio-nuclides (10 CFR Part 61).

Element	Half-Life (years)	Chemic. Mass (kg)	Isotopic Mass (kg)	Isotop. Purity	Limit for Class A	Total Activity (Cie)	Dilution Volume (m^3)
^{99}Tc	$2.11\ 10^5$	1586.84	1586.84	1.00	0.3	22445.56	74818.52
^{129}I	$1.57\ 10^7$	493.3	367.87	0.75	0.008	53.86	6732.06
^{93}Zr	$1.53\ 10^6$	7547.7	1530.9	0.20	3.5	6379.36	1822.67
^{126}Sn	$1.0\ 10^5$	182.48	55.75	0.31	3.5	3655.06	1044.3
^{135}Cs	$2.3\ 10^6$	5901.14	834.62	0.14	1	796.77	796.77
^{79}Se	$6.5\ 10^4$	119.7	12.43	0.10	3.5	71.71	20.49

Such total storage volume is not extravagant, since it will be accumulated over the 40 years of active operation of the EA at the rate of 2130 m^3/year and it corresponds to 2098 tons of processed PWR fuel, namely to an ultimate packing of 40 m^3 for each ton of spent fuel. For instance Coal Ashes for the same energy produced will involve a much larger volume.

The proposed incineration technique will reduce this figure further to about 4 m^3 for each ton of spent fuel. Such considerable benefit in the volume is achieved by prior incineration of ^{99}Tc and ^{129}I. Assuming a 92 % efficiency, the ultimate, Class A volume drops to about 8200 m^3. This method is described later on in this report. In itself the gain in volume is primarily based on cost considerations which are not determinant and "per se" would not entirely justify the effort of developing transmutation technologies. However, as already pointed out, this additional transmutation may be driven by environmental considerations, beyond what is required by 10 CFR Part 61. Indeed the fate at very long times of Technetium and Iodine are cause of some concern [11], as evidenced for instance by the low limits for Class A.

[1] A cube of about 44 m on each side.

Table 6. Strontium and Cesium related conditions for LLW storage (10 CFR Part 61).

Element	half-life years	Class A (Cie/m^3)	Class B (Cie/m^3)	Class C (Cie/m^3)	B $\rightarrow$ A years	C $\rightarrow$ B years	C $\rightarrow$ A years
Sr-90	29.1	0.04	150	7000	346.3	161.7	508.0
Cs-137	30.1	1	44	4600	164.7	202.4	367.0

Once the total dilution volume has been set for long times, one can examine the fulfilment of conditions for LLW at earlier times, which as pointed out, are determined by our policy on Strontium and Cesium (medium-lived), and the consequent period of their Cool-down, summarised in Table 6. From an initial Class C, the transition to Class A occurs after some 508 years for ^{90}Sr and 367 years for ^{137}Cs. These numbers are reduced to 346 and 164 years respectively for an initial Class B. For an initial Class C, active surveillance will be required until Class B is reached, which corresponds to the first 162 and 202 years respectively. The Class C allows a ten times larger concentration of long-lived isotopes and therefore 90% of this volume can be allocated to the medium lived isotopes in an initial, class C mixture. When the medium-lived isotopes have decayed, the package will become naturally Class A, dominated by the long-lived.

A number of scenarios may be envisaged and they relate primarily to the sharing between the Cool-Down facility and the LLW repository for the medium-lived elements. They will be illustrated with the help of Figure 9. We list the extreme cases, with an obvious continuum in between.

(1) The LLW final disposal is performed only for Class A waste. For a total waste volume of 10'000 m^3 and transmutation, the ultimate transfer from the Cool-Down facility is performed 640 years from the start of the EA plant (point 1 of Figure 9). Without transmutation and a total waste volume of 100'000 m^3 this ultimate transfer is performed about 100 year earlier (point 2 of Figure 9).

(2) The LLW final disposal is performed for an initial Class C waste, to evolve naturally to a Class A waste, according to the timings of point (1) above. For a total waste volume of 10'000 m^3 and transmutation,

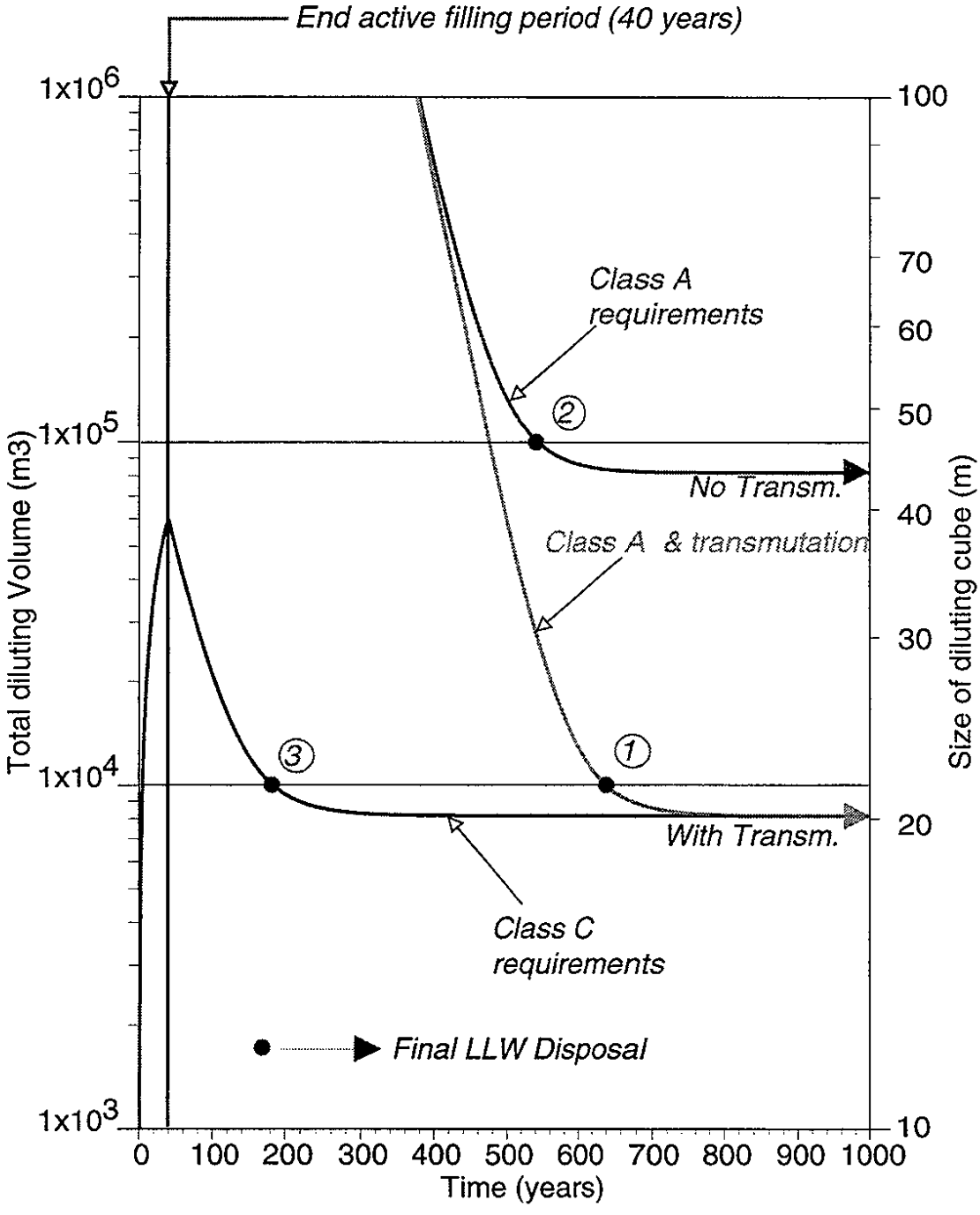

Figure 9. Total diluting volume for LWR Waste and strategy leading to the final permanent Class A LLW Storage. For details see text. The active period is set to 40 years, during which 2098 tons of spent fuel from LWR's are processed. After a cool-down period, the waste is transferred to the LLW Repository. Reduction of the waste volume is achieved with ^{99}Tc and ^{129}I transmutation with about 92% efficiency.

the ultimate transfer from the Cool-Down facility is performed 150 years from the start of the EA plant (point 3 of Figure 9). Without transmutation and a total waste volume of 100'000 m^3 this ultimate transfer is performed any time, since conditions above Class C are

never exceeded during the active period of the EA plant. Note that medium-lived and long-lived are mixed before disposal.

The pyro-processing method has a small leakage of TRU's in the FF stream. The consequences of the α-emitting transuranic elements has been investigated. The total α-activity of the TRU waste, once the correction factors indicated in Table 4 for ^{241}Pu and ^{242}Cm have been introduced, amounts to about 10^7 Cie at the end of the 40 years active period and it decays to about 3×10^6 Cie after 500 years. According to Figure 6 taken from Ref. [6] the leaked fraction onto the FF's is extremely small, of the order of 4×10^{-6}. Taken at face value, this would imply only some 40 Cie and 12 Cie respectively in the FF's. In order to dilute them to the required levels of Class C and Class A of 100 nCie/gr and 10 nCie/gr, we would need a diluting mass of $40/(100 \times 10^{-9}) = 400$ ton (≈ 100 m^3) and $12/(10 \times 10^{-9}) = 1200$ ton (≈ 300 m^3) respectively. It is clear that even a much higher leak rate, f.i. a factor 100 larger, like it is realistically to be assumed, will only slightly affect the estimated diluting volume (+30%). There is no doubt however that our scheme critically depends on the high efficiency in separating the TRU's promised by the pyro-processing technology.

In order to leave room for the leaked actinides as well for the activated waste the total nominal volume of for each EA has been set to 12'000 m^3.

4.—THE EA OPERATED AS A WASTE INCINERATOR

4.1.- General Considerations. The EA has been extensively described in several previous papers [1], to which we refer for details. The possibility of incinerating TRU's has also been already discussed in Ref. [2]. The main virtue of the EA is the fact that the fission probability of TRU's is very large for all elements because of the very hard spectrum of the neutrons, only slightly moderated in the high atomic number medium (Figure 10). As a consequence incineration becomes a very efficient process. The TRU's cannot be introduced in the EA without prior blending with a fertile element. We have chosen Thorium[1], rather than Uranium, since the bred fissionable element produced in this way is ^{233}U rather than additional Plutonium.

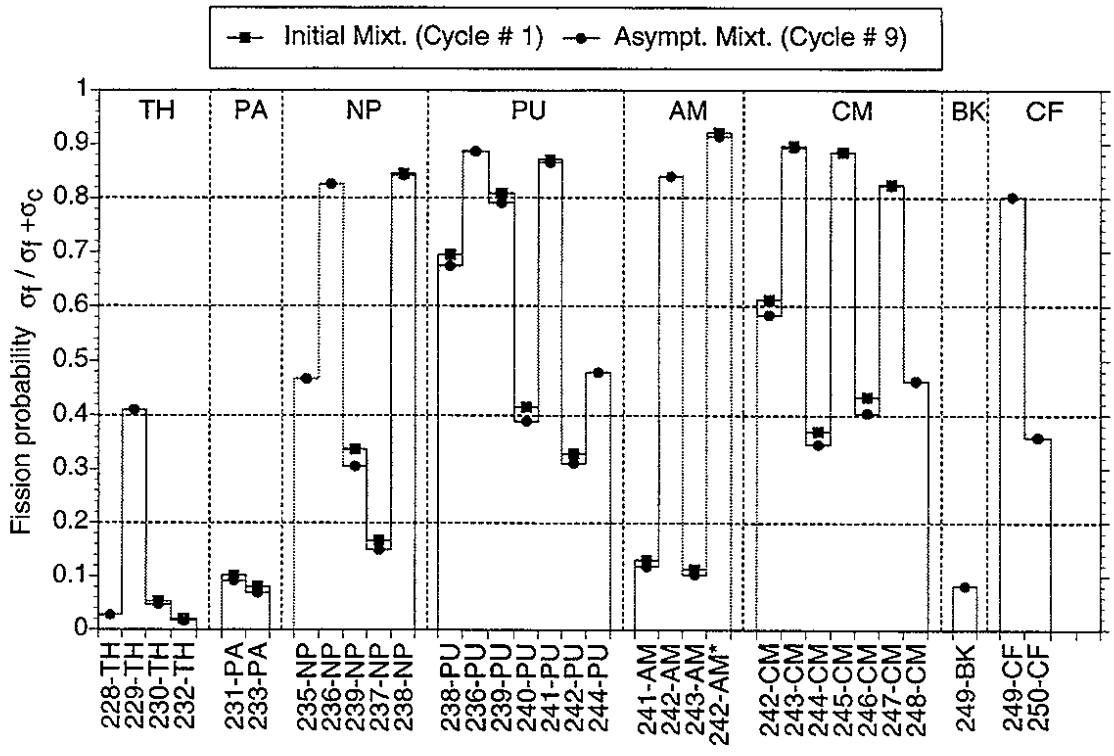

Figure 10. Fission fractional probabilities for the actual spectrum of the EA operated with metal fuel. The differences between the initial fill of TRU's and the asymptotic mixture after many incineration cycles is very small.

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[1] Actually the fuel of the EA will contain a significant amount of ^{238}U coming from the reprocessing stream, though much smaller than the Thorium. The presence of this Uranium isotope will ensure that the ^{233}U thus produced is not "bomb grade". Typically the isotopic content of the Uranium produced will in fact be 60% ^{233}U and 40% ^{238}U.

Table 7. Main parameters of the EA operated as a Waste incinerator.

Gross Thermal Power/unit	1500	MW
Coolant	Liq. Lead	
Sub-criticality factor k, (nominal)	0.97	
Scram systems (CB_4 rods)	3	
Main Vessel		
Gross height	30	m
Diameter	6	m
Material	Steel	
Walls thickness	7.0	cm
Weight (excluding cover plug)	515	tons
Fuel Core (double structure, inner, outer core)		
Fuel mixture (asympt.)	^{232}Th +30% TRU	
Fuel mass (asympt.)	9.2	ton
Number of bundles inner core	30	
Number of bundles outer core	90	
Specific power	160	W/g
Core Power density, aver.	176	W/cm^3
Core Power density, max.	305	W/cm^3
Fuel Pins		
Outer diameter	8.2	mm
Cladding thickness	0.35	mm
Cladding material	HT-9	
Active length	150	cm
Fuel-Cladding gap thickness	0.1	mm
Inner Void diameter	6.26	mm
Sub-Assemblies (Bundles)		
Configuration	Hexagonal	
No hex. rounds of pins (inner, outer core)	10 , 11	
No of pins in each bundle (inner, outer core)	331 ,397	
Flat to Flat	234	mm
Pitch between pins (inner, outer core)	12.45 , 11.38	mm
No units	120	

At the end of the cycle, Uranium is easily separated from the TRU's by the same reprocessing facility used to prepare the LWR fuel, the volume of spent fuel of the EA being much smaller, of the order of 10 tons discharged every two years. The ^{233}U thus produced, of the order of 170 kg/year is either preserved for future operation of the EA without incineration or blended with the recovered Uranium from the LWR in order to produce new fuel for LWR[1].

With respect to the model of Ref. [1], a number of changes (see Table 7) have been introduced in order to optimise the incinerating power and to optimise the parameters to the actual TRU discharge from Spain, which has been assumed the one after 15 years of cool-down, which increases significantly the Americium content. In particular:

(1) *the Fuel is metallic rather than mixed-oxide.* Metallic fuels have been studied in great detail by the IFR project in the US. These fuels were made of cast U—xPu—10Zr alloy with x=0 ÷ 26 and a smear density of 75%. The plenum/fuel ratio was 1.0 ÷ 1.5. The reasons for our choice is primarily related to the need to achieve the hardest spectrum in order to enhance fission and that metal fuel are most convenient for pyro-processing. They however present many additional advantages:

(i) large burn-up. Before the shut-down of EBR-II values of the order of 20% at. were achieved, corresponding to 200 GWatt d/t.

(ii) ease of Fuel refabrication (casting) and reprocessing (pyro-electric);

(iii) high fuel density;

(iv) high fuel thermal conductivity, which ensures low centreline temperatures, with the same T_{max}/T_{melt} as oxides for the same power;

(v) Reactor can operate with breached pins.

The technology of the EBR-II should be easily extrapolated to Thorium based Fuels with more favourable higher temperatures, since

[1] In order to prepare fresh fuel with a reactivity similar to the one used in the Spanish reactors, in addition to the 1.1 % surviving ^{235}U already present one has to add about 2,5 % of ^{233}U. The fresh fuel produced in this way will then be about 10.3 tons, including the mass of the cladding, Oxygen etc. Note that this represents 10.3/52.45= 20% of the spent LWR fuel reprocessed.

(i) alloy redistribution, which limits Fuel centreline melting to occur at higher temperatures. For instance U-20Pu-10Zr is liquid at 1068 °C, while Th-20Pu is liquid at 1450 °C.

(ii)Fuel-cladding metallurgical interactions occur at temperatures which are ≈ 150 °C higher. For instance the minimum temperature of the U-Fe Alloy is 725 °C while the minimum of a Th-Fe Alloy is 875 °C. These values have to be compared with the rapid creep limit of the HT-9 cladding which occurs starting at 650 °C.

(2) *The amount of blending Thorium has been minimised,* to reduce the fraction of the neutrons producing ^{233}U, since it is assumed that the primary purpose is incineration rather than energy production. As a consequence the fuel total mass which was in Ref. [1] of the order of 25 tons has been reduced to about 10 tons. The breeder has also been removed. The main result of these changes are:

(i) a larger fraction of the neutrons escape from the core. These excess neutrons, as discussed later on will be used to eventually incinerate the most offending FF's, namely ^{99}Tc and ^{129}I;

(ii) the cycle time is reduced from 5 years to 2 years for a nominal burn-up of 120 GWatt d/t and

(iii) the power produced in the fuel is of the order of 160 W/g, still quite acceptable for a metallic fuel.

(3) The *fuel pin geometry has been modified* in order to leave space around the core for the ^{99}Tc and ^{129}I incinerators. Therefore the long, empty tubular appendixes (plenum) have been removed. The dedicated space for gases build up is now a wider, central hole in the fuel pin. A larger central hole has also advantages on the maximum temperature of the fuel and the over all design of the fuel cladding is simplified. The additional space and the shorter cycle time reduce considerably the pressure build-up due to FF gases.

4.2.-Thermo-hydraulic considerations. These modifications imply some differences in the thermo-hydraulic which have been extensively checked with a commercial code called Star-CD [21], which implements the finite element method. The current numerical model includes more than 30000 cells and 64000 boundaries and consists of an axial-symmetric representation of the

entire machine including the core structure, the structural materials (vessel, target container, beam window, beam pipe, flow separators), the heat exchangers and the RVACS (Radiation Vessel Air Cooling System). Such a model has been used to study the Lead flow in the vessel and in the target. The geometry and the results of the thermal hydraulic simulation have also been imported in ANSYS [22] to evaluate the thermal stresses in the beam window [23]. Transient simulations have also been used to simulate accidents such as Total Loss of Power [24]. We refer to Ref. [1] for more details of the general layout of the EA.

The primary cooling loop of the EA is substantially similar to what has already been described in Ref. [1] and Ref. [25]. The changes affect mainly the core and the beam target. The total height of the core is still 1.5 m but the upper and lower plenums have been removed, since the function of collecting the fission gases is now taken over by a bigger central void in the metallic fuel pin. Also the breeder has been removed while the Inner Core and Outer Core regions have been left untouched. Since plenums pressure drops are no longer present, the natural flow is enhanced.

Table 8. *Main parameters of the primary cooling system.*

Approximate weight of the coolant	9350	ton
Coolant	Liq. Lead	
Pumping method	Nat. Conv.	
Convection column height	25	m
Convection generated primary pressure	0.54	bar
Heat exchangers	4×375	MW
Decay heat removal	RVACS	
Inlet temperature, Core	400	°C
Outlet temperature, Core	612	°C
Maximum Cladding temperature	656	°C
Maximum heat flux in fuel pin	128	W/cm^2
Coolant Flow in Core	50.3	ton/s
Coolant speed in Core, average	1.382	m/s
Average coolant speed in ascending column	0.764	m/s

In Table 8 we give a list of the main parameters of the primary cooling system. We can observe the following:

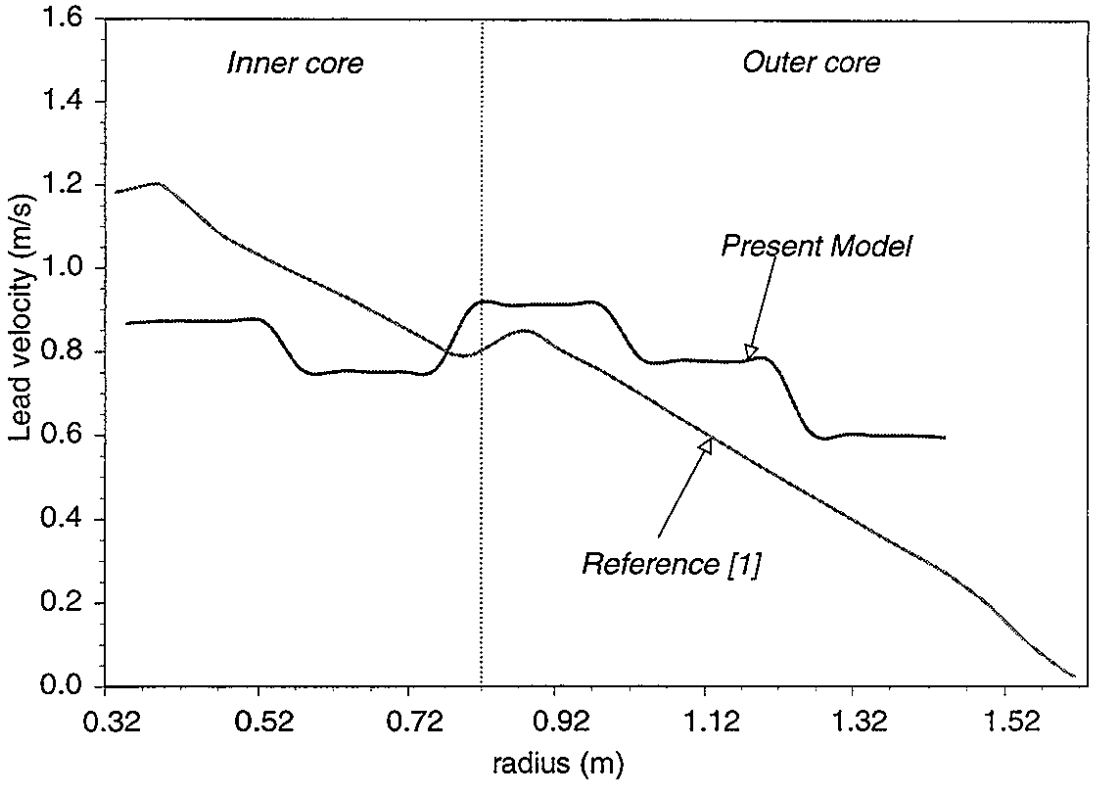

Figure 11. Core outlet Lead speeds in Ref. [1] and in the present model.

(1) Cooling is more effective: the Lead outlet temperature is in fact 612 °C, about 38 °C less than in Ref. [1].
(2) In view of the lower outlet temperature, the convection generated primary pressure is reduced to 0.54 bar.
(3) The heat flux in the fuel pins and the maximum cladding temperature have decreased, compared to the values of Ref. [1].

A comparison between the core outlet Lead velocities in Ref. [1] and the one corresponding to the present model is shown in Figure 11. Lead speeds across the core are more uniform. The modulation is due to the additional pressure losses introduced at the core inlet grid of each bundle ring to uniformise the core outlet temperatures.

The beam target is enclosed in a vertical cylindrical container of 32.54 cm external radius (in order to fit in the buffer region of the core), about 26 m height (the size of the machine) and 6 mm thickness, made of HT-9. The beam pipe is in the centre of the container and is an HT-9 tube of about 26 m length and 20 cm external diameter. Around the beam pipe there is an insulating flow guide of 34 cm internal diameter, separating the hot Lead flow channel

from the cold one inducing a "chimney effect", that strongly enhances the natural circulation. The flow guide is a sandwich of a 3 mm insulating material between two 3 mm HT-9 concentric layers. In Table 9 we give a list of the main thermal hydraulics parameters of the beam target.

Table 9. Main parameters of the beam target.

Beam kinetic energy	1.5	GeV
Nominal beam current	12	mA
Maximum beam current	18	mA
Beam radius at spallation target	7.5	cm
Beam pipe material	HT-9	
Beam pipe shape	cylindrical	
Beam pipe length	≈ 30	m
Beam pipe external diameter	20	cm
Beam pipe thickness	3	mm
Spallation target and cooling material	Liq. Lead	
Pumping method	Nat. Conv.	
Separated spallation source	yes	
Container material	HT-9	
Container thickness	3	mm
Container radius	32	cm
Beam window material (hemispherical shape)	W-Re (26)	
Beam window thickness (edge, centre)	3.0, 1.5	mm
Maximum heat flux in window (@ 12 mA)	730	W/cm^2
Maximum temperature in window (@ 12 mA)	909	°C
Heat deposition in the window (@ 12 mA)	90	kW
Lead maximum coolant speed (@ 12 mA)	3	m/s
Lead coolant mass flow (@ 12 mA)	588	kg/s
Heat deposition in the Lead (@ 12 mA)	1.02	MW
Maximum Lead temperature (@ 12 mA)	714	°C
Average outlet Lead temperature (@ 12 mA)	545	°C

4.3.-The Physics of the Incineration Process. The incineration process as described in Ref. [2] consists in the gradual elimination of the unwanted TRU's waste with the help of fission, which as shown in Figure 10 occurs abundantly for all elements of relevance. The details of the incineration process are shown in Figure 12 where only transitions due to (1) decays with half-life relevant to the incineration process (2) neutron captures (n,γ) and fissions have been considered for simplicity.

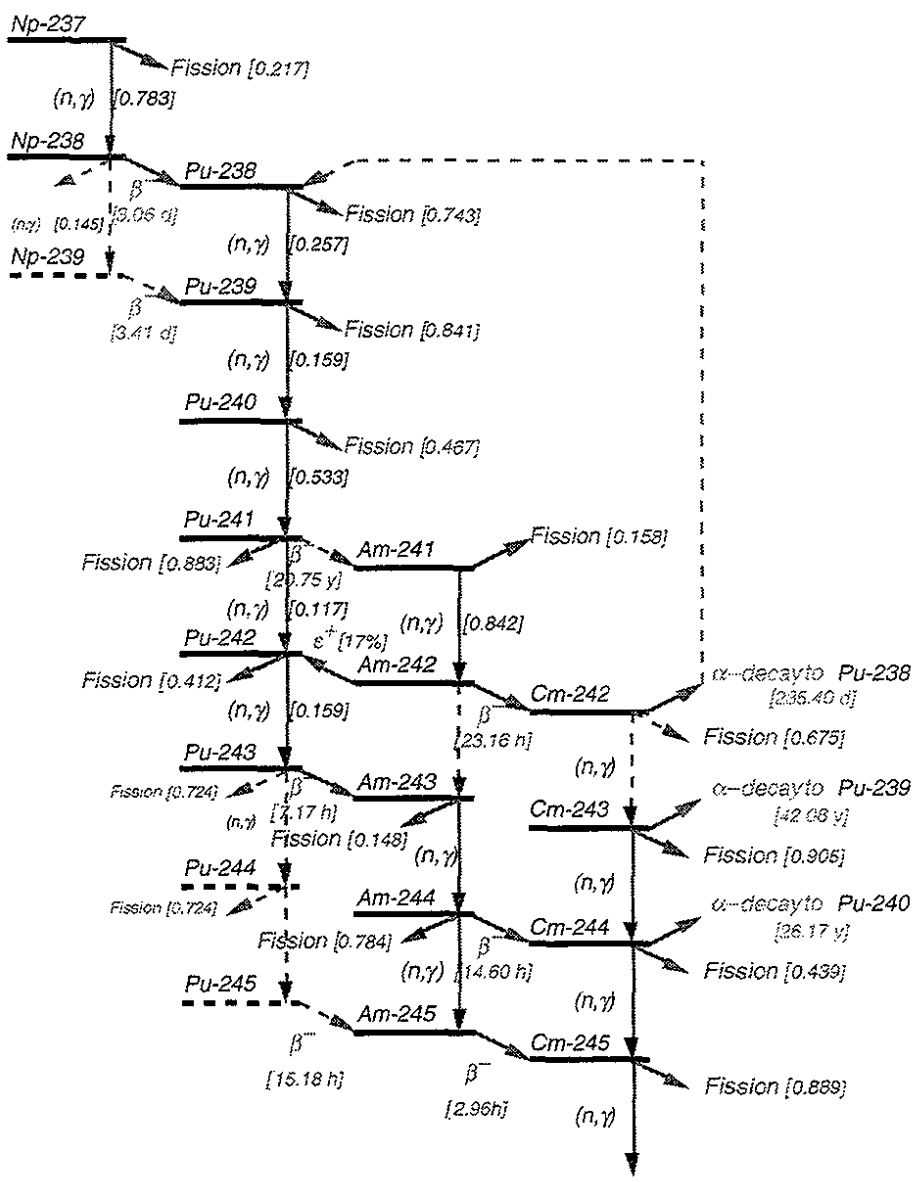

Figure 12. *Main chain of elements involved in the incineration process. Values within square bracket are branching ratio for fission and capture respectively for the actual spectrum of the EA. Only decays with relevant half-life are shown.*

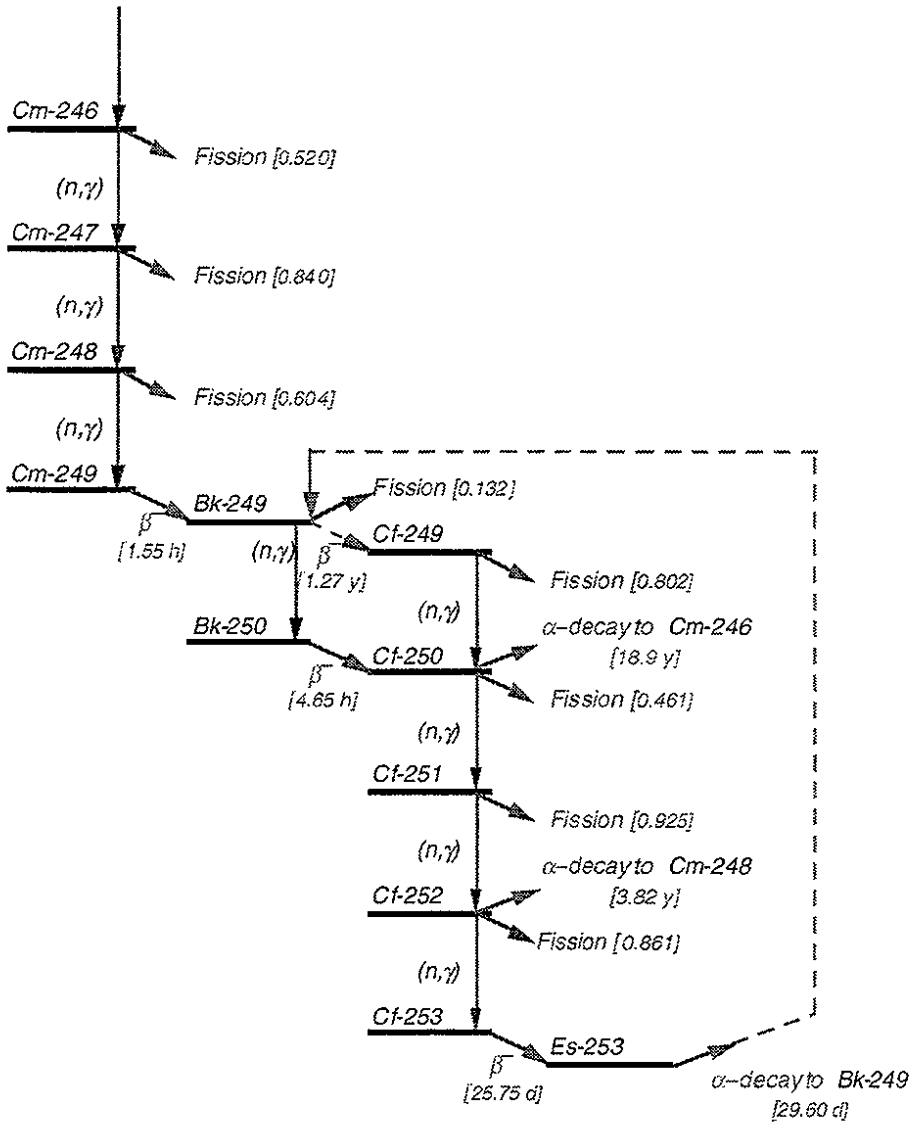

Figure 13. Continuation of Figure 12, for higher elements. Values within square bracket are branching ratio for fission and capture respectively for the actual spectrum of the EA. Only decays with relevant half-life are shown.

Under irradiation of neutrons, an equilibrium situation, reminiscent of the radioactive family decay sets in. Values within square bracket represent the neutron induced branching ratio between fissions and captures and are calculated for the actual energy spectrum of the EA[1]. We remark that the neutron flux φ in the EA operated as incinerator is quite large, of the order of $\varphi \approx 10^{16}$ n/cm^2/sec. Typical transmutation cross sections are of the order of $\sigma \approx 2$ barn, leading to a trasmutation rate $1/\tau = (1/n)\ dn/dt = \sigma\ \varphi = 2 \times 10^{-24} \times 10^{16} = 2 \times 10^{-8}$ sec^{-1}, namely $\tau = 1.58$ years.

[1] As one can see from Figure 10, the changes in spectrum during the composition changes due to the evolution are very small.

The progress through the chain, starting from an initial concentration spectrum occurs with a progressive reduction in the number of surviving atoms, since at each step a substantial fraction is subtracted because of fission. In particular all Plutonium isotopes are strongly fissionable. There are some radio nuclides, like ^{237}Np, ^{241}Am and ^{243}Am which have a fission probability of the order of 10 ÷ 20 % and for which an additional neutron capture is required in order to reach a well fissionable element. They act in the chain as "breeding" elements, and in order to compensate for their presence, the ^{232}Th blending has to be slightly trimmed to lower value. However, as shown by the exact evolution calculation described later on, their presence can be easily accommodated and the required neutron multiplication in the overall chain is carried by the largely fissionable elements.

The chain of Figure 12 is partially closed by the return path due to α-decay (235 d) of ^{242}Cm to ^{238}Pu. A similar closure occurs further on in the chain, as shown in Figure 13, due to the α-decay (29.6 d) of ^{253}Es to ^{249}Bk. Finally the even higher elements have a rapidly rising probability for spontaneous Fission. Of course the closure is purely academic, since by then the concentration of elements is becoming so small because of the repeated fission probabilities accumulated along the chain, that the amount of atoms reaching the end of the chain is truly negligible. What is of importance instead is that, as confirmed by the precise simulation of the whole process, there is no anomalous build-up of concentration anywhere along the chain.

As already mentioned, Thorium blending is required in order to operate in stable conditions. The reaction chain for an initial ^{232}Th element is shown in Figure 14. Incidentally, this is also the main chain for the EA operated as a pure energy generator, namely with no incineration task. The small amount of transuranic elements produced at the end of this chain are of course following the incineration path of the previous figures.

In view of the large concentration of Thorium in the fuel, also the main (n,2n) reactions have been shown, leading to ^{231}Th production with a fractional probability of 7×10^{-3} of all captures in ^{232}Th. The long-lived ^{231}Pa is thus accumulated by the beta decays of ^{231}Th, which introduces a significant radiotoxicity. The direct fission/incineration of this element is not sufficient (0.135) and it occurs through the production of ^{232}Pa and its fast decay into ^{232}U, which is strongly fissionable (0.937), as also the subsequent elements of the chain of Figure 14. Additional production of ^{232}U occurs also

through the (n,2n) reaction on ^{233}U which is a relatively abundant element in the fuel, though with a branching ratio of only 2.1×10^{-4}.

Of course the exact behaviour of the EA and in particular the change of elementary concentrations during burning is simulated with an elaborated Montecarlo calculation in which both interactions and decay are taken into account. For more details on the computing method we refer to Ref. [26]. The evolution through a number of cycles of the actual incineration of the Spanish LWR fuel has been calculated and the results are discussed in the following section.

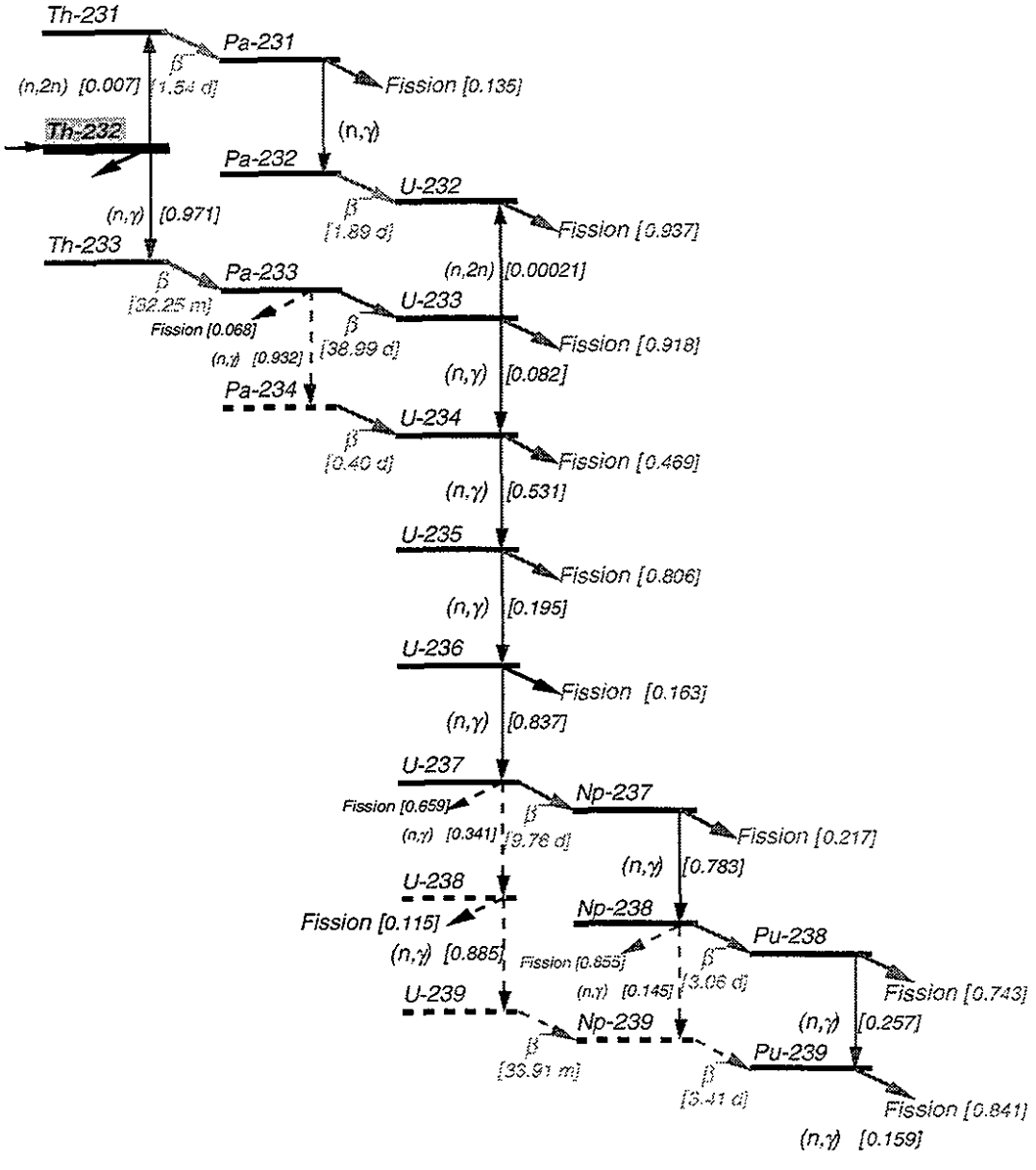

Figure 14. Same as Figure 12 but for the main Thorium breeding process. Values within square bracket are branching ratio for fission and capture respectively for the EA. Only decays with relevant half-life are shown.

4.4.- Fuel composition at each cycle. The fuel composition at each cycle has to be chosen precisely in order to satisfy a number of operational requirements. Indeed in order to ensure a relatively constant multiplication factor k over the long burn-up of each fuel load, one has to ensure that a number of compensating effects are at work:

(1) The decaying reactivity of the TRU's due to their progressive burning must be compensated by an appropriate rise of the concentration of ^{233}U due to breeding on Thorium.

(2) The loss of reactivity due to the emerging captures of the FF's as result of the incineration and the one due to the reduction in the mass of active fuel due to burning must be compensated by an increase of reactivity due to the rising concentration of ^{233}U.

(3) The quantity of freshly added TRU's must be tailored to the residual reactivity of the TRU's from the previous cycle.

An optimisation programme has been written and used in order to simulate the operation of the EA as incineration over a large number of successive cycles. At each cycle, all the actinides surviving from the previous load — with the exception of the Uranium isotopes which are separated out for further use — are mixed with an amount α of fresh TRU's and an amount β of fresh Thorium. Parameters $[\alpha,\beta]$ are then varied in order to reach the best optimisation of requirements (1) (2) and (3) above and a pre-determined value of the initial k coefficient, equal for all cycles. We note that the total fuel mass

Table 10. *Composition of TRU Waste From Spain.*

Radio-Nuclide	Relative Concentration	Radio-Nuclide	Relative Concentration
^{237}Np	0.05360	^{241}Am	0.07382
^{239}Np	0.0	^{242}Am	0.95464E-04
^{236}Pu	0.0	^{243}Am	0.98244E-02
^{238}Pu	0.01641	^{242}Cm	0.22320E-06
^{239}Pu	0.51499	^{243}Cm	0.33610E-04
^{240}Pu	0.21358	^{244}Cm	0.16900E-02
^{241}Pu	0.06614	^{245}Cm	0.12218E-03
^{242}Pu	0.04968	^{246}Cm	0.13857E-04

is not kept constant in the procedure, though the final result is that it changes only moderately over the cycles. The new concentrations are then inserted in the full Montecarlo simulation which is run until the specified burn-up has been reached. This procedure is repeated for a large number of times. In the first filling of course there is only Thorium and fresh TRU's and the mass of the initial fuel is specified by the predetermined k-value. The composition of the TRU fuel from the Spanish LWR's is listed in Table 10.

The results of these calculations are summarised in a number of graphs. In Figure 15 we show the variation of the multiplication coefficient as a function of the burn-up and for nine different successive cycles.

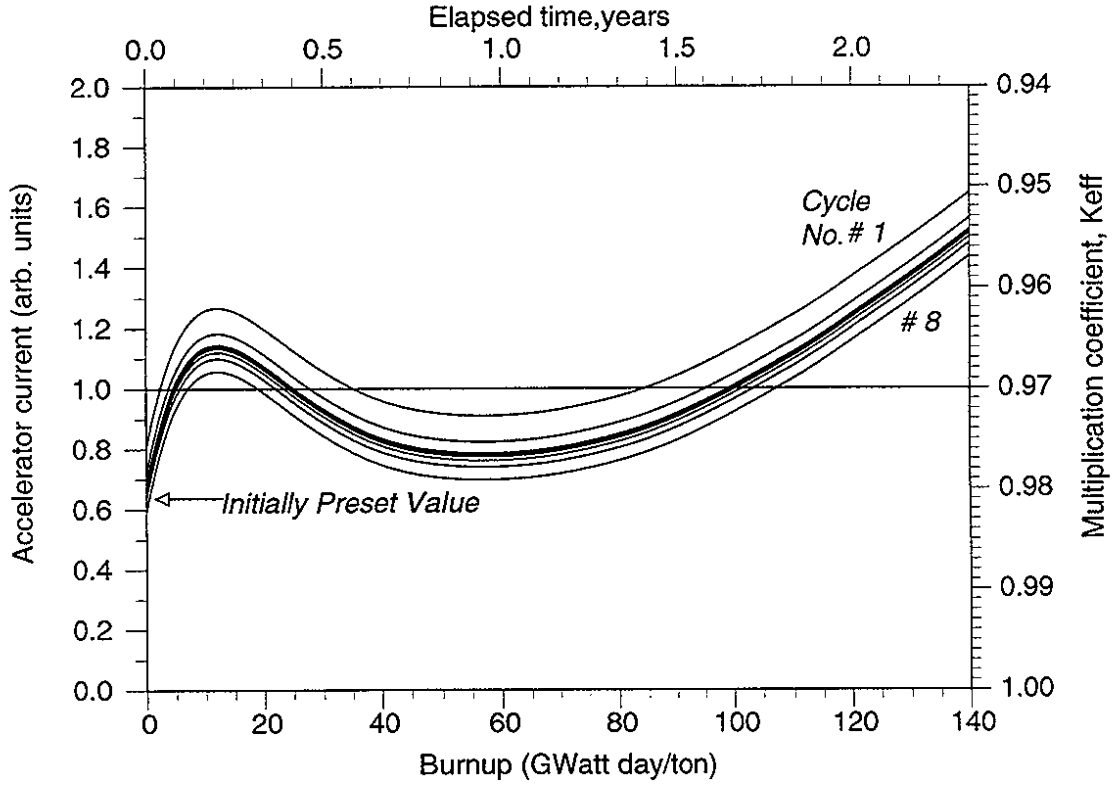

Figure 15. Variation of the required accelerator current and of the corresponding multiplication parameter k as a function of the burn-up for different successive incineration cycles. The initial fuel composition has been optimised at each cycle according to the procedure described in the text.

The optimisation is excellent and it is indeed possible to operate the full EA without external intervention over the specified burn-up of 120 GW d/t, corresponding to a nominal 2 years of operation for each cycle. The power produced is kept constant varying the accelerator current during the cycle in order to compensate for the residual reactivity variations [1]. As one can see, the current must vary from about 0.6 i_o to 1.3 i_o in order to ensure constant

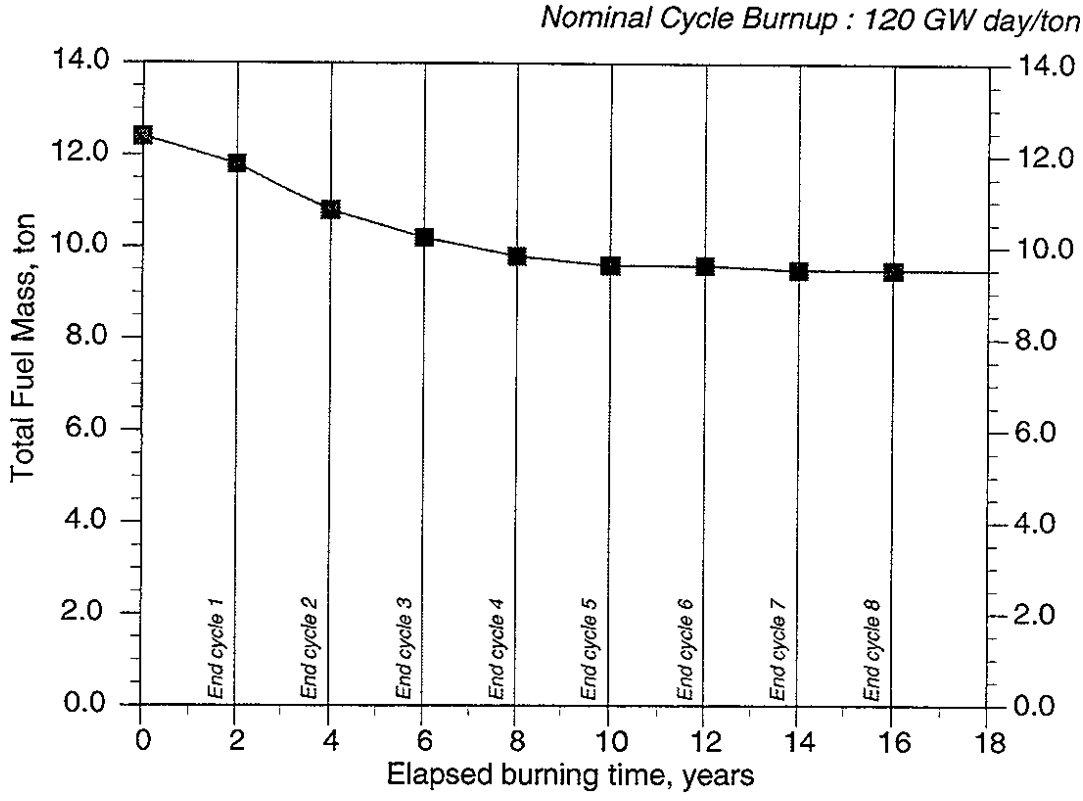

Figure 16. Optimised fuel mass as a function of the cycle.

power output at all times. The k value corresponding to the nominal current i_o has been set to k = 0.97. The energetic gain (heat produced/beam power) is G_o = 83 corresponding to a beam current $i_o = 18/T_p$ mA, where $T_p \geq 1$ is the kinetic energy of the beam in GeV.

The total fuel mass as a function of the cycle is shown in Figure 16. As already pointed out there is a significant drop in the fuel mass after a number of fuel cycles, likely due to the growing presence of the Americium which partially replaces the function of Thorium as a breeder and the fact that indeed less Thorium is needed since less ^{233}U is needed to compensate for the smaller variations of reactivity of the progressively asymptotic TRU's. However after the first ≈ 20% drop in fuel mass due to the cycle initialisation, the fuel mass is tending rapidly to a constant value in the vicinity of 10 tons.

The actual splitting amongst the three main components of the fuel is shown in Figure 17. The residue from the previous load is rising, tending exponentially to a limiting value of about 2.5 tons.

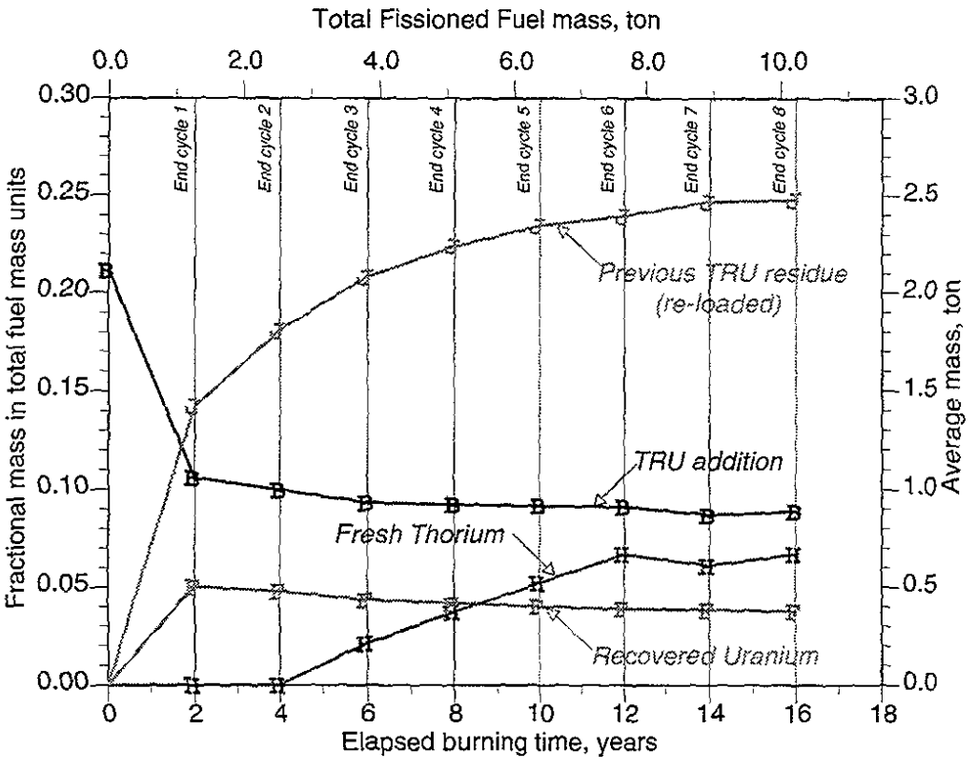

Figure 17. Optimised Fuel composition as a function of the cycle.

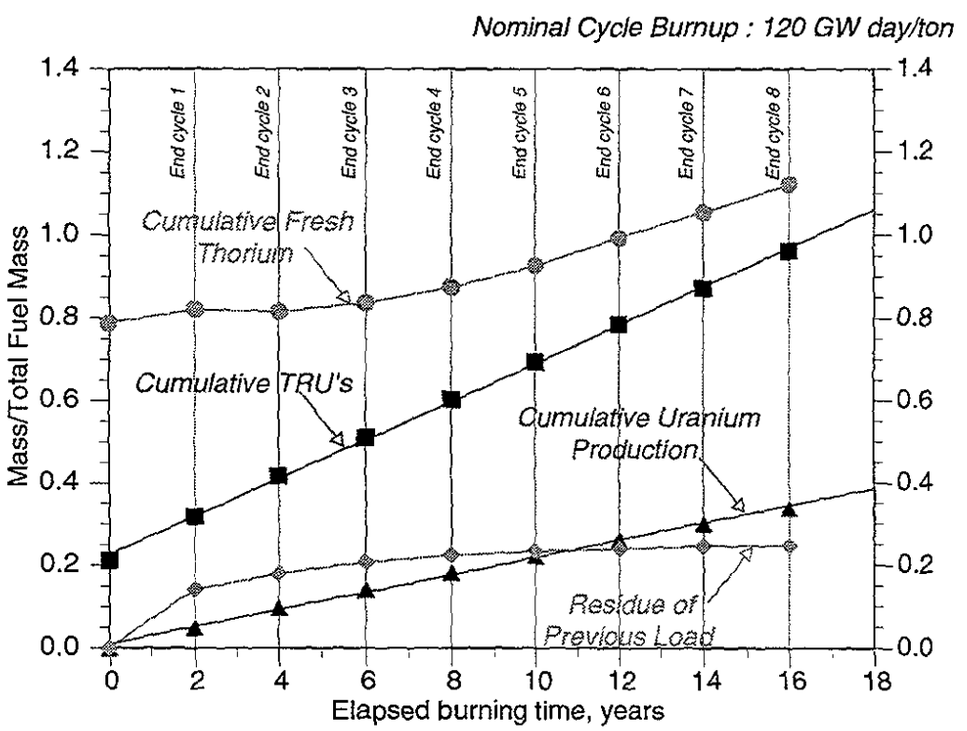

Figure 18. Cumulative, optimised amounts as a function of the cycle.

Table 11. Asymptotic yields of the EA module as Incinerator.

General Parameters		
Nominal Power, thermal	1500	MW
Nominal k value	0.97	
Burn-up/cycle	120	GW × day/t
Fuel Power density	160	Watt/g[1]
Fuel refill period	2.0	EFPY
Overall performance		
TRU incineration rate	402	kg/year
^{233}U-rich Uranium mix production rate	175	kg/year
TRU→ ^{233}U conversion (atom by atom)	0.435	
Refill composition		
Fresh TRU's	0.85	ton
Fresh Thorium	0.74	ton
Residue of previous (actinides ≠ U)		
-Thorium	5.35	ton
-Rest	2.26	ton
Fuel metal total initial mass	9.2	ton
Discharge composition		
Recovered U-mix	0.35	ton
Fraction ^{233}U in U-mix	0.94	
Fission Fragments	1.24	ton

The recovered Uranium is slightly dropping as the consequence of the smaller amount of Thorium in the fuel, also tending to an asymptotic value of slightly less than 350 kg. The addition of fresh TRU's is also essentially constant of the order of 800 kg, after the first large excess required to start up the cycles, which is over 2 tons. The fresh Thorium initially very small also tends to about 700 kg.

The cumulative distribution of these amounts is shown in Figure 18. Note the linear rises in the cumulative amount of TRU's processed and of the

[1] Heavy Metal.

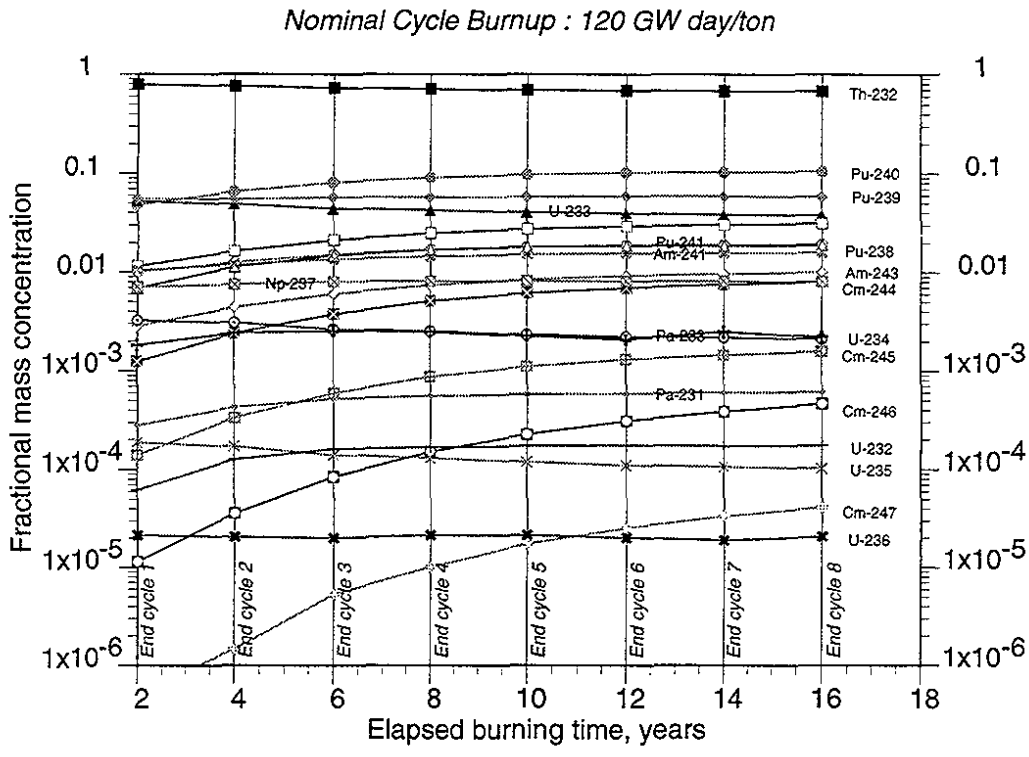

Figure 19. Fractional fuel composition at discharge as a function of the cycle.

Uranium produced. The asymptotic values of the fuel related quantities are summarised in Table 11.

The actual composition of each isotope at load (BOC) and discharge (EOC) is summarised in Table 12. The values of Table 12 for the end of cycle are graphically displayed in Figure 19. We remark the smooth drop in Thorium composition which is required to compensate for the not readily fissionable elements of the accumulating residue. Plutonium's are readily reaching an asymptotic concentration with ^{240}Pu as dominant element, followed by the ^{239}Pu and ^{238}Pu. Also Americium's are quickly stabilised, with ^{241}Am and ^{243}Am at levels of $\approx$ 1% of the total fuel compositions. Curium at the level of $\leq$ 1 % is dominated by the isotope ^{244}Cm which has incidentally a rather short (18.10 year) decay half-life to ^{240}Pu. The more remote ^{245}Cm, ^{246}Cm and ^{247}Cm have relative concentrations dropping by about one order of magnitude at each step. They are not yet completely stabilised at the ninth cycle. The Uranium's should maintain a roughly constant concentration, since they are extracted at every cycle. This is generally so, with the outstanding exception of the ^{232}U which as already pointed out is produced both directly through (n,2n) reactions from ^{233}U and also by neutron captures from ^{231}Pa. It is the growing concentration of this last element, which is part of the recycled waste, which is the reason of the concentration change.

Table 12. Inventory (kg) of EA operation at initial load and after 120 GW day/ton.

	Cycle # 1		Cycle # 2		Cycle # 3		Cycle # 4		Cycle # 5		Cycle # 6		Cycle # 7		Cycle # 8	
Elements	BOC	EOC	BOC	EOC	BOC	EOC	BOC	EOC	BOC	EOC	BOC	EOC	BOC	EOC	BOC	EOC
232-TH	9449.40	8495.30	8663.10	7824.30	7575.70	6907.90	6936.00	6334.30	6510.30	5970.70	6291.20	5782.20	6242.90	5747.40	6146.50	5664.40
231-PA	0.00	3.03	3.12	4.43	4.78	4.95	4.78	4.97	4.78	4.98	4.78	4.97	4.78	5.11	4.78	5.14
233-PA	0.00	19.76	0.00	25.18	0.00	24.01	0.00	22.10	0.00	19.47	0.00	17.69	0.00	21.22	0.00	18.57
232-U	0.00	0.67	0.00	1.30	0.00	1.53	0.00	1.50	0.00	1.49	0.00	1.47	0.00	1.46	0.00	1.46
233-U	0.00	564.31	0.00	504.57	0.00	418.53	0.00	380.02	0.00	349.67	0.00	333.71	0.00	326.27	0.00	318.47
234-U	0.00	34.89	0.00	31.68	0.00	24.95	0.00	22.57	0.00	20.15	0.00	18.79	0.00	18.57	0.00	17.70
235-U	0.00	2.04	0.00	1.76	0.00	1.33	0.00	1.17	0.00	1.02	0.00	0.93	0.00	0.90	0.00	0.86
236-U	0.00	0.23	0.00	0.21	0.00	0.19	0.00	0.19	0.00	0.18	0.00	0.17	0.00	0.16	0.00	0.17
237-U	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
238-U	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
237-NP	138.13	76.73	136.63	77.64	127.09	76.36	119.26	72.43	112.65	70.10	109.65	68.97	108.48	68.95	105.60	67.56
238-NP	0.17	0.00	0.12	0.00	0.11	0.00	0.09	0.00	0.21	0.00	0.11	0.00	0.21	0.00	0.14	0.00
239-NP	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
238-PU	4.42	74.35	82.09	117.06	127.10	140.03	147.77	150.96	156.60	156.11	160.71	157.99	162.40	158.17	163.78	159.14
239-PU	1298.00	599.73	1171.60	565.47	1040.10	544.25	960.23	512.83	905.32	500.96	885.19	498.09	878.06	499.23	857.07	492.15
240-PU	541.32	519.01	751.10	673.40	866.78	765.95	926.99	811.63	957.92	840.16	980.22	860.59	998.51	877.01	1003.70	882.06
241-PU	167.02	109.47	169.21	129.59	176.49	144.54	181.30	151.89	183.52	155.93	185.33	159.21	188.01	162.87	187.98	162.99
242-PU	125.88	124.33	177.97	168.66	214.33	201.01	238.76	222.16	255.56	237.11	268.39	248.73	279.18	258.43	285.88	264.42

Table 12. Inventory (kg) of EA operation at initial load and after 120 GW day/ton (cont.).

	Cycle # 1		Cycle # 2		Cycle # 3		Cycle # 4		Cycle # 5		Cycle # 6		Cycle # 7		Cycle # 8	
Elements	BOC	EOC	BOC	EOC	BOC	EOC	BOC	EOC	BOC	EOC	BOC	EOC	BOC	EOC	BOC	EOC
244-PU	0.00	0.00	0.00	0.01	0.00	0.01	0.01	0.02	0.02	0.03	0.03	0.04	0.04	0.05	0.05	0.05
241-AM	189.74	113.02	207.17	124.01	204.69	128.95	202.15	129.20	199.48	130.93	200.13	132.58	201.08	133.51	198.96	134.00
242-AM*	0.31	3.86	3.84	5.12	4.83	5.43	5.12	5.51	5.20	5.58	5.27	5.67	5.33	5.70	5.35	5.69
243-AM	25.39	30.55	41.10	45.32	54.65	57.19	64.86	66.00	72.44	72.72	78.47	77.88	83.27	81.96	86.64	84.76
242-CM	0.47	10.21	0.60	14.65	0.60	15.65	0.59	15.39	0.98	14.69	0.65	15.04	1.03	16.47	0.77	14.97
243-CM	0.09	1.12	1.05	1.70	1.57	1.87	1.71	1.90	1.72	1.87	1.70	1.88	1.70	1.95	1.79	1.89
244-CM	4.41	13.43	13.68	24.76	24.43	35.56	34.20	45.47	42.19	52.80	48.54	58.78	53.70	64.05	57.98	67.67
245-CM	0.32	1.51	1.62	3.45	3.76	5.66	5.96	7.79	7.92	9.52	9.57	11.10	11.07	12.29	12.15	13.29
246-CM	0.04	0.12	0.14	0.37	0.42	0.80	0.88	1.35	1.40	1.95	2.00	2.62	2.64	3.26	3.26	3.89
247-CM	0.00	0.01	0.00	0.02	0.02	0.05	0.06	0.09	0.09	0.15	0.16	0.21	0.22	0.28	0.28	0.35
Total Th's	9449.40	8495.30	8663.10	7824.30	7575.70	6907.90	6936.00	6334.30	6510.30	5970.70	6291.20	5782.20	6242.90	5747.40	6146.50	5664.40
Total Pa's	0.00	22.79	3.12	29.61	4.78	28.97	4.78	27.07	4.78	24.46	4.78	22.66	4.78	26.33	4.78	23.70
Total U's	0.00	602.14	0.00	539.53	0.00	446.53	0.00	405.45	0.00	372.52	0.00	355.08	0.00	347.36	0.00	338.65
Total Np's	138.30	76.73	136.75	77.64	127.20	76.36	119.35	72.43	112.86	70.10	109.76	68.97	108.69	68.95	105.74	67.56
Total Pu's	2136.64	1426.89	2351.97	1654.19	2424.80	1795.79	2455.06	1849.49	2458.94	1890.30	2479.87	1924.65	2506.20	1955.76	2498.46	1960.81
Total Am's	215.43	147.43	252.11	174.45	264.17	191.57	272.13	200.71	277.12	209.23	283.86	216.12	289.68	221.17	290.94	224.45
Total Cm's	5.32	26.41	17.08	44.94	30.80	59.59	43.40	71.99	54.30	80.99	62.62	89.62	70.36	98.30	76.24	102.05

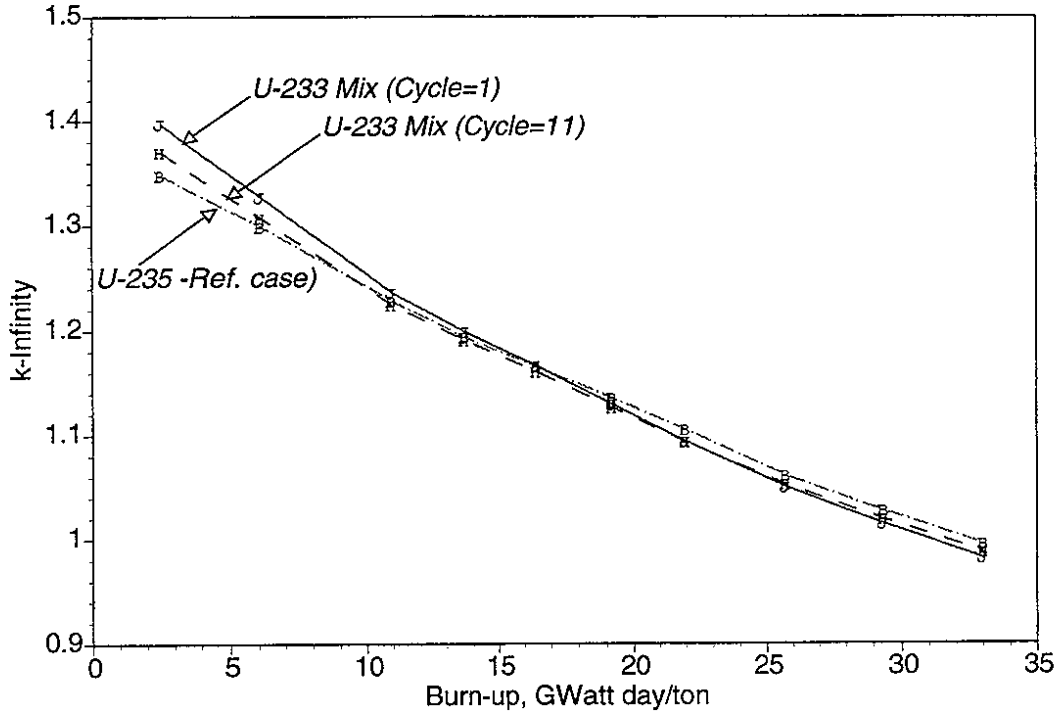

Figure 20. Comparison between standard ^{235}U enriched fuel and ^{233}U mix from the EA in a standard LWR. From Ref.[2].

4.5.- Recovery of ^{233}U-rich Uranium. As already pointed out, an inevitable feature of the scheme is the breeding of a substantial amount of ^{233}U out of the Thorium matrix, whose presence is indispensable to ensure a constant multiplication coefficient without interventions over the long burn-up time. Part of this ^{233}U is actually burnt especially during the latter part of the cycle, since it has to compensate for the losses of reactivity, mainly due to the incineration of the TRU's.

In addition at the discharge the produced ^{233}U rich Uranium is recovered (175 kg/year), since it is no longer necessary and in fact actually replaced by the fresh amount of TRU's added to the mixture. This is an added bonus, since the energetic content of the fissile material thus recovered is substantial. As already pointed out two possible further uses can be envisaged:

(1) the ^{233}U is stocked for a subsequent phase of operation of the EA without TRU's continuous injection at each cycle. This is for instance necessary in order to ultimately eliminate also the 2.26 tons of residue of TRU's from previous cycles. As shown in Ref. [2] this waste is

gradually eliminated and the mode of the EA operation progressively shifts to the one of TH-U mixture [1]. In principle there is no limit to the extent in which such an ultimate incineration can be pursued, with a 1/e reduction of the "old" residue in about 5.4 years, assuming the same energy production of 1.5 GWatt is continued at all times. A factor 100 in the last waste elimination is thus achieved in 25 years.

(2) The ^{233}U is used to blend the recovered Uranium from the LWR's in order to fabricate new fuel with the same criticality coefficient as fresh fuel, to be used to produce power in the LWR's, thus saving the expense of isotopic enrichment. As shown in Ref. [2], from which we have taken Figure 20, the ^{233}U and ^{235}U give rise to fuels which are extremely similar. Note however that the ^{233}U contains as much as 1% of ^{232}U, which is very radioactive because of the Thallium presence in its decay chain[1].

Since the ^{233}U-rich mixture will be therefore burnt either in the EA or eventually in the LWR's it does not constitute a "waste" and no long time storage has to be foreseen.

4.6.- The Particle Accelerator. The particle Accelerator needed to drive the EA has been already described in a number of previous papers [27]. Its detailed discussion is outside the purpose of this paper. Therefore we shall limit ourselves here to a brief summary of its main characteristics.

Particles accelerated are protons of sufficiently high energy as to produce an efficient yield of spallation neutrons. An experiment at CERN [15] has shown that the optimum kinetic energy should be not less than about 1 GeV. Then the choice of the energy is mainly related to the power required from the beam and the corresponding accelerated current, in turn primarily determined by the power to be dissipated in the target window. The nominal, calculated energetic gain for the EA and k= 0.97 is G = 83 corresponding to a beam current $i_o = 18/T_p$ mA, where $T_p \geq 1$ is the kinetic energy of the beam in GeV. The current must vary from about 0.6 i_o to 1.3 i_o in order to ensure constant power output at all times. Therefore the maximum current that the

[1] As already pointed out this represents a powerful deterrent against military diversions of the produced ^{233}U.

accelerator has to produce is $i_{max} = 24/T_p$ mA. Setting T_p = 1.5 GeV, the nominal maximum current is therefore 16 mA. In the design a current of 20 mA is being used, to provide for some operational margin.

The accelerator uses "status of the art" technologies and it consists in three separate stages:

(1) A warm LINAC injector, for a final energy of 25 MeV, made of a RFQ stage for the first 2 MeV, followed by a Drift tube structure (DTL), operating at 350 MHz. The average energy gain is about 1 MeV/m. This component is probably commercially available.

(2) A six sector cyclotron, operating at 1/5 of the LINAC frequency (70 MHz) to accelerate proton energy to about 350 MeV. The machine is largely inspired from the PSI accelerator, presently in operation. The third stage is either:

(3a) a superconducting LINAC structure, derived from the presently operating LEP200 accelerating cavities [9] which accelerates particles to a final energy of 1.5 GeV. The length of the accelerating structure is about 500 m long and the operating frequency is 350 MHz. Or, alternatively

(3b) a 10-12 sector cyclotron of the type described in Ref. [27]. This second solution is much more compact and requires the same technologies as point (2).

The elements (1) and (2) can also constitute a viable accelerator for a lower power demonstration EA incinerator [28] which should be realised first.

The overall efficiency of the accelerator complex is of the order of 50 %. Efficiency is intended as the ratio between the beam power and the mains load of the full accelerator complex.

5.— INCINERATION OF LONG LIVED FISSION FRAGMENTS ?

5.1.- General considerations. The main purpose of the EA incineration is the one of eliminating the TRU's elements which represent the bulk of the residual toxicity after a LWR cycle. However, once this task has been successfully accomplished there are a few long-lived elements which represent the main contribution to the long term radio-toxicity of long lived waste. It is therefore reasonable to consider the possibility of incinerating (transmuting) them as well. While incineration of TRU's proceeds through fission and therefore it eliminates the element producing a large amount of energy, in the case of FF's we can only transform them by neutron capture. However, since the FF's are intrinsically neutron rich, the addition of one extra neutron is normally transforming the long lived element in other elements which have a much shorter lifetime.

This method in order to work, requires a high isotopic purity of the isotope to be eliminated, otherwise parasitic neutron captures in the other presumably stable isotopes can in some circumstances not only be costly in terms of neutrons, but also produce additional radio nuclides. As shown in Table 5, it is a fortunate circumstance that the two elements ^{99}Tc and ^{129}I which are the largest contributors to the residual radioactivity of the waste, are also essentially pure isotopes. The total dilution volume without them will be reduced to a mere 4.32 % of the total volume. Therefore our considerations will be limited to the task of eliminating only these two isotopes. While the Technetium is essentially a pure ^{99}Tc isotope, Iodine contains also 22.9 % of the stable isotope ^{127}I. Technetium should be separated from the Noble Metal stream for instance by pyro-electrolysis. Iodine is present in the waste stream as a compound and it should be possible to isolate it chemically, since Iodine is specifically reacting.

The neutron capture reactions relevant to the transmutation are then the following

$$^{99}Tc + n \rightarrow {}^{100}Tc \xrightarrow{\beta-dec(15.8\,sec)} {}^{100}Ru \quad (stable)$$

$$^{129}I + n \rightarrow {}^{130}I \xrightarrow{\beta-dec(12.36h)} {}^{130}Xe \quad (stable)$$

$$^{127}I + n \rightarrow {}^{128}I \xrightarrow{\beta-dec(24.99m)} {}^{128}Xe \quad (stable)$$

The final products are all stable. While the daughter Xenon nucleus is easily extracted from the transmutation volume before it has the time to

capture another neutron, Ruthenium is a metal which will make an alloy with Technetium and therefore it will remain exposed to the neutron flux for the whole duration of the transmutation process. It is however a fortunate circumstance that ^{100}Ru, ^{101}Ru and ^{102}Ru are all stable isotopes and all have a rather small neutron capture rate. Therefore, as we shall see, their presence is harmless.

The amounts from the LWR discharge that a single EA unit has to transmute are 39.6 kg/year of ^{99}Tc and 9.20 kg/year of ^{129}I (contained in 12.3 kg/year of Iodine). The additional, intrinsic production of these isotopes by the EA itself are 9.5 kg/year and 5.0 kg/y respectively [1]. Therefore the total quantities to be eliminated are of the order of 50 kg/year and 15 kg/year respectively. In order to achieve this goal about 10% of the neutrons produced in the EA must be dedicated to the task. This implies a very high capture efficiency to these elements and new, promising methods are under study [16]. They are based on the so called "Adiabatic Resonance Crossing" (ARC) which is briefly described. At this stage it appears that they hold the potentials to transmute effectively both the contribution from the LWR waste and the one produced by the EA as the result of the incineration of the TRU's. However more work is needed in order to conclusively prove this point.

5.2.- Capture enhancement by Adiabatic Resonance Crossing (ARC). At present, neutron capture is normally performed by introducing the sample in a strong neutron flux, for instance in the core of a Nuclear Reactor. We can however achieve a vastly increased neutron capture efficiency introducing a small mass of the material to be activated in a highly transparent, diffusive medium for neutrons which is the Lead volume surrounding the core. We can exploit in this way the large value of the capture cross section in correspondence to the nuclear resonances which are all traversed in very tiny energy steps in a medium of large mass number, which has consequently a small "lethargy"[1] and a smooth, otherwise unperturbed, energy slow down of the initially high energy (MeV) neutrons. Neutrons used for the FF transmutation task are those which have already leaked out from the core and

[1] Lethargy is defined as the fractional average energy loss at each neutron elastic collision.

have a small probability of re-entering it. Therefore the transmutation is performed with little or no consequence on the operation of the EA core.

The neutron propagation in a medium of small lethargy and highly transparent to neutrons has peculiar properties which are exploited for the transmutation. The absorbing sample is diffused inside the bulk of the diffusing medium. Using an optical analogy, the target-moderator sphere is chosen to be diffusive but highly transparent to neutrons. Doping it with small amount of additional material makes it "cloudy". As a consequence most of the neutrons are captured by the absorbing impurities.

Assume a large volume of transparent, diffusing medium of sufficiently large size to contain most of the neutrons, as it is the case of the Lead volume surrounding the core of the EA. The source, assumed point-like is located at its centre. Consider a neutron population in a large, uniform medium of N scattering centres per unit volume, with very small absorption cross section σ_{abs} and a large scattering cross section σ_{sc}. All other cross sections are assumed to be negligible, as it is generally the case for neutrons of energy substantially smaller than 1 MeV. Since the angular distribution of these collisions is almost isotropic, they have also the important function of making the propagation of neutrons diffusive, and therefore maintain the neutrons "cloud" within a smaller containment volume. The neutron flux[1] $\phi(x,y,z)$ in such a volume is defined as the number of neutrons crossing the unit area from all directions per unit time. The reaction rate ρ_x, defined as the number of events per unit time and unit volume, for a process of cross section σ_x is given by $\rho_x = \phi\ N\ \sigma_x = \phi\ \Sigma_x$, where we have defined with $\Sigma_x = N\ \sigma_x$ the macroscopic cross section for the process x. For a steady state, Fick's law leads to the well known differential equation :

$$\nabla^2\phi - \frac{\Sigma_{abs}}{D}\ \phi = -\frac{S}{D}$$

where S is the neutron source strength (rate) and $D = 1/(3\ \Sigma_{sc})$ the diffusion coefficient for isotropic scattering. For anisotropic scattering, a correction must be introduced, $D = 1/3\ \Sigma_{sc}(1 - \mu)$, where $\mu = < \cos\theta >$. Note that for Lead $\mu \approx 0$ and $D = 11.2$ mm.

[1] At this point the energy spectrum of the neutrons is not considered, namely the flux is the one integrated over the energy spectrum.

If we place a point source at the origin of the co-ordinate system, equation (1) will hold everywhere with S = 0, except at the source. The approximate solution of the differential equation is:

$$\phi(r) = S_o \frac{e^{-\kappa r}}{4 \pi D r} \quad ; \quad \kappa = \sqrt{\frac{\Sigma_{abs}}{D}} = \sqrt{3\, \Sigma_{sc}(1-\mu)\Sigma_{abs}}$$

S_o is the rate of neutrons from the source per unit of time (n/sec). The elastic scattering being relatively large ($\sigma \approx 11$ barn for Lead) and the absorption probability very small, D is a small number, while $1/\kappa$ is large (metres). For a region close to the source, namely $\kappa r << 1$ the flux is given by $\phi(r) \approx S_o/4\pi D r$, namely is considerably enhanced with respect to the flux in absence of diffuser $\phi_o(r) \approx S_o/4\pi r^2$ by the factor $F = \phi(r)/\phi_o(r) = r/D$. For a typical sample distance of r =30 cm, the enhancement factor is very substantial, especially for Lead, for which F = 30/1.12 = 27. The diffusing medium is acting as a powerful flux enhanced, due to multiple traversals.

As we have seen, in order to achieve an effective rate of activation, the neutron flux must be as high as possible. But, in addition, also the energy spectrum of the neutrons must be matched to the energy dependence — usually rather complicated — of the capture cross section of the relevant isotopes. In general, the energy spectrum of the bare source is not optimal because its energy is too high to produce effective capture rate. Therefore an energy matching (moderation) must be performed before utilisation. This second, important function must also be performed by the diffusing medium.

In the case of Lead, the energy of such neutrons is progressively shifted in a multitude of small steps by a large number of multiple, elastic collisions. The logarithmic energy decrement for Pb is very small $\xi = 9.54 \times 10^{-3}$ and the average number of collisions to slow down from 5 MeV to 0.025 eV (thermal energies) is $n_{coll} = \ln(5\ \text{MeV}/0.025\ \text{eV})/\xi = 2.0 \times 10^3$. The elastic cross section, away from the resonances is about constant down to thermal energies and large (around 11 barns). The total path l_{coll} to accumulate n_{coll} is then the enormous path of 61.1 meters ! The actual drift distance travelled is of course much smaller, since (1) not all neutrons will survive such a long path and (2) the process is diffusive. Neutrons loose at each step a constant fraction of their energy, the resultant energy spectrum is then flat in the variable dE/E = d(ln(E)). Neutrons scan the full energy interval down to thermal energies, "seeking" for large values of the capture cross section of the added impurities due to strong resonances. This method is evidently profitable if such strong

resonances exist elsewhere than at thermal energies. It is a fortunate circumstance that this is the case for ^{99}Tc and ^{129}I.

If a small amount of impurity to be activated is added to the transparent slow moderating medium, captures by the added element will occur. We introduce the quantity $P_s(E_1, E_2)$ which we call survival probability, defined as the probability that the neutron moderated through the energy interval $E_1 \rightarrow E_2$ is not captured. Evidently such probability is defined for a large number of neutrons in which the actual succession of energies is averaged. In the good approximation that the elastic cross section is energy independent, we can write the formula:

$$P_s(E_1,E_2) = \exp\left(-\frac{1}{\xi\,\Sigma_{sc}}\int_{E_2}^{E_1}\frac{\Sigma_{abs}(E)}{1+\Sigma_{abs}(E)/\Sigma_{sc}}\frac{dE}{E}\right) =$$

$$= \exp\left(-\frac{N_u}{\xi\Sigma_{sc}}\int_{E_2}^{E_1}\frac{1}{1+\sigma_{abs}(E)N_u/\Sigma_{sc}}\sigma_{abs}(E)\frac{dE}{E}\right) \le \exp\left(-\frac{1}{\xi\,\sigma_{sc}}\frac{N_u}{N_m}\int_{E_2}^{E_1}\sigma_{abs}(E)\frac{dE}{E}\right)$$

where the last equality holds for infinitely diluted impurity and N_u and N_m

Table 13. Resonance Integral and corresponding capture probabilities.

Element	Integral (barns)	Conc. by weight for 62% captures
^{99}Tc	300	1.53×10^{-4}
^{100}Ru	11	4.19×10^{-3}
^{101}Ru	100	4.65×10^{-4}
^{102}Ru	4.7	1.00×10^{-2}
^{129}I	50	1.19×10^{-3}
^{127}I	150	3.90×10^{-4}

are the number atoms per unit volume for the impurity and the moderating medium respectively.

The main resonance integrals are listed in Table 13. In such approximation we can estimate the magnitude of the capture probability. For instance, setting the exponent equal to 1 (namely $P_s(E_1, E_2) = 1/e = 0.367$, corresponding to 63.2% of neutrons captured by the impurity) we find $N_u/N_m = \xi$ (11 barn)/(300 barn) = 3.5×10^{-4}, or about only 2.28×10^{-4} Technetium concentration by weight. The capture probability of ^{129}I is about a factor 6 smaller, which is not dramatic, in view of the smaller amounts to transmute. Note also that the ^{127}I contamination has a factor three higher

cross section. Since its fractional concentration is smaller (25%), roughly equal amounts of captures occur in each of the two isotopes. The daughter nuclei of Iodine transmutation are Xenon gases which are extracted before they accumulate appreciably. The ^{99}Tc daughter nucleus (^{100}Ru) has fortunately a capture probability which is only 3% of the one of the father nucleus, and its residual presence has no adverse consequence.

The above formulae neglect the absorption in the moderating medium[1]. The flux is significantly attenuated during slow down. Hence the fraction of captures will have to be corrected for the self-absorption. These formulae above are approximately valid and they can give only the qualitative features of the phenomena. In such linear approximation each element is contributing, so to say, independently. However, if a resonance is strong enough to absorb a major fraction of neutrons, it may "shield" other resonances occurring at lower energy. Then the element which has a dominating resonance group at higher energies can void the captures of the elements "downstream". This effect may be very important. Finally the lethargy is modified by the elastic part of the resonance. The flux is locally decreased (dip) due to the shorter path needed to make the collision.

In practice, computer simulations with the appropriate time evolution are the only valid method to predict with precision the performance of the device.

5.3.- Computer simulated Technetium transmutation. The Montecarlo simulation programme used to calculate the evolution through the cycles of the EA operation has been extended to simulate the transmutation of ^{99}Tc in the "halo" of the neutrons emitted by the core and drifting in the surrounding Lead medium. To that effect a small ^{99}Tc contaminant has been introduced in two volumes about 1 metre above and below the core. This distance and position are not critical and the potential transmutation volume is effectively around all the core volume.

The effects of the presence of ARC and consequent captures are striking on the neutron spectrum, (Figure 21), especially in view of the very small

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[1] The resonance integral for natural Lead is 0.137 barn, namely 1.247% of the elastic cross section.

concentration used (260 p.p.m.). The spectrum is plotted as a function of dE/E, such that a lethargy-dominated, diffusing spectrum will be flat. One can notice a significant drop in flux due to the adiabatic crossing of two strong 5.6 eV and 20.3 eV resonances, as well as of the continuum above them. Above 1000 eV one can observe the resonance lines due to Lead. The energy spectrum is of course much softer than the one in the core and the build-up is due to diffusion into the surrounding Lead, with corresponding lethargic energy losses.

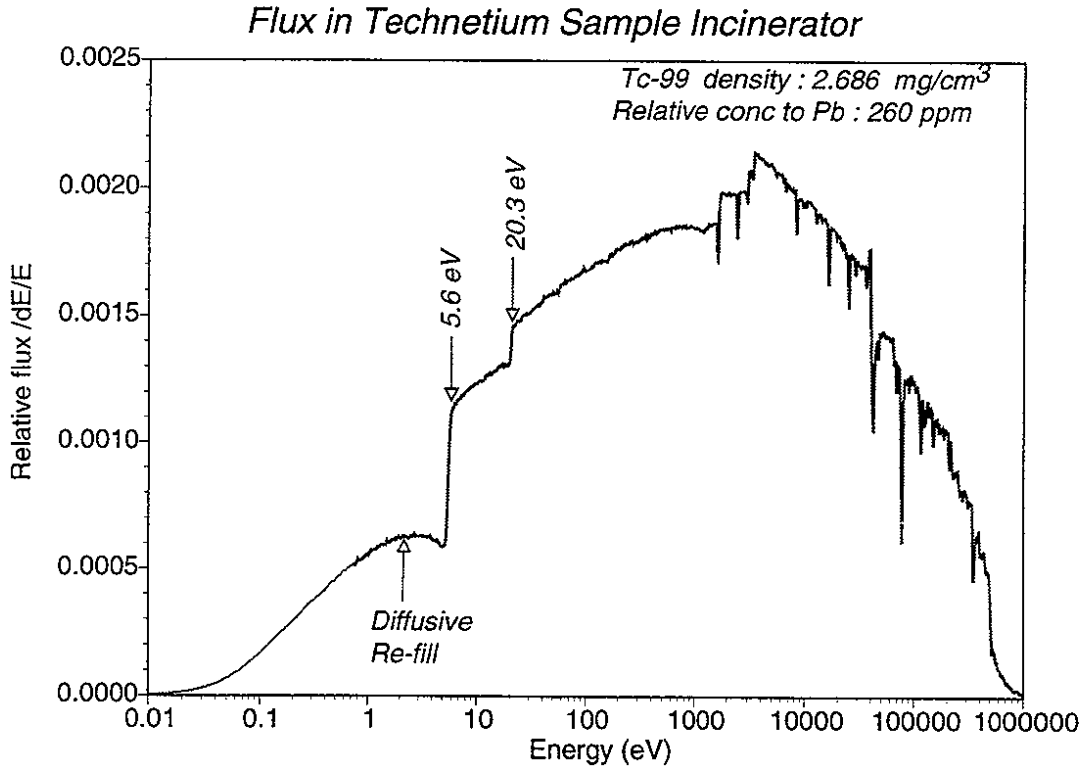

Figure 21. Spectral absorption due to adiabatic resonance crossing in the Lead volume surrounding the core due to 260 p.p.m. Technetium contamination.

The time evolution of the ^{99}Tc concentration in the volume of the sample transmuter (0.409 m^3) has been calculated over the 120 GWatt d/t standard burn-up of the EA and it is shown in Figure 22. The total mass of initial ^{99}Tc is 1.1 kg, corresponding to 2.686 mg/cm^3. One can observe the almost exact exponential decay of the father element with a time constant of 82.1 GWatt d/t (1.36 years) and the corresponding build-up of ^{100}Ru daughter isotope. The higher order captures, as expected, are negligibly small. This somewhat arbitrarily chosen initial concentration corresponds to an (initial) transmutation rate of the order of 1.1/1.36/0.409 = 1.98 $kg/m^3/y$. The

complete transmutation of the 50 kg/year requires then a transmutation volume of 50/1.98 = 25.2 m^3, which is easily arranged around the core.

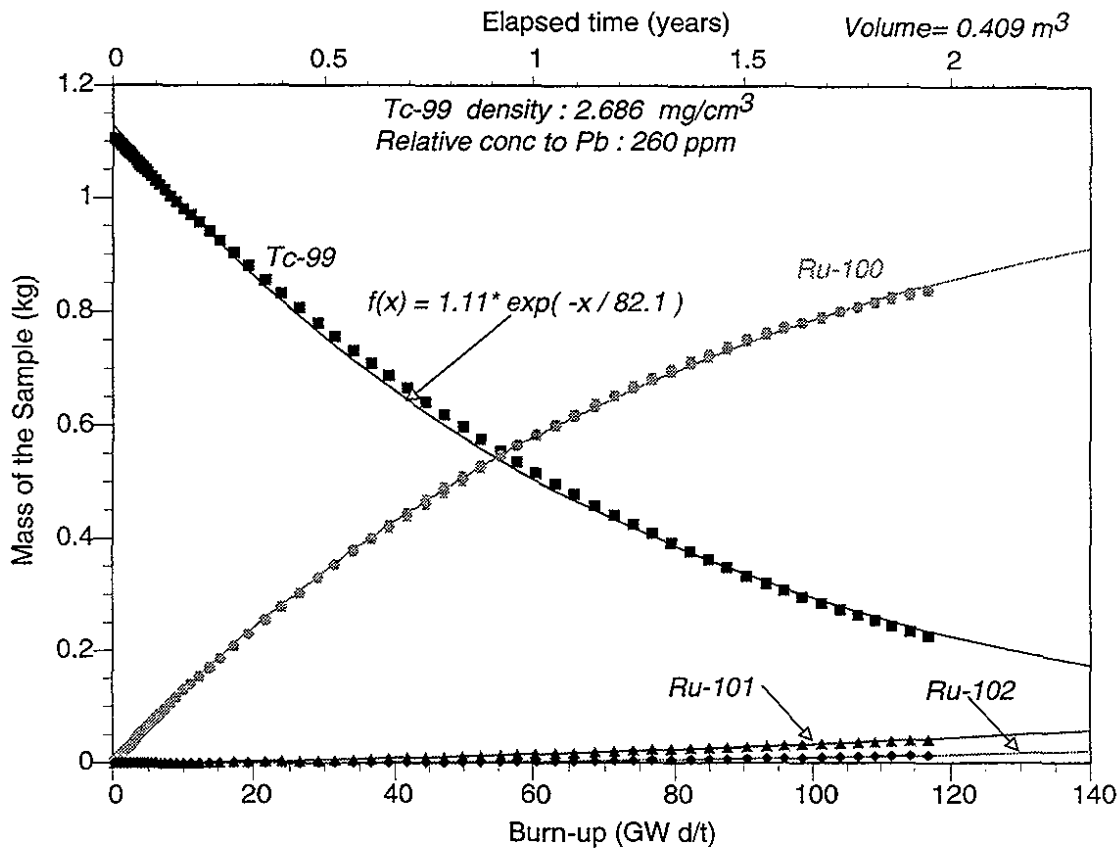

Figure 22. Time evolution of the transmutation of the ^{99}Tc by ARC.

We would like to stress the preliminary nature of this figure, which however shows that a massive transmutation of unwanted FF's is promising and most likely feasible. The engineering of the device has to be studied, for instance in which way Technetium and the Iodine are introduced within the volume etc. However we anticipate no major problems.

6.— CONCLUSIONS.

The total expected (2029) fuel waste from the Spanish LWR's (14400 units, 9628 tons) can be divided in three streams of decreasing radiological concern and which can be separated by the pyro-processing techniques and once the bulk of the material, the unburned Uranium (6296.83 tons), is recovered for further use:

(1) the TRU's (73.42 tons) which carry criticality and high activity at very long times. They are incinerated in the EA's and thus eliminated from the waste stream. This is made possible because they represent only 1% of the initial spent fuel mass.

(2) The specific activity of ^{90}Sr (3.81 tons) and ^{137}Cs (22.5 tons) amongst the volatile FF's which have a very intense initial activity, but characterised by a half-life of about 30 years. Their time of decay is sufficiently short to justify a specialised Cool-Down facility. In addition the ^{85}Kr , which instead of being sent into the atmosphere is retained until it has decayed with a half-life of 10.7 years.

(3) The remainder of the FF's (244 tons, 3.87 % of the fuel mass) can easily meet at the natural duration of the reprocessing procedure Class A LLW conditions for definitive surface disposal, according to the US NRC Regulations [8]. However environmental concerns, beyond the requirements of 10 CFR Part 61.55, suggest that at least the bulk of ^{99}Tc (6.06 tons) and ^{129}I (1.40 ton) should be transmuted parasitically in the EA, leading to stable, harmless elements. It has been shown that this is possible with the ARC method. Some questions remain on the procedure to be followed in the case of ^{135}Cs (3.16 tons), which is initially mixed with the rest of strongly active Volatiles (Strontium and Cesium, see point (2)). Again according to 10 CFR Part 61.55, it could be disposed in a LLW or eventually in excess to those Rules, transmuted after the long cool-down period in the Cool-Down facility. This is a problem for future generations, but with a technology we can fully define with today's know-how.

The general scheme is summarised in Figure 23. The incineration of the TRU's in the 5 EA units can be performed in about 37 years. At the end of this process an additional phase of energy production is proposed without periodic injection of TRU's from the LWR's in order to completely incinerate the residual TRU's from the LWR's. The duration of this second phase

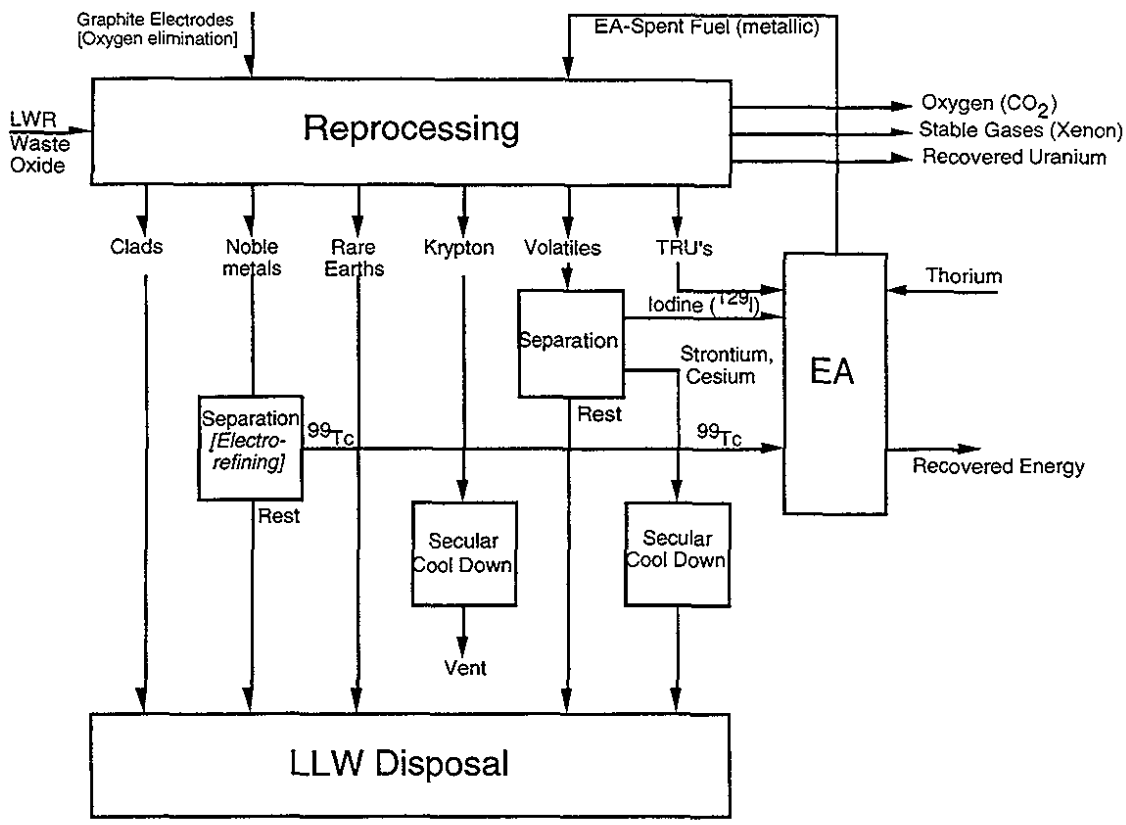

Figure 23. Simplified, general diagram of the Incineration procedure.

coincides with the end of the life of the EA Plant, which has been assumed to be >60 years.

It would seem to us that there should be no major difficulty in packing the more resilient radionuclides of the Secular Cool-Down in such a way as to make full use of the reduction due to decays, for instance several hundreds of years.

At the end of the active life of the Plant, the Secular Cool-Down will remain in activity until such a time that Krypton can be safely vented into the atmosphere and that Strontium and Cesium have decayed sufficiently, as to be transferred to the ultimate LLW repository on surface or at shallow depth.

The size of the ultimate LLW repository on surface or at shallow depth which eventually will collect all the residual waste from the LWR's of Spain, is comparable to and will comply with the requirements of the now operational site "El Cabril".

This strategy, also taking into account the relatively small volume of the surviving active waste, is offering all the guarantees of containment and it is considerably simpler than the Geologic Repository alternative.

Finally it is worth underlying once again the considerable amount of energy recovered through incineration and the benefits of recycling the unburned Uranium.

In a nutshell, incineration makes a lot of sense environmentally and financially. We believe it is technically perfectly feasible .

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