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Review

Basics of accelerator driven subcritical reactors

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Abstract

This paper is an introduction to the physics of Accelerator Driven Subcritical Reactors (ADSR) and some technologies associated with them. The basic neutronics is presented with a specific discussion of modifications with respect to that of critical reactors. The fuel evolution in ADSR's is discussed, including the influence of reactivity surges and drops on the limitation of the design reactivity. The application of ADSRs to nuclear waste management is examined and the different options are discussed. Finally, some practical proposals are briefly discussed.

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1. Introduction

In recent years great interest has been displayed, worldwide, for accelerator driven subcritical reactors (ADSR or ADS), also called here subcritical reactors on hybrid systems, to produce energy and transmute radioactive wastes in a, possibly, cleaner and safer way than at present. Pioneers in this revival have been Furukawa [1], Bowman [2] and Rubbia [3]. Similar ideas were first proposed almost 50 years ago [4–14]. At that time they were not carried through, not so much due to technical difficulties but for lack of economic incentive. Also, while building reliable GeV accelerators achieving intensities of several tens of milliampere was by no means considered to be a trivial matter, performing critical reactors were available, and were thus thought to be the simplest and most natural way to harness nuclear fission. It was, however, acknowledged that accelerator driven

nuclear devices might give interesting transmutation possibilities. For example, without a large enough concentration of ^{235}U in natural uranium, the only way to exploit fission energy would have been the use of such subcritical systems.[1]

High energy accelerators appear to be a promising way to incinerate heavy actinides [15,16]. It has also been acknowledged that a thorium-based fuel cycle would considerably limit the amount of produced transuranic wastes. The implementation of such a cycle would be made easier with subcritical reactors, due to the improved neutron economy of such systems as compared to classical critical reactors [1–3]. Subcritical reactors appear to be a credible alternative to fast breeder and fusion reactors.

The aim of this review is to provide the reader with a basic understanding of subcritical reactors and discuss some practical examples. It focuses, in particular, to the possible contribution of subcritical reactors to the solution of the nuclear waste problem. It is based on a more complete review published in Progress in Particle and Nuclear Physics [17], to which we will, occasionally, refer as PPNP.

We have deliberately chosen to give simple, intuitive treatments of the different aspects of the physics of subcritical reactors, in order to provide the reader with the possibility to gain physical insight into these systems. Because of the limited amount of space, this choice has prevented us to give detailed presentation of present work in the field. Also, the reader should be aware that realistic calculations require the use of complex codes, most frequently of the Monte Carlo types, and that the considerations which are explicited here should not be used in place of them. However, it is our experience that starting with simple models helps understanding the results of the "real" calculations, and provide intuition of the most fruitful ways to improve systems or find new ones. As far as possible this review is self-contained and does not require a priori knowledge of the field. However, some knowledge of the physics of critical reactors is, occasionally, required and can be obtained in PPNP.

[1] Using any kind of energy source, accelerators allow ample production of neutrons which might be used for synthesis of fissile nuclei starting from the fertile nuclides ^{238}U and ^{232}Th.

2. Energy and waste production with standard and breeding reactors

Subcritical reactors have to be appreciated in view of the general situation and possible future of power generation by nuclear reactors. Thus we give, here, a brief discussion of the general Nuclear Power paradigm.

2.1. Standard reactors

Most existing energy producing reactors are of the light water cooled type, either Pressurized Water (PWR) or Boiling Water (BWR) reactors. Although other types of commercial reactors like the heavy water CANDU have interesting characteristics, our discussion is focused on the Light Water Reactors. The power produced by commercial reactors ranges between 600 and 1500 electric MWatts (MWe), with thermodynamical efficiencies close to 33%. As a sample case, we consider a 1000 MWe reactor.

Each fission produces approximately 200 MeV (185 MeV at the moment of fission and 15 MeV produced by β-radioactivity). Accordingly the fission of 1 kg of a fissile isotope typically produces 80 Terajoules (or 1900 Toe[2]). It follows that a 3 GW (thermal gigawatts) reactor, yielding 1 GWe (electrical gigawatt), produces annually about 7 TWhe for an availability of 80%. It burns about 1 ton of fissile isotopes which is equivalent to 2 million Toe. More precise numbers are given in Table 1 where material inventories at loading and discharge are given.

In Table 1, a burn-up of 33 GWd/ton (Gigawatt-day/metric ton) is assumed. The table shows the following interesting features:

- The amount of ^{235}U which has disappeared equals 674 kg. This accounts not only for the fission of this nuclide, but also for its neutron

[2] Metric ton oil equivalent.

capture, at least the 111 kg of produced ^{236}U.

- This means that at least 383 kg of the higher isotopes, mostly ^{239}Pu have contributed to fission. This can also be considered as indirect fission of the ^{238}U isotope, which lost 673 kg corresponding essentially to the production of plutonium. Of these 673 kg only 286 kg are found in the form of plutonium isotopes and minor actinides.
- The mass balance between the initial and discharge inventories is not exact. This is due to the mass equivalence of the energy produced (about 1 kg) and to the neutrons captured in the structure elements and cooling water (2 kg corresponding to approximately 0.5 neutron per fission).

As usual, energy production is accompanied by waste production. The nuclear wastes to be considered can be divided into three categories:

1. The plutonium and Minor Actinides (Np, Am, and Cm) have very high radiotoxicities due to their dominant α decay. They have long lifetimes, up to 25 000 years for ^{239}Pu. They would require either long-term underground storage or transmutation. In the latter case they can only disappear by fission (this is usually called incineration). The fission of 280 kg of plutonium and Minor Actinides would produce about 2 TWh of electrical energy. This means that at least one incinerating reactor for 4 PWRs would be needed if one wants to incinerate completely plutonium and Minor Actinides.
2. The long-lived Fission Fragments. These are nuclides with lifetimes larger than 1000 years which decay by β emission. The main fission fragments involved are shown in Table 2 together with the amounts produced yearly by a 1 GWe reactor.
3. The medium-lived Fission Fragments, essentially ^{90}Sr and ^{137}Cs which have very high activities at discharge and small neutron capture cross-sections. It does not seem realistic to transmute them and they would, then, set a minimum time length of around 300 years over which the waste activity will require special storing care.

Table 1
Inventories at loading and discharge of a 1 GWe PWR [19]

Nuclides	Initial load (kg)	Discharge inventory (kg)
^{235}U	954.0	280.0
^{236}U		111.0
^{238}U	26 328.0	25 655.0
U total	27 282.0	26 047.0
^{239}Pu		56.0
Pu total		266.0
Minor actinides		20.0
^{90}Sr		13.0
^{137}Cs		30.0
Long-lived PF		63.0
PF total		946.0
Total mass	27 282.0	27 279.0

The size of the waste problem is appreciated from Table 3 which shows the inventory of spent fuels in the OECD countries in 1995.

We recall that nuclear power only accounts for 4.5% of the total world energy production. Although small, this percentage will lead to a spent fuel inventory of about 200 000 tons by the year 2020. The yearly production of spent fuels amounts to about 8000 tons. This figure is to be compared to the present spent fuel recycling capabilities of around 200 tons, mostly by the COGEMA La Hague facility. Should nuclear power rise to 30% of the world energy production (which would be significant with respect to the greenhouse issue), the yearly production of spent

Table 2
Long-lived fission fragments with their half-lives and production rates

Nuclide	^{79}Se	^{90}Zr	^{99}Tc	^{107}Pd	^{126}Sn	^{129}I	^{135}Cs
$T_{1/2}$ years	70 000	1.5×10^6	2.1×10^5	6.5×10^6	10^5	1.57×10^7	2×10^6
Production kg/yr	0.11	15.5	17.7	4.4	0.44	3.9	7.7

Table 3
Data concerning the end of cycle in OECD countries [19]

Countries	Nuclear power (GWe)[b]	Share of nuclear power (%)[c]	Spent fuels[a]
France	58.5	76.4	11 770
Belgium	5.5	55.8	1400
Sweden	10.0	51.1	3240
Switzerland	3.0	36.8	1300
Spain	7.1	35.0	1775
Finland	2.3	29.5	975
Germany	22.7	29.3	6315
Japan	38.9	27.2	8600
UK	11.7	25.8	7000[d]
USA	98.8	22.0	28 600
Canada	15.8	19.1	20 000[e,f]
Netherlands	0.5	4.9	150
Total	274.8		91 125

[a] Cumulated tons in 1995 (E.U. estimate).
[b] January 1st 1995 (LAFA).
[c] As compared to the total electric energy production.
[d] Authors' estimate.
[e] Canada uses natural uranium fueled reactors (CANDU), hence the large stock.
[f] Authors' estimate for 1995 from Ref. [20].

fuels would be close to 40 000 tons. To appreciate the meaning of such an amount it is interesting to compare it to the storage capability of discussed nuclear waste repositories. We take the example of the US Yucca Mountain site which is considered as the possible unique site for deep underground storage of nuclear wastes in the US. The site would cover about 6 Km^2 honeycombed with about 100 km of tunnels [21], while the maximum storage capacity should be 70 000 tons. The cost of the site would be more than 15 billion dollars. These figures show that while the option of deep underground storage seems realistic in the perspective of a withdrawal from nuclear power in the first half of the next century (only a few sites like Yucca Mountain would be needed), it appears much more problematic if nuclear power is to contribute significantly to the reduction of greenhouse gas emission.

Aside from the back-end waste question, the poor use of uranium in the present thermal reactors has consequences on the amount of mining required, as well as on the level of resources. In the absence of recycling, each 1 GWe reactor requires, annually, 200 tons of fresh natural uranium.[3] Typically, currently used uranium ores have grades around 0.25% [21]. This means that a 1 GWe reactor requires extraction of 80 000 tons of ore, to be compared to the 2 million tons of oil which would be needed to produce the same amount of energy. The rather large amount of mill tailings is associated to radioactivity due to the progeny of uranium, especially to a continuous flow of radon during a long period (75 400 years as defined by the lifetime of the parent ^{230}Th). This radon gas escapes more readily from the tailings than from the unmined uranium ore.

The uranium reserves are estimated around 15 million tons at costs close to the present. The present world power production is about 350 GWe, requiring 70 000 tons of natural uranium yearly. Thus, the present reserves are estimated to last 200 years. Again, it is not a problem as long as the present small contribution of nuclear power to the overall energy production is kept. However, as in the wastes case, should the nuclear share increase to a 30% level, the reserves

[3] This includes process losses as well as the effect of incomplete burning of ^{235}U.

would be reduced to approximately 40 years, not more than the oil reserves. One should note, however, that there is a very large reserve of uranium in sea water, amounting to 4 billion tons of uranium [21], at a concentration of 3.2 parts per billion. It seems possible to extract this uranium at a cost 10 times larger than the current price, which would increase the cost of the produced electricity by 50%.

2.2. Breeding reactors

The use of breeding or converter[4] reactors would change considerably the above picture. Converters and breeders allow full use, not only of the fissile ^{235}U isotope, but also of the fertile ^{238}U and ^{232}Th isotopes. Thus, in principle, a 1 GWe reactor requires only 1 ton of natural uranium or an equivalent amount of thorium. This means that, at the current market cost, assuming a production capacity of 2500 GWe corresponding to a nuclear share of 30% in the total energy production, the reserves would amount to 6000 years for natural uranium and about 4 times more for thorium. In fact, the very effective use of the uranium and thorium would allow the use of very poor ores, including the sea water uranium, which means that the resources would be practically, infinite. The mill tailings would also be considerably reduced by more than a factor 100.

While the plutonium present in spent fuels has to be considered as a waste, it is the fissile material for breeders and converters. Only long-lived fission products (LLFP) and minor actinides (MA)[5] can, thus, be considered as nuclear wastes. In the absence of specific transmutation of these wastes, their radiotoxicity, after a cool down period of 300 years,[6] would be, at least, one order of magnitude smaller than that of the PWR spent fuels, for an equivalent energy production. Since fuel reprocessing is a prerequisite for most breeding or converting cycle it is quite natural to consider the possibility of trasmuting the LLFP and MA. We shall discuss such a possibility in some details below. It is shown that incineration of MA and transmutation of some of the most significant LLFP appear to be feasible. Nuclear wastes would, then, be reduced to the reprocessing losses. Modern reprocessing is claimed to have 99.9% efficiency in the recovery of plutonium and 99% in the recovery of MA [22]. It would, then, be possible to reduce the total radiotoxicity of the wastes by several orders of magnitude after a few hundred years of cooling time. With such a reduction, long-term storage might not be necessary.

While the reliability and safety of PWRs has been widely demonstrated in industrialized western countries,[7] the experience with breeder or converter reactors is limited and ambiguous. By far, the best-known breeders belong to the Liquid Metal Fast Reactors type. Practically all of these liquid metal reactors have used Sodium as a coolant, with the exception of several recent Russian submarines propulsion reactors which are cooled with liquid lead–bismuth eutectic. While it seems that the records of the Russian sodium cooled reactors like BOR60, BN350 and BN600 appear to be very good, those of such western reactors is much more questionable. The small American reactor EBR2 worked satisfactorily until its closure in 1995. Another American reactor, Enrico Fermi, could never work. The small French reactor RAPSODIE worked very nicely until it stopped. The 250 MWe Phenix reactor did work satisfactorily during 10 years until unexplained reactivity fluctuations led to its stopping. It is planned to start again, pending safety improvements. The large, 1200 MWe Super Phenix reactor was plagued by sodium leaks and administrative imbroglio until it was decided to

[4] Breeding reactors produce more fissile material than they consume while converter reactors produce as much fissile material as they consume.

[5] Np, Am and Cm are produced in relatively small quantities in normal reactors and are thus called Minor Actinides.

[6] This cooling time is necessary to allow for decay of ^{137}Cs and ^{90}Sr whose transmutation would be very difficult and costly. Due to their short lifetime these isotopes dominate the waste's radiotoxicity, in the short run.

[7] Even in the former USSR the reactors analogous to the PWR, the VVER are considered to be safe by the international experts, while the RBMK reactors, such as those of Tchernobyl, are unanimously considered as unsafe.

stop it definitively. The Japanese Monju reactor is, also, suffering from sodium leaks. It seems that the combination of more and more stringent safety constraints and the use of the very reactive liquid sodium has led to difficult running conditions. Furthermore, the investment costs for a reactor like Superphenix are about 2 times larger than those necessary for a standard PWR with the same power. The cost of electricity which would have been produced by Superphenix in normal running conditions would, also, have been about twice more than that produced by a PWR. Of course, it can be argued that, for industrial series of reactors, the investment cost as well as the fuel cost would have decreased, and that such costs are similar to those of much advocated power from renewable energies like wind.

In conclusion, while the interest of breeding reactors is clear in the hypothesis of an extension of nuclear power, one cannot consider that the present type of sodium cooled reactors should be the only solution. In this context it is possible that subcritical reactors could offer an interesting alternative to sodium cooled reactors. They may also facilitate a switch to the thorium breeding cycle which would lead to a much reduced production of Minor Actinides.

3. Principles of reactor operation

3.1. Chain reaction

A neutron created in a medium (which we first consider infinite) with fissile nuclei gives birth to k_∞ second generation neutrons. The total number of neutrons following the apparition of a neutron in the multiplying medium is[8]

$$n_{\text{chain}} = 1 + k_\infty + k_\infty^2 + \cdots + k_\infty^n + \cdots = \frac{1}{1-k_\infty} \quad (14). \tag{1}$$

The total number of neutrons created in the medium per source neutron is simply $k_\infty n_{\text{chain}}$. One defines a neutronic "gain" as the ratio of the total number of neutrons (source + created) to the number of source neutrons. This gain is then $1/(1-k_\infty)$. Since all neutrons are, ultimately, absorbed, the number of absorption reactions is, thus, $n_{\text{reac}} = n_{\text{chain}}$. For finite media one has to replace k_∞ by an effective value of k_{eff}[9] which is less than k_∞ due to neutrons escaping from the system. One should also consider local values k_s dependent on the specific location of the apparition of the initial neutron. If k_{eff} is larger than unity the reaction diverges, i.e. from one initial neutron one obtains a final number of neutrons going to infinity. A controlled divergence allows to start a reactor. When uncontrolled it leads to a criticality accident. When k_{eff} is kept equal to unity one obtains a critical reactor. If k_{eff} is less than unity an incident neutron gives birth to a finite number of secondary neutrons. The medium is said to be multiplying. The multiplication factor is $1/(1-k_{\text{eff}})$.

3.1.1. Expression of k_∞

It is shown in PPNP that k_∞ can be expressed as

$$k_\infty = \langle \nu \rangle \frac{\int\int\int\int \Sigma_f(E,\boldsymbol{r})\varphi(E,\boldsymbol{r})\,\mathrm{d}E\,\mathrm{d}^3r}{\int\int\int\int \Sigma_a(E,\boldsymbol{r})\varphi(E,\boldsymbol{r})\,\mathrm{d}E\,\mathrm{d}^3r} \tag{2}$$

where Σ_f and Σ_a are the fission and absorption macroscopic cross-sections. If we consider a medium involving n nuclei, and use cross-sections averaged over $\boldsymbol{r}$ and E, like in Eq. (2), we can write

$$k_\infty = \frac{\sum_i \nu_i \Sigma_f^{(i)}}{\sum_i \Sigma_a^{(i)}}. \tag{3}$$

Consider the simple case where the medium involves only three types of nuclei, one fissile, one fertile and one capturing. Then,

$$k_\infty = \nu \frac{\Sigma_f^{(\text{fis})}}{\Sigma_a^{(\text{fis})} + \Sigma_a^{(\text{fert})} + \Sigma_a^{(\text{abs})}} = \eta \frac{\Sigma_a^{(\text{fis})}}{\Sigma_a^{(\text{fis})} + \Sigma_a^{(\text{fert})} + \Sigma_a^{(\text{abs})}} \tag{4}$$

where we have used the relation $\eta = \nu\sigma_f/\sigma_a = \nu\Sigma_f/\Sigma_a$, since it is clearly valid when there is only one fissile species.

[8] For $k_\infty < 1$.

[9] See Section 3.1.2 for a more systematic discussion of the different multiplication factors.

3.1.2. Neutron importance

We have already mentioned the difference between k_∞ and k_{eff}. The definition of k_∞ was unambiguous: it was the number of neutrons produced in an homogeneous, infinite medium following the absorption of a neutron. It is, therefore, a property of the medium. For finite reactors, some neutrons escape the medium, so that the effective multiplication coefficient k_{eff} is less than k_∞. For example, in the case of the most simple slab reactor (see Section 3.3.4 of PPNP) the criticality condition became $k_\infty = 1 + \pi^2 D/a^2\Sigma_a$, rather than $k_\infty = 1$. Thus, close to criticality,

$$k_{\text{eff}} = k_\infty - \frac{\pi^2 D}{a^2 \Sigma_a} \tag{5}$$

the criticality being reached when $k_{\text{eff}} = 1$. In an under-critical finite system it seems evident that the progeny of a neutron created at the center will not be the same as that of a neutron created on the boundary. This leads to the concept of a source multiplication factor $k_s(\boldsymbol{r}, E)$, depending on the initial position and energy of the neutron. The progeny of such a neutron created at $(\boldsymbol{r}, E)$ will be $k_s(\boldsymbol{r}, E)/(1 - k_s)(\boldsymbol{r}, E)$. This progeny number is called the importance of the neutron $\phi^\dagger$ which is also the adjoint flux

$$\phi^\dagger(\boldsymbol{r}, E) = \frac{k_s(\boldsymbol{r}, E)}{1 - k_s(\boldsymbol{r}, E)}. \tag{6}$$

For the sake of simplicity, in the following we leave out the explicit dependence of k_s and $\phi^\dagger$ on $(\boldsymbol{r}, E)$, unless necessary. There is, a priori, no reason why the first generation neutrons would be in number k_s. Rather, k_s can be viewed as defined by

$$\frac{k_s}{1 - k_s} = k_1 + k_1 k_2 + \cdots + k_1 k_2 \ldots k_i + \cdots. \tag{7}$$

Fig. 1 shows the result of a Monte-Carlo calculation which shows how the values of k_i change with i.

The number of first generation neutrons k_1 also depends upon the position and energy of the neutron. It is also called (unfortunately) the adjoint flux

$$\varphi^\dagger = k_1. \tag{8}$$

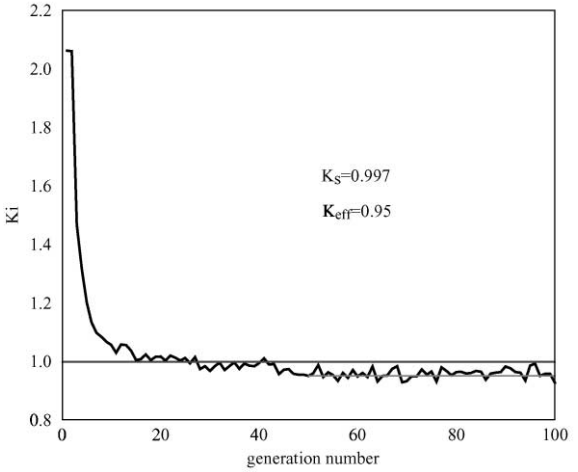

Fig. 1. Evolution of the multiplication factor as a function of the neutron generation number for a model reactor made of a central plutonium sphere with a radius of 4.62 cm surrounded by a plutonium shell with an inner radius of 10 m and a thickness of 1.54 cm. Each single component is characterized by $k_\infty = 0.95$.

The adjoint flux $\phi^\dagger$ (in the first sense) obeys an integral equation, which is particularly instructive in the simple case of the one-group theory (see PPNP, Chapter 3). Finally, one gets

$$\phi^\dagger(\boldsymbol{r}) = \int\int\int \mathrm{d}^3 r \frac{\mathrm{e}^{-\bar{\Sigma}_T|\boldsymbol{r}-\boldsymbol{r}'|}}{|\boldsymbol{r}-\boldsymbol{r}'|^2} \phi^\dagger(\boldsymbol{r}')| \\ (\Sigma_s(\boldsymbol{r}') + \nu\Sigma_f(\boldsymbol{r}')). \tag{9}$$

This equation has exactly the form of the one-group version of the Boltzmann equation (Eq. (3.23) of PPNP) for the normal flux. Thus, the flux and the adjoint flux, or importance, are proportional in the one-group theory. This need not be the case in multi-group theories. Since the kernel of the integral of Eq. (9) is regular in three dimensions, it follows that if $\phi^\dagger(\boldsymbol{r})$ becomes infinite somewhere, it is infinite everywhere. A local criticality ($k_s = 1$) leads to a global criticality ($k_{\text{eff}} \geqslant 1$), since at least neutron chains originating from the corresponding position become infinite. For subcritical systems it is quite possible that $k_s > k_{\text{eff}}$; however, the series of k_i converges towards k_{eff} when $i \to \infty$. Similarly, the i generation neutron density $n_i(\boldsymbol{r})$, although decreasing in magnitude like k_{eff}^i, converges towards the critical distribution $n(\boldsymbol{r})$.

Consider a neutron created in the medium with a probability following the asymptotic density distribution. If absorbed, it produces k_∞ new neutrons. However, in a finite system it only produces $k_{\rm eff}$ neutrons, with some neutrons escaping the system. Thus, $k_{\rm eff} = P_{\rm cap} k_\infty$. The escape probability is $P_{\rm esc} = 1 - k_{\rm eff}/k_\infty$. If we consider a system with N_0 injected neutrons and multiplication $k_{\rm eff}$, the number of escaping neutrons will be $N_0/(1 - k_{\rm eff})(k_\infty - k_{\rm eff})/k_\infty$.

In most cases, in this article, we have not made the difference between $k_{\rm s}$ and $k_{\rm eff}$.

3.2. Subcritical systems

We have seen that, if the neutron multiplication factor of a reactor assembly $k_{\rm eff} < 1$, the chain reaction cannot be sustained. However, if one introduces a source of neutrons inside the multiplying medium, the initial neutron number is multiplied by a factor which can be very large. Since neutrons are produced by fissions, a large energy could be produced with a subcritical system, provided a neutron source is available. In subcritical reactors, these external neutrons are provided by the interaction of accelerated charged particles with matter. The most widely proposed systems use high energy protons. The nuclear reaction of high energy protons with nuclei is called the spallation process. A few other propositions resorted to electrons or deuterons as well as muons as originators of neutron producing reactions (see Section 4.2 of PPNP and references therein).

3.2.1. The neutron source

Following an initial proton–nucleus interaction, a particle cascade is generated. These cascades are simulated by transport codes, which are all built on the same scheme. Nuclear reactions induced by particles with energy above a specified value (at the moment 20 MeV for neutrons) are treated by Intranuclear Cascade (INC) modules.[10] Neutrons below the cut-off are, then, followed with specific neutron Monte-Carlo transport codes like MCNP [25], MORSE [26], Tripoli [27] or MC2 [28]. In principle, the calculation can proceed until all neutrons have been absorbed or have escaped the medium, whatever its properties. However, in the case of a neutron multiplying medium, it is much more efficient to distinguish between source neutrons and secondary neutrons produced by the multiplication. Then if N_0 is the number of primary neutrons following, for example, interaction of a proton with a target surrounded by a multiplying medium and characterized by a multiplication factor k,[11] the total number of created neutrons, after multiplication, is

$$\frac{N_0}{(1-k)}. \tag{10}$$

The number of secondary neutrons (produced after at least one multiplication) is

$$\frac{kN_0}{(1-k)}. \tag{11}$$

The distinction between source and secondary neutrons is by no means trivial. It is relatively easy if the source and multiplying media are distinct. In this case one could define the source neutrons as those coming out from the source medium and penetrating into the multiplying medium. However, even for this simple case, neutrons can originate in the multiplying medium, penetrate the source medium and be scattered back into the multiplying medium. Furthermore, high energy neutrons penetrating the multiplying medium have different multiplication properties than "average secondary neutrons": for example they can produce more (n, xn) reactions, and, in case of fission, lead to a fission with higher than average neutron multiplicity. In practice, it is fortunate that spallation (evaporation) neutrons have energy spectra which are close to fission neutron spectra. Thus, in most cases, one ceases following neutrons which are born with an energy below a cut-off value $E_{\rm cut}$, considered high with respect to fission

[10] These incorporate a high energy part like the Bertini [23] cascade, an evaporation part like the Dresner EVAP code [24], and, possibly a preequilibrium part.

[11] In principle, k depends on the properties of the source neutron (energy, spatial distribution) and on the geometry of the reactor and is equal to $k_{\rm s}$("k source"). For simplicity, we keep the notation k at this stage.

and evaporation neutrons (typically 5–20 MeV). Such neutrons with energy $E < E_{cut}$ are then considered as the source neutrons and kept as inputs for calculations with the neutron propagation codes. This procedure allows a very convenient possibility to decouple the study of the multiplying medium from that of the neutron source. However, this operational definition of source neutrons makes the comparison with experiment difficult. Indeed, experimentally [29] one usually measures the number of neutrons coming out from a thick target.

Most simulation codes account reasonably well for spallation neutron multiplicities. However, the traditional approach which combines the Bertini INC code [23] with the Dresner [24] evaporation code for the high energy part of the cascade and the MCNP [25] or MORSE [26] codes for neutrons below 20 MeV, has serious failures, especially for the prediction of the neutron energy spectra and angular distributions and the residual nuclei mass and charge distributions. Significant improvement is obtained when either the ISABEL [30,31] or the Cugnon [32] codes are used, especially in combination with the relatively new GSI evaporation code [33]. The extension of the MCNP [34] type calculations up to 150 MeV, which is being carried out at Los Alamos and Bruyères le Chatel is also a very significant improvement. In this respect a large amount of work, both experimental and calculational, has to be made for the completion of the evaluated data file for neutrons and protons between 20 and 150 MeV.

3.2.2. Energy gain

The goal of subcritical reactors is to produce energy as well as a neutron excess which could be used for nuclear waste transmutation. It is, consequently, important to evaluate to what extent the energy produced by fission in the multiplying medium exceeds the energy of the primary particle beam.

As seen from Eq. (11), the number of secondary neutrons is $kN_0/(1-k)$. Each of these neutrons is produced by a fission (we neglect (n, xn) reactions), which, itself, produces ν neutrons. Thus, the number of secondary fissions in the system is $kN_0/\nu(1-k)$. Since each fission produces about 0.2 GeV energy,[12] the thermal energy produced in the medium will be $0.2kN_0/\nu(1-k)$. This energy has to be compared to the energy of the incident protons E_p to define an energy gain of the system:

$$G = \frac{0.2kN_0}{\nu(1-k)E_p} = \frac{G_0 k}{1-k}. \tag{12}$$

The CERN FEAT experiment [35] gives a constant value of $G_0 = 3.3$,[13] for incident proton energies larger than 1 GeV and for a uranium target. The experiment consisted in mapping out the number of fissions produced in a multiplying array surrounding a uranium target bombarded by the CERN PS proton beam. The multiplying array consisted of natural uranium bars immersed in a light water swimming pool. The fission density within the uranium bars was obtained from the measurements, after careful corrections for the flux depression within the bars and influence of the surroundings of the detectors. The value of k was deduced from a measurement using a known ^{252}Cf source. From the value of G_0 it is possible to deduce a value of $N_0 = C_0 \nu E_p/0.2 = 41$ neutrons per GeV proton. The ratio of neutron multiplicities for uranium and lead amounted to 1.35, to be compared to the value of 1.4 corresponding to the multiplication in uranium. Another important result of the FEAT experiment was that the neutron multiplicity per GeV saturated for proton energies above 0.8 GeV. For lead and 1 GeV protons the value of $G_0 = 2.65$, was retained by the CERN group for its calculations of the Energy Amplifier [36].

The proton beam is produced with a finite energy efficiency, which is the product of the thermodynamic efficiency for producing electricity from heat (typically 40% in foreseen reactors[14])

[12] This energy corresponds to the total of the kinetic energy of the fission fragments, of their prompt deexcitation (neutrons and photons), and of the β radioactivity energy, which amounts to approximately 15 MeV.

[13] Taking into account a value $k = 0.9$.

[14] Thermodynamic efficiency of present PWRs is around 0.33, while that of gas combined cycle turbines reach 0.5. The lead (and sodium) cooled reactors allow one to reach efficiencies of 0.4. High Temperature Gas Reactors would allow one to reach even higher efficiencies of 0.5.

and the acceleration efficiency. For high current intensity accelerators, most of the power is used for the high frequency cavities, at variance with low intensity accelerators where most of the power is spent in the magnetic devices. High intensities, therefore, are expected to allow 40% efficiencies. Finally, the total efficiency for proton acceleration is expected to be in the vicinity of 0.16. This leads to a minimum value of the multiplication factor for obtaining a positive energy production (ignition), $k_m = 0.68$. For a value $k = 0.98$ one gets a net energy gain of 16.

3.2.3. Neutron balance

Aside from energy production, it is important to evaluate the potential of subcritical reactors for transmutation, i.e. to what extent they produce excess neutrons. A standard reactor can be viewed as a device producing energy and neutrons, both primarily by fission. Fission releases about 200 MeV and 2.5 neutrons. It follows that one may say that 80 MeV are needed to produce one neutron. The spallation process requires only 30 MeV to produce one neutron. Should 200 MeV of fission energy be available for proton acceleration one would, then, get more than 9 neutrons per fission (2.5 fission neutrons and 6.5 spallation neutrons)! True enough one would not produce any more usable energy. Taking into account an acceleration efficiency of 0.4 and an electricity production efficiency of 0.4, one finds that about 6 GeV of fission energy are needed to accelerate a proton to 1 GeV. It would, then, be possible to obtain about 3.5 neutrons per fission in the ignition condition. In general, using an accelerator allows one to increase the number of neutrons available for transmutation at the expense of usable energy.

It is interesting to see if, as far as neutron availability is concerned, subcritical reactors are more or less efficient than the association of a critical reactor and an accelerator. The number of neutrons produced in the subcritical reactor is

$$N = \frac{N_0}{1-k}. \tag{13}$$

while the number of fissions is

$$N_F = \frac{N_0 k}{v(1-k)}. \tag{14}$$

On the average a fission is produced by $(\sigma_F + \sigma_c)/\sigma_F$ neutrons. The total number of neutrons needed for producing N_F fissions is

$$N_{nf} = N_F \frac{\sigma_F + \sigma_C}{\sigma_F} = N_F(1+\alpha) = \frac{N_0 k}{\eta(1-k)} \tag{15}$$

where η is the number of neutrons following capture of an initial neutron by a fissile nucleus. The total number of neutrons available for transmutation is, therefore

$$N_{Dhyb} = N - N_{nf} = \frac{N_0}{1-k}\left(1 - \frac{k}{\eta}\right). \tag{16}$$

We now consider a critical reactor, and, independently, an accelerator. N_{Dr} is the number of neutrons available when using a reactor producing N_F fissions, in addition to the N_0 spallation neutrons. The number of neutrons necessary per fission is

$$\frac{\sigma_F + \sigma_c}{\sigma_F} = 1 + \alpha \tag{17}$$

while the number of neutrons produced per fission is v. It follows that the number of neutrons available per fission is $v - 1 - \alpha$. The total number of neutrons available in the reactor is, then

$$N_{Df} - N_F(v - 1 - \alpha) = \frac{N_0 k}{v(1-k)}(v - 1 - \alpha) \tag{18}$$

and the total number of neutrons available for the system reactor + accelerator is

$$N_{Dr} = N_0\left(1 + \frac{k}{v(1-k)}(v - 1 - \alpha)\right) = \frac{N_0}{1-k}\left(1 - \frac{k}{\eta}\right). \tag{19}$$

Thus,

$$N_{Dhyb} = N_{Dr}. \tag{20}$$

It follows that the choice of a specific value of k is irrelevant as far as the transmutation capabilities are concerned. Whatever the method of coupling between the fission reactor and the accelerator, the number of available neutrons is

$$N_D = N_0 + N_F(v - 1 - \alpha). \tag{21}$$

From the preceding, it is seen that using 10% of the available energy allows one to obtain about 0.1 additional neutrons per fission. Although small, this number has to be compared to the number of neutrons which are effectively available in critical reactors. We know that the maximum number of available neutrons per fission amounts $\nu - 1 - \alpha$. In practice, the real number is smaller than this value due to captures in structural materials and to transmutations of fertile nuclei. Let the number of such captured neutrons to ν_c. The number of available neutrons is, then, $\nu - 1 - \alpha - \nu_c$. Captures in structural materials and losses cannot be much less than 0.2 neutrons per fission, so much so that it is necessary to compensate for reactivity changes by using consumable neutronic poisons. For each fissioning nucleus α such nuclei suffer neutron capture leading, in general, to a fertile nucleus. If one requires regeneration of the nuclear fuel, one sees that $\nu_c = 0.2 + 1 + \alpha$ at least. The number of available neutrons amounts to $\nu - 2(1 + \alpha) - 0.2$. We consider four cases:

1. The thermal ^{238}U–^{239}Pu system. Then, $\nu = 2.871$, $\alpha = 0.36$. The number of available neutrons is $2.871 - 2 \times 1.36 - 0.2 = -0.05$. Regeneration is not possible and no neutron is available for transmutation. The 0.1 additional neutrons made available by the use of an accelerator would allow regeneration.
2. The thermal ^{232}Th–^{233}U system. In this case $\nu = 2.492$, $\alpha = 0.09$. The number of available neutrons becomes $2.492 - 2 \times 1.09 - 0.2 = 0.11$. Regeneration is possible and 0.1 neutrons are available for transmutation. The additional number of neutrons given by the accelerator is significant.
3. The fast ^{238}U–^{239}Pu system. In this case $\nu = 2.98$, $\alpha = 0.14$. The number of available neutrons becomes $2.98 - 2 \times 1.14 - 0.2 = 0.5$. Regeneration is easy. The advantage of an accelerator is not compelling.
4. The fast ^{232}Th–^{233}U system. In this case $\nu = 2.492$, $\alpha = 0.093$. The number of available neutrons becomes $2.492 - 2 \times 1.093 - 0.2 = 0.10$. Regeneration is possible. The additional number of neutrons given by an accelerator is significant.

3.3. Fuel evolution in subcritical reactors

Since subcritical reactors would lose some of their appeal if requiring control rods, it is important to check that the reactor cannot become critical at any time. We address this question in the present section, having in mind, especially, the possible evolution of the fuel.

3.3.1. Evolution equations

During irradiation the nuclear fuel evolves due to several processes, the most significant being:

- fission of heavy nuclides;
- β or α disintegrations;
- Transformation of fertile nuclei into fissile nuclei due to neutron captures followed by radioactive decay;
- production of fission fragments which may act as neutronic poisons.

In general, the evolution of the nuclear fuel is followed by solving the Bateman equations which read, for neutron fluxes independent of time:

$$\frac{\mathrm{d}n_i(t)}{\mathrm{d}t} = -\left(\sigma_i^{\mathrm{T}}\varphi + \sum_j \lambda_{i,j}\right) n_i + \sum_{j \neq i} (\sigma_{j,i}^{a}\varphi + \lambda_{j,i}) n_j \tag{22}$$

where n_i is the number of nuclei of type i per unit volume. $\lambda_{i,j}$ is the decay constant of nucleus i to nucleus j, $\sigma_{i,j}^{\alpha}$ is the capture cross-section of nucleus i resulting in nucleus j, σ_i^{T}, the total cross-section of nucleus i is the sum of the capture and fission cross-sections. These equation are summarized in the vector–matrix form:

$$\frac{\mathrm{d}\boldsymbol{n}}{\mathrm{d}t} = \bar{\bar{A}}\boldsymbol{n} \tag{23}$$

where the elements of the matrix $\bar{\bar{A}}$ are deduced from Eq. (22).

In order to stress the main trends of the evolution of the nuclear fuel we consider a model where only three types of nuclei are present:

1. the fertile nuclei (cap);
2. the fissile nuclei (fis);
3. the fission products (pf).

The fuel is replenished in fertile nuclei at a rate $S(t)$.

Absorption cross-sections are denoted $\sigma^{(a)}$, and fission cross-section $\sigma^{(f)}$.

The evolution of the nuclei is given by the system:

$$\frac{dn_{cap}}{dt} = -n_{cap}\sigma_{cap}^{(a)}\varphi + S(t) \tag{24}$$

$$\frac{dn_{fis}}{dt} = n_{cap}\sigma_{cap}^{(a)}\varphi - n_{fis}\sigma_{fis}^{(a)}\varphi \tag{25}$$

$$\frac{dn_{pf}}{dt} = n_{fis}\sigma_{fis}^{(f)}\varphi. \tag{26}$$

Here φ is the neutron flux.

In order to discuss the dominant features of the fuel evolution we shall make the simplifying assumption that the amount of fertile nuclei is kept constant. This assumption is approximately valid as long as the characteristic evolution time of the fissile part is much shorter than that of the fertile part, i.e. $\sigma_{fis}^{(a)} \gg \sigma_{cap}^{(a)}$.[15] Then,

$$\frac{dn_{cap}}{dt} = 0 \tag{27}$$

and the number of fissile nuclei is obtained:[16]

$$n_{fis}(t) = \frac{1}{\sigma_{fis}^{(a)}}(n_{cap}\sigma_{cap}^{(a)}(1 - \exp(-\sigma_{fis}^{(a)}\varphi t)) + n_{fis}(0)\sigma_{fis}^{(a)}\exp(-\sigma_{fis}^{(a)}\varphi t)). \tag{28}$$

The term $n_{cap}\sigma_{cap}^{(a)}(1 - \exp(-\sigma_{fis}^{(a)}\varphi t))$ of the equation expresses the rise of $n_{fis}(t)$ due to the conversion of fertile nuclei into fissile ones. The term $n_{fis}(0)\sigma_{fis}^{(a)}\exp(-\sigma_{fis}^{(a)}\varphi t)$ corresponds to the disappearance, by fission, of the fissile nuclei present at the initial time. It appears that $n_{fis}(t)$ tends towards an equilibrium value $n_{fis}^{(equ)} = n_{cap}\sigma_{cap}^{(a)}/\sigma_{fis}^{(a)}$ at large times.

- If $n_{fis}(0) < n_{fis}^{(equ)}$ the amount of fissile nuclei will increase with time, so that the reactor is of the breeder type.
- Inversely, if $n_{fis}(0) > n_{fis}^{(equ)}$ the reactor will be an incinerator.
- It is important to note that a subcritical reactor can always be a breeder, contrary to critical reactors.

The evolution of the number of fission fragments is given by

$$n_{pf}(t) = tn_{cap}\sigma_{cap}^{(a)}\varphi + n_{cap}\frac{\sigma_{cap}^{(a)}}{\sigma_{fis}^{(a)}}(\exp(-\sigma_{fis}^{(a)}\varphi t) - 1) + n_{fis}(0)(1 - \exp(-\sigma_{fis}^{(a)}\varphi t)). \tag{29}$$

Here the first term corresponds to the linear consuming of fertile nuclei, the second term to the building up of the fissile nuclei from the fertile ones, and the last term to the disappearance of the initial load of fissile nuclei. For large times, the first term dominates.

Knowing the evolution of the concentrations, one gets the evolution of the multiplication factor[17]

$$k_\infty(t) = \frac{\eta n_{fis}(t)\sigma_{fis}^{(a)}}{n_{cap}\sigma_{cap}^{(a)} + n_{fis}(t)\sigma_{fis}^{(a)} + n_{pf}(t)\sigma_{pf}^{(a)} + P(t)} \tag{30}$$

where $P(t)$ is the number of neutrons lost in structural materials or control rods or by escaping the reactor. In critical reactors the condition $k = 1$ is kept via modulation of $P(t)$. For subcritical reactors the value of k is allowed to evolve within prescribed limits around a nominal value provided it remains sufficiently smaller than unity. This result may be obtained by periodical regeneration of the fuel as well as by defining working conditions between two regeneration events that minimize the variations of k_∞. These conditions are realized differently in systems using liquid fuels and in those using solid fuels.

[15] This condition is fulfilled for both fast and thermal reactors. However, if a reactor could be made to work in the resonance region, it might be incorrect. In this case, the fertile part would, progressively, disappear in favour of the fissile part. Such system would lead to a very high breeding ratio.

[16] Here, we neglect the effect of radiative captures in fissile nuclei except for the difference between $\eta = \nu\sigma_{fis}^{(f)}/\sigma_{fis}^{(a)}$ and ν. Thus, in the evolution equation $\sigma_{fis}^{(f)} = \sigma_{fis}^{(a)}$.

[17] In the following considerations the values of k_∞ may be significantly larger than unity. In such cases additional neutron absorbers or leakage have to assure the subcriticality.

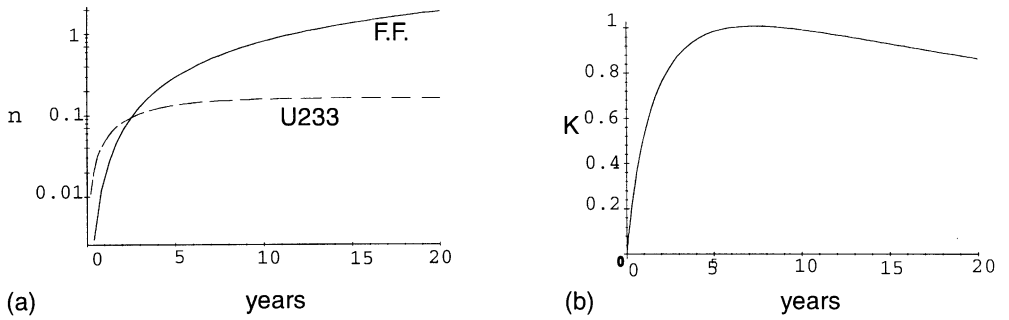

Fig. 2. Model fuel evolution in a Th–U hybrid system. The fast neutron flux is 4×10^{15} n/cm^2 s^{-1}. The evolution of the concentrations of ^{233}U and fission fragments (FF) with respect to ^{232}Th are shown in (a), the evolution of k_∞, in (b)

3.3.2. Liquid fuel systems

In this case fission products are, generally, extracted from the fuel soon after they are produced. It is also possible to keep the concentration of fertile elements constant by continuous feeding. The relative proportion of fissile and fertile nuclei evolves towards an equilibrium:

$$\frac{dn_{fis}}{dt} = n_{cap}\sigma_{cap}^{(a)}\varphi - n_{fis}\sigma_{fis}^{(a)}\varphi = 0, \tag{31}$$

$$\frac{n_{fis}^{(eq)}}{n_{cap}^{(eq)}} = \frac{\sigma_{cap}^{(a)}}{\sigma_{fis}^{(a)}}. \tag{32}$$

In the case of simple fissile nuclei regeneration, one sees from relations (30) and (32) that the maximum value of k_∞ is equal to $\eta/2$.

Proposed liquid fuels have been molten salts. A reactor using a mixture of uranium, thorium, beryllium and lithium fluorides has worked with success for several years in Oak Ridge [37]. This reactor used the ^{232}Th$-^{233}$U cycle. ^{233}Pa may capture neutrons which tend to decrease the reactivity of the reactor.[18] This is why, in the experimental reactor of Oak Ridge, an on-line processing of the fuel was carried out. This processing aimed at extracting both the fission products and the protactinium. After decay of protactinium, the resulting ^{233}U was re-injected in the reactor. This procedure allowed to reach a breeding of the order of 5% per year. This was the only case where breeding was demonstrated for a thermal reactor.

Using liquid fuels has also been considered for fast reactors. In this case chlorides rather than fluorides have been proposed [38].

3.3.3. Solid fuels

In systems using solid fuels, as small a variation of k_∞ as possible between two refueling events is looked for. From Eqs. (28)–(30) it is seen that the value of $k_\infty(t)$ depends upon the initial concentration of the fissile element. An initial breeding value of this concentration induces an increase of $k_\infty(t)$ with time. This increasing trend may be more or less exactly compensated by the decrease of k_∞ caused by the increase of the concentration of fission products. Rubbia [36] has shown that such a compensation is possible over long periods of time. To illustrate the mechanism of this compensation, we use our simple 3 components model where we choose representative values of the cross-sections for a fast reactor using the thorium cycle. The capture cross-section of the fertile nucleus is taken to be 0.45 b, the fission cross-section of the fissile nucleus to be 2.75 b. The average capture cross-section of fission products is taken to be 0.15 b, according to recent calculations.[19] Starting from a state without fissile component, Fig. 2 shows the evolution of the fissile part and of the fission products part (a), and that of the multiplication factor k_∞(b). The evolution of k_∞ shows

[18] In the case of the uranium–plutonium cycle, effects due to captures in ^{239}Np, analogous to those due to ^{233}Pa in the thorium–uranium cycle, are 10 times smaller.

[19] D. Heuer, private communication.

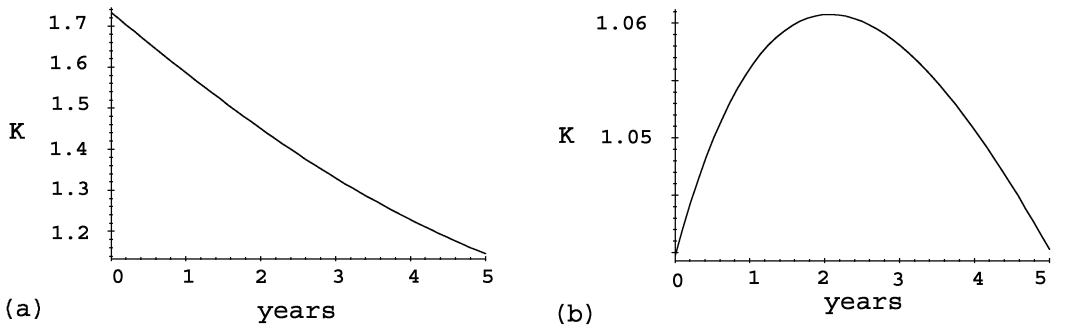

Fig. 3. Evolution of the model Th–U fuel with an initial concentration of 0.5(a) and 0.135(b) for ^{233}U with respect to thorium.

a maximum after about 7 years, starting from zero concentration of ^{233}U. After 3 years the concentration of ^{233}U is close to 0.135. Starting with this concentration one finds that the value of k_∞ is reasonably constant for at least 5 years, as shown in Fig. 3(b). The maximum value of k_∞ of 1.06 shows that the neutron economy for a critical reactor would be difficult since only 6% of the neutrons are available for parasitic captures and leakage. This point will be discussed later, in more realistic terms. Fig. 3(a) shows the evolution of k_∞ when the initial load in the fissile component exceeds noticeably the equilibrium value. Here there is a fast and continuous decrease of the reactivity. This means that solid fuels in subcritical reactors would not be good choices for incinerating without regeneration a nucleus like ^{239}Pu, for example, which is highly fissionable.

Fig. 4(a) and (b) are equivalent to Figs. 2(b) and 3(b), but for a thermal reactor with the same specific power corresponding to a flux of 4×10^{14} n/cm^2 s^{-1}.[20] One sees that, if the neutron economy is slightly improved (higher value of k_∞ at maximum) the stability of k_∞ is only assured for the very short time of less than 1 year. This difference between fast and thermal systems was stressed by Rubbia [36]. Fig. 4(a) also shows that the electro-breeding[21] of ^{233}U is much faster for thermal reactors than for fast reactors. This is a reflection of the fact that the equilibrium concentration of ^{233}U is 7 times smaller for thermal reactors.

3.3.4. Properties of fuels

More complete calculations than those just presented are needed in order to characterize the behavior of specific fuels which might be used in subcritical reactors. These calculations are based on the generalized Bateman evolution equation:

$$\frac{\mathrm{d}n_i(\boldsymbol{r},t)}{\mathrm{d}t} = -\left(\sigma_i^{\mathrm{T}}(\boldsymbol{r},t)\varphi(\boldsymbol{r},t) + \sum_j \lambda_{i,j}\right) n_i(\boldsymbol{r},t) + \sum_{j\neq i} (\sigma_{j,i}^{a}(\boldsymbol{r},t)\varphi(\boldsymbol{r},t) + \lambda_{j,i}) n_j(\boldsymbol{r},t). \tag{33}$$

The cross-sections and fluxes involved in these equations are one energy group quantities defined as energy averages or sums:

$$\sigma_i(\boldsymbol{r},t) = \frac{\int \sigma_i(E)\varphi(E,\boldsymbol{r},t)\,\mathrm{d}E}{\int \varphi(E,\boldsymbol{r},t)\,\mathrm{d}E} \tag{34}$$

$$\varphi(\boldsymbol{r},t) = \int \varphi(E,\boldsymbol{r},t)\,\mathrm{d}E. \tag{35}$$

The time dependence of the neutron flux and thus of the one-group cross-sections, reflects the evolution of the concentrations $n_i(\boldsymbol{r},t)$. Eq. (33) is, in fact, an implicit, non-linear, differential equation which has to be solved numerically. Detailed calculations, either of the deterministic type or, more often, of the Monte-Carlo type, are carried out at successive times close enough for the cross-sections to be considered constant. In these time intervals the evolution of concentrations is

[20] We shall see in the next section that such a high thermal flux may be difficult to accept, due to the Protactinium Effect.

[21] Electro-breeding denotes the process by which the fertile-to-fissile conversion is achieved by the spallation neutrons produced by the accelerator.

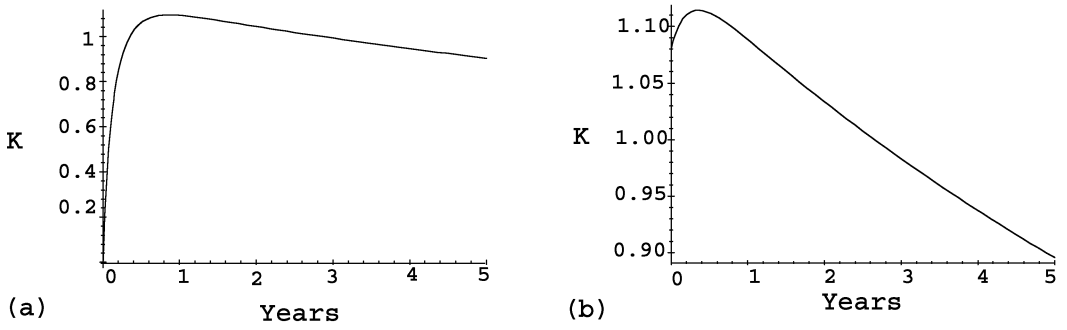

Fig. 4. Variation of k_∞ for a thermal system using the Th–U cycle. (a) starting without ^{233}U present in the system at time 0. (b) starting with an initial concentration of ^{233}U slightly below the equilibrium value.

followed, at each position, according to the Bateman equation with time-independent coefficients. By integrating Eq. (33) over the reactor volume it is possible to obtain an evolution equation for the total number of nuclei of a given species

$$\frac{\mathrm{d}N_i(t)}{\mathrm{d}t} = -\left(\sigma_i^{\mathrm{T}}(t)\varphi(l) + \sum_j \lambda_{i,j}\right) N_i(t) + \sum_{j\neq i} (\sigma^{\mathrm{c}}_{j,\,i}(t)\varphi(t) + \lambda_{j,\,i}) N_j(,\,t) \tag{36}$$

with

$$N_i(t) = \int\int\int n_i(\boldsymbol{r}, t)\,\mathrm{d}^3\boldsymbol{r} \tag{37}$$

$$\varphi(t) = \frac{\int\int\int \varphi(\boldsymbol{r}, t) n_i(\boldsymbol{r}, t)\,\mathrm{d}^3\boldsymbol{r}}{\int\int\int n_i(\boldsymbol{r}, t)\,\mathrm{d}^3\boldsymbol{r}} \tag{38}$$

$$\sigma(t) = \frac{\int\int\int \sigma(\boldsymbol{r}, t)\varphi(\boldsymbol{r}, t) n_i(\boldsymbol{r}, t)\,\mathrm{d}^3\boldsymbol{r}}{\int\int\int \varphi(\boldsymbol{r}, t) n_i(\boldsymbol{r}, t)\,\mathrm{d}^3\boldsymbol{r}}. \tag{39}$$

It is legitimate to simplify the treatment by assuming a time independent neutron flux since this can be obtained by a modulation of the accelerator intensity. The time dependence of the cross-sections is, usually, found to be relatively modest and can be neglected if qualitative discussions like these given below are considered sufficient. On the other hand, as shown in Table 4, very strong dependence of the cross-sections upon the composition of the fuel and upon the reactor geometry require exact calculations. Table 4 compares important cross-sections as obtained in three cases:

1. The Super Phenix fuel composition (U–Pu) and geometry (sodium coolant).
2. A lead cooled reactor (50% lead in volume) with Minor Actinide oxide fuel.
3. A lead cooled reactor (50% lead in volume) with Minor Actinide metal fuel.

In case 1, it is assumed that the presence of MA does not affect the energy dependence of the neutron flux. In cases 2 and 3 the neutron flux is obtained from the Monte-Carlo calculation with the initial MA load.

The neutron spectra are successively harder for cases 1–3. The presence of sodium in case 1, and oxygen in both cases 1 and 2 is responsible for the softening of the spectra. It is also found that the nature of the fissile and fertile components have a strong influence on the hardness of the neutron spectrum. The strong absorption cross-sections of minor actinides at low neutron energies lead to flux depression at low energies and, therefore, to hard spectra.

With the simplifying assumption of time-independent cross-sections, Fig. 5 compares the evolution of k_∞ for a fuel consisting of a plutonium mixture (^{238}Pu(2.5%), ^{239}Pu(60.8%), ^{240}Pu(24.9%), ^{241}Pu(11.7%)) originating from PWR used fuel and for a fuel consisting of a ^{232}Th(90%)$-^{233}$U(10%) mixture. In both cases the shape of the neutron flux was chosen to be that of Super-Phenix.

Table 4
Comparison of mono-group fission and capture cross-sections for three different fast neutron spectra: (1) Super Phenix spectrum, (2) Minor Actinides oxide fuel in lead coolant, (3) Minor Actinides metallic fuel in lead coolant

Z	A	fis.SPX	fis. ox.	fis. met.	cap.SPX	cap. ox.	cap. met.
90	232	0.0104	0.0125	0.0178	0.4240	0.2441	0.2097
92	233	2.9120	2.3077	2.2249	0.2790	0.2013	0.1772
92	234	0.3300	0.4104	0.5205	0.6700	0.3951	0.3610
92	235	2.0150	1.5325	1.4366	0.6190	0.3562	0.2958
92	236	0.1040	0.1243	0.1701	0.6020	0.2855	0.2525
92	238	0.0427	0.0530	0.0746	0.3030	0.2068	0.1804
93	237	0.3077	0.4026	0.5188	1.6540	1.0131	0.8097
94	238	1.0630	1.1350	1.2592	0.5650	0.4909	0.4222
94	239	1.8230	1.6368	1.6713	0.5710	0.2627	0.2195
94	240	0.3570	0.4505	0.5620	0.5670	0.3521	0.3601
94	241	2.4920	2.0462	1.9443	0.4670	0.2630	0.2280
94	242	0.2360	0.3269	0.4272	0.4450	0.3165	0.3780
94	243	0.8630	0.6806	0.7220	0.4070	0.2289	0.1895
94	244	0.2140	0.2760	0.3633	0.2540	0.0945	0.0808
95	241	0.2750	0.3388	0.4528	2.1690	1.1062	0.9011
95	242	3.2300	3.0218	2.8838	0.4940	0.1962	0.1531
95	243	0.2010	0.2660	0.3611	1.7660	0.9440	0.7316
96	242	0.5620	0.2041	0.2819	0.5670	0.1529	0.1171
96	243	3.2880	2.1123	2.1288	0.2440	0.1385	0.1142
96	244	0.4180	0.5126	0.6504	0.6190	0.5802	0.4650
96	245	2.7570	1.8327	1.7246	0.3460	0.2168	0.1745

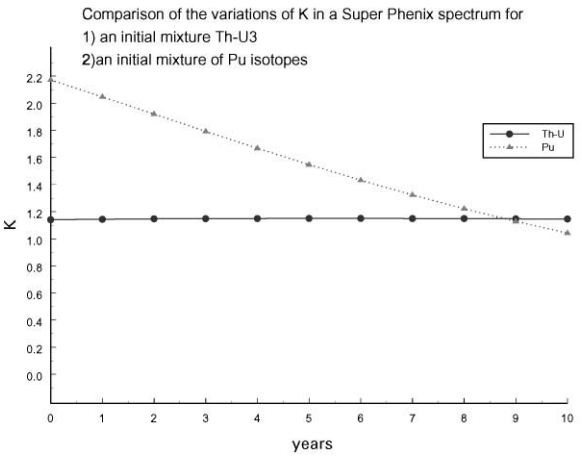

Fig. 5. Variations of k_∞ for: (1) a mixture of ^{232}Th(90%)–^{233}U(10%), (2) a mixture of ^{238}Pu(2.5%), ^{239}Pu(60.8%), ^{240}Pu(24.9%), ^{241}Pu(11.7%), as considered by Rubbia et al. [40]. The flux assumed in the calculations was 4×10^{15} n/cm^2 s^{-1}.

The flux assumed in the calculations was 4×10^{15} n/cm^2 s^{-1}. Fig. 5 shows that the reactivity decreases strongly and continuously in the case of a plutonium fuel. This means that large compensation for reactivity changes should be provided, and, consequently, that reactivity accidents related to wrong manipulation of the reactivity compensating devices could become possible, obliterating much of the appeal of subcritical reactors in that case. The plutonium mixture has a fissile behavior. On the other hand, the mixture ^{232}Th$-^{233}$U has a rather constant reactivity as first noted by Rubbia et al. [40]. Fig. 6 compares the evolution of k_∞ for a mixture of minor actinides (^{237}Np(33.3%), ^{241}Am(21.6%), ^{243}Am(40%), ^{242}Cm(2.1%), ^{243}Cm(0.032%), ^{244}Cm (1.4%), ^{245}Cm(0.9%)) in the three neutron spectra discussed above, and for a thermal PWR spectrum. The fast neutron flux was chosen to be 4×10^{15} n/cm^2 s^{-1} and the thermal flux to be 3×10^{14} n/cm^2 s^{-1}, so that fission densities were, roughly, the same in the fast and thermal cases. Under these conditions it was found that the lifetimes of the minor actinides in the flux were similar in all cases: the inventory of minor actinides decrease by a factor of 2 after 10 years of irradiation.

The minor actinide fuels behave like a mixture of fissile and fertile nuclei. By adding a small admixture of a fissile mix like industrial plutonium, a rather constant value of k_∞ can be obtained for all fast spectrum cases. Fig. 6 shows that the MA mix behaves like a strong neutron poison for thermal neutrons. In fast neutron flux the initial reactivity of the MA mix depends markedly on the hardness of the spectrum. In this respect metallic fuels have a clear advantage and are the only possibility to obtain criticality for the initial load. Fig. 7 shows the evolution of the inventory of the different minor actinides and of the fission products. The americium and neptunium isotopes decrease steadily, behaving like fertile parts, while the more fissile plutonium and curium isotopes go through a maximum. It appears that the stabilization of the variation of k_∞ is chiefly due to the formation of ^{238}Pu, which has a high fast-neutron fission probability. It is formed by neutron capture up ^{237}Np, which behaves like a fertile species. To a lesser extent the rise of ^{244}Cm fissions counteracts the decrease of the ^{241}Am and ^{243}Am fissions. Fig. 7 also gives an idea of the time required for a significant decrease (about 10 years) of the total number of transplutonium nuclei.

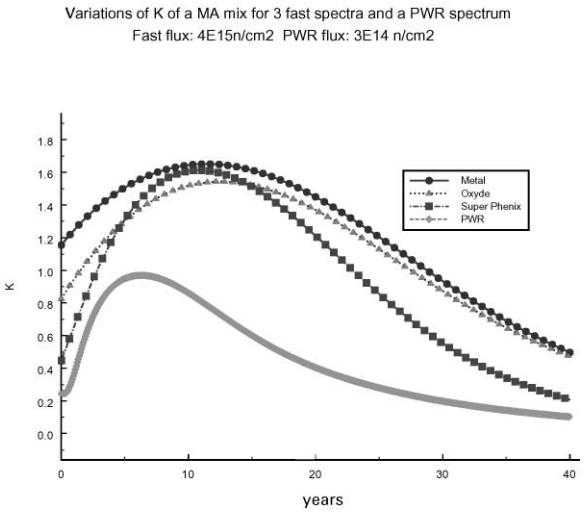

Fig. 6. Evolution of k_∞ for a mixture of Minor Actinides (^{237}Np(33.3%), ^{241}Am(21.6%), ^{243}Am(40%), ^{242}Cm(2.1%), ^{243}Cm(0.032%), ^{244}Cm(1.4%), ^{245}Cm(0.9%) [40]) in four different neutron fluxes: (1) similar to Super Phenix, (2) calculated for the minor actinide oxide fuel, (3) calculated for the minor actinide metal fuel, (4) in a PWR spectrum. In the first three cases the fast neutron flux was 4×10^{15} n/cm^2 s^{-1}. In the last case the thermal neutron flux was 3×10^{14} n/cm^2 s^{-1}.

3.4. Reactivity excursions in subcritical reactors

Short-term fuel evolution as well as temperature changes may lead to reactivity changes. For thermal reactors the very large capture cross-section of ^{135}Xe and ^{149}Sm lead to such effects. In the case of the thorium–uranium cycle, a specific effect arises, both for thermal and fast reactors, due to the 27-day half-life of ^{233}Pa. We first examine this effect.

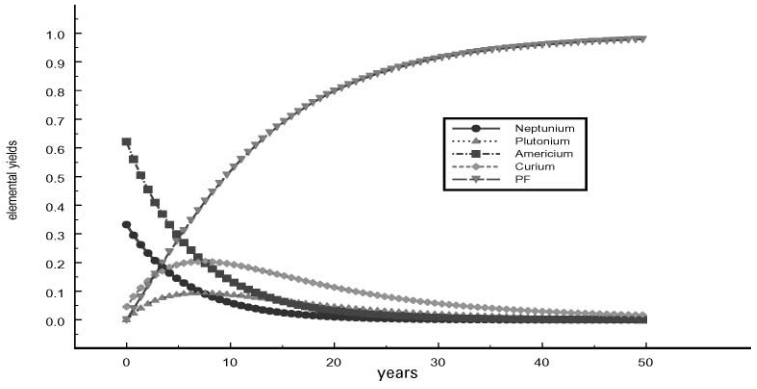

Fig. 7. Evolution of the inventories of minor actinides and fission products, for the minor actinide fuel, as a function of time. The neutron flux assumed was 4×10^{15} n/cm^2 s^{-1}.

3.4.1. Protactinium effect [36]

^{233}U is formed by neutron capture of ^{232}Th followed by two β decays:

$$^{232}\mathrm{Th} + n \rightarrow {}^{233}\mathrm{Th} \underset{22.3\ \mathrm{min}}{\overset{\beta^-}{\rightarrow}} {}^{233}\mathrm{Pa} \underset{26.97\ \mathrm{d}}{\overset{\beta^-}{\rightarrow}} {}^{233}\mathrm{U}. \quad (40)$$

The presence of protactinium imposes limits on the admissible neutron flux when using solid fuels. This limitation is due to two detrimental effects of protactinium:

1. Protactinium captures neutrons but has a small fission probability, and thus decreases the reactivity of the reactor.

2. After a reactor stop, the ^{233}Pa inventory transforms into ^{233}U, which leads to an increase of the reactivity and of k. This increase may lead to criticality of the reactor. The characteristic time for such an evolution is of the order of the lifetime of ^{233}Pa, i.e. about 1 month. Corrective actions could be easily taken by insertion of a negative reactivity. However, the advantage of passive safety of subcritical systems would be lost. It is thus of interest to keep the system subcritical under all circumstances.

In Section 5.2 of PPNP it is shown that, during the neutron irradiation, k_∞ is decreased by

$$\frac{\Delta k}{k} = -\frac{\sigma_{\mathrm{Pa}}^{(\alpha)} \nu_c (1+\alpha)}{\lambda n_{\mathrm{Th}} \sigma_{\mathrm{Th}}^{(a)}} \quad (41)$$

where ν_c is the fissions density, λ the decay constant of ^{233}Pa, n_{Th} the density of thorium nuclei, $\alpha = \sigma_{\mathrm{U3}}^{(c)}/\sigma_{\mathrm{U3}}^{(a)}$. For thermal reactors the ratio $\sigma_{\mathrm{Pa}}^{(a)}/\sigma_{\mathrm{Th}}^{(a)} = 7.4$ while $\sigma_{\mathrm{Pa}}^{(a)}/\sigma_{\mathrm{Th}}^{(a)} = 2.4$ for fast reactors. It follows that if one accepts a given decrease of k_∞, fast reactors allow specific powers 3 times larger than thermal reactors, and hence 3 times more compact cores.[22]

After a reactor stop the protactinium will decay into ^{233}U, leading to an increase to k_∞. In Section 5.2 of PPNP, it is also shown that the increase of k_∞ is given by

$$\frac{\Delta k_\infty}{k_\infty} = \frac{\nu_c (1+\alpha)}{\lambda n_{\mathrm{Th}} \sigma_{\mathrm{Th}}^{(a)}} (0.5\sigma_{\mathrm{U}}^{(a)} + \sigma_{\mathrm{Pa}}^{(a)}). \quad (42)$$

For fast reactors we get

$$\frac{\Delta k_\infty}{k_\infty} = 1.8 \times 10^7 \frac{\nu_c (1+\alpha)}{n_{\mathrm{Th}}}$$

and, for thermal reactors

$$\frac{\Delta k_\infty}{k_\infty} = 1.6 \times 10^8 \frac{\nu_c (1+\alpha)}{n_{\mathrm{Th}}}.$$

One sees that the limit on the specific power is 10 times more stringent for thermal systems than for fast ones. For $\Delta k_\infty/k_\infty = 2 \times 10^{-2}$ the corresponding capture densities are of order 2.8×10^{13} for fast systems and 2.6×10^{12} for thermal ones. The corresponding fluxes are, then, 4×10^{15} for fast reactors and 4×10^{13} for thermal reactors.

In conclusion, it appears that the protactinium effect favors greatly fast reactors if solid fuels and the thorium–uranium cycle are to be used. This is not true for the uranium–plutonium cycle.

3.4.2. Xenon effect [41]

It is well known that some fission products like ^{135}Xe[23] have very large absorption cross-sections for thermal neutrons. ^{135}Xe is not produced directly by fission but by beta decay of a precursor fission fragment. The decay chain by which it is produced is

$$^{135}\mathrm{I} \underset{6.7\ \mathrm{h}}{\overset{\beta}{\rightarrow}} {}^{135}\mathrm{Xe} \underset{9.2\ \mathrm{h}}{\overset{\beta}{\rightarrow}} {}^{135}\mathrm{Cs} \underset{2.6\times10^6\ \mathrm{y}}{\overset{\beta}{\rightarrow}} {}^{135}\mathrm{Ba\ (stable)}. \quad (43)$$

^{135}Xe has an absorption cross-section of 2.7×10^6 b for thermal (0.025 eV) neutrons. One can find a detailed treatment of the xenon effect in Ref. [17].

Fig. 8, obtained in [17], shows the evolution of the xenon-induced reactivity decrease after shutdown of a thermal reactor at two flux levels: 4×10^{13} and 2×10^{14} n/cm^2 s^{-1}.

The initial xenon concentration is independent of the neutron flux, at least for not too small fluxes. In critical thermal reactors the reactivity decrease prevents restarting of the reactor if a large enough positive reactivity reserve is not available. Hybrid systems can be restarted at any time,

[22] We have assumed that α is the same for fast and thermal reactors. This is not strictly true but does not change significantly the argument.

[23] See Lamarsch [41] for the treatment of the less serious ^{149}Sm effect.

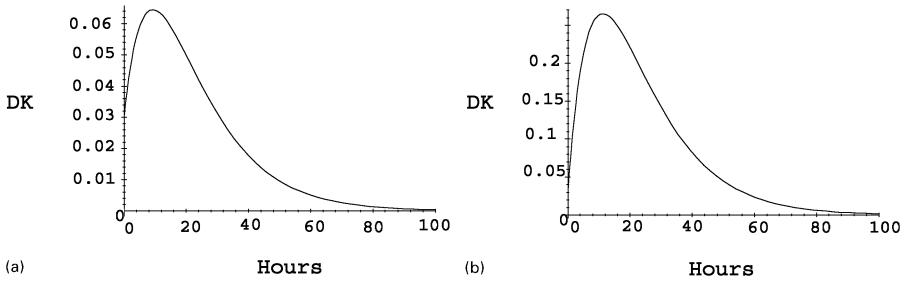

Fig. 8. Variation of the xenon-induced reactivity decrease after reactor shutdown. The thermal neutron flux was: (a) 4×10^{13} and (b) 2×10^{14} n/cm^2 s^{-1}.

although the gain will be smaller if the xenon concentration is high. However, if the reactor is stopped long enough, the xenon concentration vanishes and, thus, the reactivity is larger by 0.0035 than the reactivity in operation. This is true for any thermal reactor, and has to be added to the protactinium related reactivity increase in the case of thorium reactors. For fast reactors the xenon effect is negligible.

3.4.3. Temperature effect

The reactivity of any reactor is generally temperature dependent. Critical reactors have, for obvious safety reasons, a negative reactivity temperature coefficient. For example PWRs have a coefficient between 5×10^{-5} and $10^{-4}/^{\circ}$C [42]. This means that a PWR has a reactivity at zero power between 0.03 and 0.015 higher than at nominal power. The temperature coefficient of fast reactors is usually smaller than that of thermal reactors. For sodium cooled fast reactors it is around $10^{-5}/^{\circ}$C [42]. A similar value has been calculated by Rubbia et al. [36] for their Fast Energy Amplifier.

3.4.4. Impact of reactivity excursions

From the above considerations it is apparent that fast subcritical reactors have more favorable neutronic characteristics than do thermal reactors. Practically, it seems difficult to design a subcritical thermal reactor with k_{eff} larger than 0.95 for the uranium–plutonium cycle and 0.92 for the thorium–uranium cycle. The corresponding values for fast reactors would be 0.99 and 0.98, respectively.

3.5. Safety features in subcritical reactors

One expects that the response of a subcritical reactors to a reactivity injection will be very different from that of critical reactors. A reactivity insertion of more than 1\$[24] in a critical reactor leads to a fast exponential divergence (see Section 3 of PPNP).

$$W(t) = W_0 \exp\left(\frac{\rho_{\mathrm{prompt}}}{\tau_{\mathrm{n}}}\right). \tag{44}$$

The reactivity ρ is equal to $k_{\mathrm{eff}}/(k_{\mathrm{eff}} - 1)$, and τ_{n} is the time between two generations of neutrons, or, equivalently, the neutron lifetime in the medium. For lead cooled fast reactors $\tau_{\mathrm{n}} = 3 \times 10^{-8}$ s [7]. It follows that the power would be multiplied by 100 after $14 \times 10^{-8}/\rho_{\mathrm{prompt}}$ s. Even for $\rho_{\mathrm{prompt}} = 0.001$, i.e. a total reactivity insertion of 0.4% (for a ^{233}U fueled reactor) the power is multiplied by 100 after 0.14 ms! Consider now, the case of a subcritical system with $k = 0.98$, and the same reactivity increase of 0.4%. The energy gain is proportional to $1/(1 - k)$ and increases by 25% only!

The preceding considerations are very schematic. An example of a realistic calculation [36] is displayed in Fig. 9 where a comparison between the behaviors of a critical and a subcritical system is made. In the figure the total reactivity insert is as much as 2.55\$. Temperature reactivity dependence is taken into account. The advantage of subcritical

[24] By definition, in reactor physics 1\$ reactivity insertion corresponds to a reactivity insertion equal to the fraction of delayed neutrons. For ^{235}U it is equal to 0.65%.

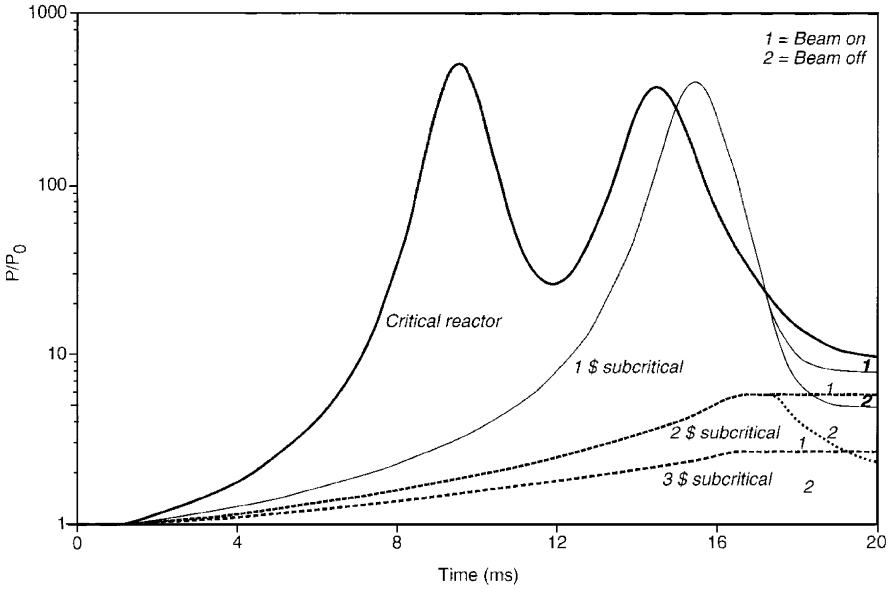

Fig. 9. Comparison of the power increase of a critical reactor and of different subcritical systems after insertion of an additional reactivity. The additional reactivity amounts to an increase rate of 170$/s for 15 ms, after which the reactivity remains constant. Note that hybrid reactors are not supposed to be less than 6$ subcritical. Figure from Ref. [36].

reactors, even with a moderate amount of subcriticality, is quite compelling.

As far as residual heat extraction is concerned, subcritical reactors have essentially the same properties as a critical reactor using the same technology, hence, the potential interest in Lead Cooled, Molten Salt and High Temperature Gas reactors, which are known to be very safe in handling the residual heat.

4. Nuclear waste incineration [18,43–46]

This section examines to which extent subcritical reactors might contribute to solve the nuclear waste problem. We begin by recalling the general context of this question.

4.1. General considerations

As noted in Section 2.1, nuclear energy production is accompanied by production of radioactive wastes of different nature:

- Fission products.
- Activation products obtained from neutron capture by nuclei belonging to the structure of the reactor, such as, for example, cobalt 60.
- Transuranic nuclei obtained from neutron capture by the nuclear fuel.

Nuclear wastes are characterized by their radiotoxicity and their lifetime. Only wastes with lifetimes exceeding about 10 years are considered to be significant storage problems. These are essentially some fission products (LLFP) and transuranic elements. Their noxiousness is, traditionally, measured by their ingestion radiotoxicity.

4.1.1. Radiotoxicities

The ingestion radiotoxicity of an element is a measure of the biological consequences of its ingestion. The radiotoxicity is, then, defined as

$$R(\mathrm{Sv}) = F_{\mathrm{d}}(\mathrm{Sv/Bq})\mathscr{A}(\mathrm{Bq}) \tag{45}$$

where R(Sv) is the radiotoxicity in Sievert per mass unit, F_d(Sv/Bq), is the dose factor in Sievert per Becquerel activity, and A(Bq) is the mass specific activity. For 1 kg mass

$$\mathscr{A}(\mathrm{Bq/kg}) = \frac{1.32}{T_{1/2}(\mathrm{yr})}\frac{10^{19}}{A}. \quad (46)$$

The International Commission of Radiation Protection (ICRP) has evaluated the dose factors [47], some of which can be found in the relevant section of PPNP.

Fission products decay by β radiation, while transuranic elements decay essentially through α radiation. For the same disintegration rate, α emitters are much more radiotoxic than β emitters, with the exception of ^{129}I which has very peculiar biological properties, with a very high affinity for the thyroid gland.

The use of ingestion radiotoxicity as a measure of noxiousness is subject to question. For example, in the case of underground storage, the probability for the radioactive species to enter the biosphere is of paramount importance. Plutonium and, generally, other actinides, have a very low mobility, especially in clay, so that they contribute little to the radiotoxicity released to the biosphere. On the other hand, elements like technetium and iodine are very mobile and are, potentially, the chief contributors to radiotoxological release from deep underground storage.

4.1.2. Waste management options

Two different strategical approaches are proposed for high activity nuclear wastes disposal:

- Direct underground storage of spent fuel elements, without any reprocessing.
- Spent fuel reprocessing with the aim of optimized extraction of transuranics and fission products and their transmutation by nuclear reactions into less radiotoxic or short-lived species. Available nuclear reactions for nuclear waste processing are of two types:

— Transmutation which, by neutron capture, transforms a radioactive nucleus into a stable one. This method is suitable for fission products. As stable nuclei could be, simultaneously, transformed into radioactive ones, the method may require an initial separation of the isotopes to be transmuted. However, ^{99}Tc and ^{129}I do not require such separation.

— Incineration which amounts to nuclear fission following neutron capture. This method is suitable for transuranic elements. It is always associated to energy and neutron production. It is already applied, at an industrial scale, with plutonium.

4.1.3. The plutonium case

From the preceding discussion, plutonium can be considered from two different view points. In the breeding strategy, it is a nuclear fuel. In normal PWRs, it is a nuclear waste which is apt to be incinerated. Incineration is thought to be possible with thermal reactors like PWRs.

At present, plutonium obtained after reprocessing of standard PWR fuel is used, in some countries, for fabrication of MOX fuel, a mix of plutonium and depleted uranium oxides. The MOX fuel elements are used as substitutes of normal enriched uranium fuel elements in PWRs. Note that the presence of ^{238}U in the MOX fuel is needed for safety arguments: it allows one to keep a negative temperature coefficient and partially breed the nuclear fuel so as to prevent too fast a decrease of the reactivity. While being irradiated the plutonium mixture is depleted of the fissile 239 isotope and enriched in the 240 isotope which is not fissile by thermal neutrons but has a large capture cross-section (it is a neutron poison), as well as in transplutonic nuclei (minor actinides, especially americium) which are not or weakly fissile. In the course of the successive reprocessing it is, therefore, necessary to increase the total concentration of plutonium with respect to that of uranium. Such an increase is, however, not possible ad infinitum, since ^{240}Pu is a poison for thermal neutrons, but not for fast neutrons. Thus, a fuel too enriched in plutonium might yield criticality of the reactor for fast neutrons, and a divergence in case of partial or total loss of coolant. After two or three reprocessing cycles in PWRs, a "dirty" plutonium is left with an increased quantity of minor actinides. It has been proposed to incinerate this mix in fast, sodium cooled, reactors. These would incinerate more

plutonium than they would produce, contrary to breeder reactors. However, due to safety considerations, the fuel would include a minimum amount of ^{238}U which would limit the net consumption of plutonium. Fast reactors should also be able to incinerate minor actinides efficiently. Incineration of dirty plutonium and minor actinides would require 1 fast reactor for 4–5 PWRs.

Rather than transforming about one-third of the PWRs into MOX-PWRs, it seems that replacing uniformly the traditional 3.5% ^{235}U-enriched fuel elements by elements where about 2/3 of the fissile nuclei would be ^{235}U and the remaining 1/3 ^{239}Pu and ^{241}Pu would allow a stabilization of the plutonium inventory. Minor actinides should be extracted at each reprocessing, since these cannot be easily incinerated in thermal reactors.[25] The minor actinides could, then, be incinerated in fast [49] or subcritical [40,50,51] reactors.

Recently, it has been proposed [52] to incorporate in standard PWRs, special annular fuel rods highly enriched in plutonium. The PWR could, then, consume 160 kg of plutonium per year instead of producing 200 kg as in a present standard PWR. Incinerating minor actinides in dedicated fuel elements seems also possible. Such a solution would be extremely attractive, at least as long as uranium reserves do not command breeding reactors. Indeed, with minor modifications, the existing reactor system could be run with a stable plutonium inventory.

For increased efficiency in reducing the plutonium inventory it has also been proposed to substitute for uranium non-fissile matrices such as tungsten [54], which have the same properties as uranium as far as the temperature behavior of the reactivity is concerned. However, the reactivity would decrease rapidly with time, due to the rapid disappearance of the fissile nuclei. This decrease would require very frequent reprocessing or a high fuel enrichment in fissile species, associated with an initially large quantity of consumable neutronic poisons. Due to the absence of ^{238}U the delayed neutrons fraction decreases and reactor control may become difficult.

[25] Minor actinides are weakly fissile by thermal neutrons, but easily fissile by fast neutrons.

4.2. Waste incineration with subcritical reactors

Another solution, proposed, among others, by Rubbia [40], is to replace the depleted uranium by thorium. Thorium has neutronic properties close to those of ^{238}U. Incineration of plutonium would be associated with the production of ^{233}U. The proposed system could burn annually around 1.2 ton of plutonium while producing 0.7 ton of ^{233}U. This nucleus could, then, either be a substitute of ^{235}U in standard PWR fuel, or be a part of a new fuel based on the mixture ^{232}Th–^{233}U. In contrast with the ^{238}U–^{239}Pu mixture, such fuel could be reprocessed as many times as wanted in PWRs. The main difficulty of such a scheme would be the fuel element fabrication: irradiation of ^{233}U produces a significant amount of ^{232}U by $(n, 2n)$ reactions on ^{233}U and neutron capture by ^{231}Pa, the decay of which is accompanied by an intense high energy gamma activity which would require large biological shielding for fuel fabrication. The whole fuel cycle would have to be redesigned.

For the thorium–uranium cycle, ^{233}U would have a role similar to plutonium in the uranium–plutonium cycle. However, in this case, the production of transuranic elements is greatly reduced.

4.2.1. The thorium–uranium cycle

When the choice was made in favor of a plutonium economy, considerations about the level of nuclear waste production were not considered as a top priority. This is one of the reasons why the thorium–uranium cycle was disregarded. Nowadays it is clear that this cycle would offer great advantages by a considerable reduction of incineration needs. In this context, subcritical reactors, with their favorable neutron balance, might have special interest. Fig. 10 shows how the use of the thorium–uranium cycle could reduce the waste problem. The reference curve is that corresponding to a once-through PWR fuel, directly sent to the repository. For both thorium–uranium and uranium–plutonium breeding cycles, the following assumptions were made:

- Fast, lead cooled, subcritical reactors.
- Oxide fuels.

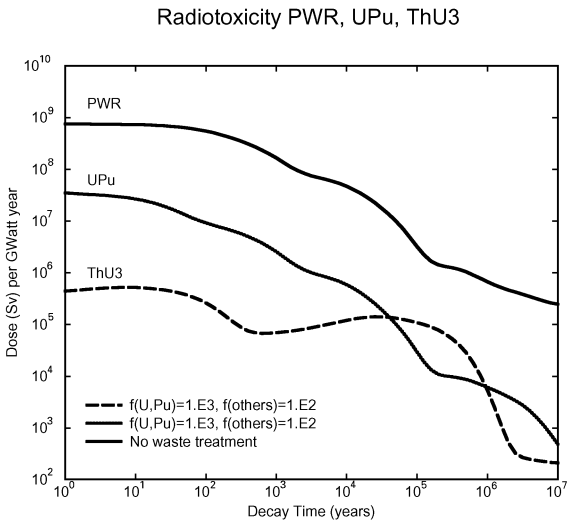

Fig. 10. Comparison of actinides ingestion radiotoxicities as a function of time. The calculation was made by M.E. Brendan. The curve labeled PWR corresponds to a one-through procedure. The two other curves assume multi-reprocessing with 0.1% losses for U and Pu elements and 1% for minor actinides.

- Burn-up of 66 Gwd/ton, a conservative figure for fast reactors.
- 5 years cooling before reprocessing and re-use.
- Reprocessing losses of 0.1% for uranium and plutonium and 1% for Minor Actinides.
- The composition of the wastes was that obtained after 5 cycles (irradiation + cooling); it is close to the asymptotic composition.
- No special treatment of the curium isotopes, although fuels with a significant amount of ^{244}Cm might be difficult, if not impossible to manufacture, due to the high spontaneous fission rate of this isotope.

The calculations were based on the coupling between the MCNP Monte-Carlo code [25] and an evolution code [53], and followed the following scheme:

- Define an initial concentration of the fissile part, in order to assure a small variation of k_s over the 5-year period between refueling events.
- Compute an initial set of one-group cross-sections through a detailed Monte-Carlo simulation.
- Use the calculated one-group cross-sections as input in the evolution code, allowing prediction of the new fuel composition 3 months later.
- Resume the calculation, using the new fuel composition.

It was observed that the one-group cross-sections did change with irradiation time due to the modification of the neutron energy spectrum.

The amount of waste depends upon the quality of the chemistry, i.e., the reprocessing losses ε, the burn-up B and the radiotoxic isotope concentration c in the fuel. Thus one can write that $w = w_0\varepsilon(B_0c/Bc_0)$ where w_0, B_0, c_0 are the values for the once-through situation. This expression shows that, for the same reprocessing techniques, fast reactors are less favorable than thermal ones since the ratio c/B is higher in the former case.[26]

Fig. 10 shows that the thorium–uranium cycle reduces the radiotoxicity of the wastes by almost two orders of magnitude with respect to the uranium–plutonium cycle, at least in the first thousand years. Furthermore, the heat released by the wastes is much less per unit volume for the thorium–uranium cycle. This should be of help in designing the storage facility, should it be still necessary.

Another advantage of the thorium cycle is that the activity of the mining refuse of thorium ore decays much more rapidly than that of uranium ore [19], because of the much smaller half-lives of the ^{232}Th progeny: 5.7 years for ^{228}Ra versus 77 000 years for ^{230}Th. In practice, due to the usual presence of uranium in thorium ore, the advantage depends upon the specific origin and nature of the ore. Furthermore, breeding reduces considerably the mining needs, in both cases.

4.2.2. Neutron balance for transmutation and incineration

The possibility of transmuting and incinerating nuclei depends on the neutron cost of these reactions. The simplest case is that of fission fragments.

[26] Although burn-ups are generally larger in fast reactors, this cannot override the larger concentration of fissile nuclei.

Table 5
Yields of technetium 99 and iodine 129 per fission of three important nuclei

Fissioning species	^{233}U	^{235}U	^{239}Pu
^{99}Tc	4.9×10^{-2}	6.1×10^{-2}	6.2×10^{-2}
^{129}I	1.8×10^{-2}	7.8×10^{-3}	1.4×10^{-2}

4.2.2.1. Fission fragment transmutation. The transmutation of fission fragments requires, evidently, at least one neutron per nucleus. The production rate of the most important long-lived fission products, ^{99}Tc and ^{129}I are given in Table 5. From this table it appears that at least 0.08 neutron per fission would be required to achieve transmutation of these two nuclei. Ideally, the most efficient way to transmute fission fragments is to use neutrons which would be lost to capture into the structural elements or which would leak out the reactor. This is why it has been proposed to capture neutrons in the resonances of fission fragments, whenever these display strong resonances [55]. In this way, it is hoped that neutrons are captured by the fission fragments before they reach thermal energies where captures in structure materials are important. We discuss these ideas in the case of a fast reactor using a lead reflector, such as was proposed by Rubbia et al. [36]. ^{99}Tc is characterized by the existence of a strong resonance at $E_R = 5584$ meV, with $\Gamma = 149.2$ meV and $\sigma_0 = 10^4$ b. It can be shown (Eq. (3.58) of PPNP) that the survival probability of a neutron after slowing down below the resonance is

$$P_{\text{surv}} = \mathrm{e}^{-\pi\Gamma/\xi E_R(\sqrt{1+\Sigma_0/\Sigma_s}-1)} \tag{47}$$

where $\xi \sim 2/A$ for heavy scattering nuclei with mass number A. After numerical evaluation ($\sigma_s =$ 10 b for lead) we obtain

$$P_{\text{surv}}(x) = \exp\left(-\frac{\pi \times 149.2}{55.84}(\sqrt{1 + x \times 10^3} - 1)\right) \tag{48}$$

where x is the concentration of ^{99}Tc nuclei with respect to lead. We find that 90% of the neutrons are captured for a ^{99}Tc concentration of 6×10^{-4}. In the original Energy Amplifier design [36], about 6% of the neutrons were captured in the lead. About half of these are captured below 5 eV and could thus be captured in the diluted Technetium. Since each fission produces 2.5 neutrons, it follows that 7.5 neutrons could be absorbed in technetium per 100 fissions. The volume of lead that must be considered is that where the neutron flux is high enough, rather than the full volume of the lead pool described in the Energy Amplifier proposal. The transport length in lead is around 1 m. It is found that the total weight of lead irradiated by high neutron flux is around 600 tons. The amount of ^{99}Tc which should be dissolved in order to capture 90% of the available neutrons would then be around 180 kg. The number of neutrons captured per year in ^{99}Tc would be

$$N_{\text{Tc}}^{(\text{cap})} = 8.4 \times 10^{25} \tag{49}$$

assuming a 10 MW beam and a value of $k_s = 0.98$. These captures correspond to a transmuted mass of 14 kg. The half-life of the ^{99}Tc in the neutron flux would be 7.5 years.

These data can be compared with those obtained with critical reactors. Calculations have been made both for fast and PWRs [48]. In the case of fast reactors best results are obtained using moderated assemblies where ^{99}Tc is mixed with hydrogeneous material like $CaII_2$. In the case of fast reactors, the shortest half-life is 15 years, while it is 21 years in the case of a PWR. Therefore, it appears that capture by Adiabatic Resonance Crossing [55], like that discussed above, might be advantageous.

For transmutation the most important parameter is the neutron flux, since the effective lifetime of a nucleus in a neutron flux is inversely proportional to the flux value. As an example, an ensemble of nuclei with a cross-section of 1 b, typical of some fission products, needs 200 years in a 10^{14} neutrons/cm^2/s flux to decrease by a factor of 2. Such numbers explain, partly, why projects such as that of Bowman et al. [2] aimed at a thermal neutron flux as high as 10^{16} cm^2/s.

4.2.2.2. Incineration of actinides. The only way that has been considered to dispose of actinides is to induce their fission. Although fission is accompanied both by energy and neutron production, several neutron captures may be necessary before

fission occurs, so that the net neutron number necessary for actinide incineration will be $N_{cap} + (1 - \nu)$ where N_{cap} is the number of captures before fission and ν the number of fission neutrons. The number of neutrons required depends on the neutron flux magnitude as well as of its hardness. Let us consider one nucleus of species $j(Z_j, A_j)$. It can suffer fission with average cross-section $\sigma_j^{(f)}$, capture a neutron with average cross-section $\sigma_{jk}^{(c)}$ (here nucleus k is $Z_k = Z_j$, $A_k = A_j + 1$), or decay to several possible other nuclei k, with partial decay rates λ_{jk} (here nucleus k is $(Z_k = Z_j + 1, A_k = A_j)$, $(Z_k = Z_j - 1, A_k = A_j)$ $(Z_k = Z_j - 2, A_k = A_j - 4)$ depending on the type of radioactivity involved). The fission probability reads $P_j^{(F)} = \sigma_j^{(f)}\varphi/((\sigma_j^{(f)} + \sigma_j^{(c)})\varphi + \lambda_j)$. The production of nucleus k from nucleus j can be defined as

$$A_{jk} = (1 - P_j^{(F)})\frac{\lambda_{jk} + \varphi\sigma_{jk}^{(c)}}{\sum_l (\lambda_{jl} + \varphi\sigma_{jl}^{(c)})}. \quad (50)$$

Starting with one nucleus i, the number of nuclei j which are ultimately produced is given by the system

$$y_j = \sum_k A_{kj} y_k + \delta_{ij}. \quad (51)$$

The Kronecker symbol expresses the fact that, initially, there was one nucleus i. Knowing the y_j, it is possible to compute the number of neutrons necessary to incinerate the nucleus i:

$$D_i = \sum_{j,\alpha} R_\alpha P_j^{(\alpha)} y_j \quad (52)$$

where the set $\{y_j\}$ is the solution of the system Eq. (51), R_α the neutron balance for reaction α (fission, capture or decay) and $P_j^{(\alpha)}$ the reduced transition rate for reaction α and nucleus j. The values of R_α are given in Table 6. The expression of D was first given in a slightly different form by Salvatores [22], and generalized to mixtures of nuclei. Table 7 gives values of D for important nuclei, as well as for commonly used fuel mixtures.

Table 7 shows [22] that incineration by fast neutrons is always a net neutrons producer. This is due to the fact that fission cross-sections of fertile nuclei, which are very small or vanishing for thermal neutrons, are large for fast neutrons.

Table 6
Values of the neutron balance for different types of reactions

	Capture	Fission	Decay
R_α	1	$1 - \nu$	0

Table 7
Values of neutron consumption D for incinerating selected nuclei per fission. The last 3 rows are the D for three representative fuel mixtures: Discharge of a PWR, D_{Tru}, transplutonium isotopes and neptunium extracted at discharge of a PWR, D_{TPu+Np}, and plutonium isotopes at discharge of a PWR, D_{Pu} [22]

Isotope or fuel	D for fast spectrum (10^{15} n/cm^2 s^{-1})	D for PWR spectrum (10^{14} n/cm^2 s^{-1})
^{232}Th (with Pa extraction)	−0.39	−0.24
^{232}Th (without Pa extraction)	−0.38	−0.20
^{238}U	−0.62	0.07
^{238}Pu	−1.36	0.17
^{239}Pu	−1.46	−0.67
^{240}Pu	−0.96	0.44
^{241}Pu	−1.24	−0.56
^{242}Pu	−0.44	1.76
^{237}Np	−0.59	1.12
^{241}Am	−0.62	1.12
^{243}Am	−0.60	0.82
^{244}Cm	−1.39	−0.15
^{245}Cm	−2.51	−1.48
D_{Tru}(PWR)	−1.17	−0.05
D_{TPu+Np}(PWR)	−0.7	1.1
D_{Pu}(PWR)	−1.1	−0.2

Neutron balance is not the only parameter to take into account for the choice of a particular system with the aim of waste incineration. The half-life of the waste in the neutron flux is also very important since it commands the necessary inventory and the amount of time necessary for disposing of the waste. In this respect, it can be shown that thermal reactors may be advantageous for incineration of fissile[27] TRU mixtures, like industrial plutonium obtained from PWR spent fuels [68]. However, for non-fissile sub-mixtures

[27] Recall that "fissile" refers to the fissionability with slow neutrons.

like americium and neptunium, thermal reactors become very subcritical and are no longer a practical choice.

5. Size of subcritical reactors

While critical reactors may have an arbitrary size, the presence of a localized primary neutron source imposes constraints on the size and total power of subcritical reactors. In this section we shall examine this question, first using a simple, intuitive model of a spherical reactor, and then taking the example of the optimization of the size of a possible demonstration set-up. Since the fuel evolution rate is a very important quantity, especially concerning inventories and incineration of transuranics, we first estimate, schematically, the fastest evolutions achievable for thermal and fast neutron's fluxes. The following considerations are valid both for critical and subcritical systems, provided the amount of subcriticality is not too large. For very subcritical systems an additional, very important, parameter will be the energy cost of the neutrons.

5.1. Maximum flux

For a given type of neutron's energy spectrum, the fuel evolution rate is, chiefly, determined by the value of the neutron flux, since the lifetime of a nucleus in a neutron flux is $\varphi\sigma^{(a)}$, independent of the nucleus concentration. We first discuss the maximum neutron flux which could be achieved in either a fast or a thermal reactor. The maximum heat density which can be extracted provides the maximum value of the product $\Sigma_f\varphi$. At present, values of 500 W/cm^3 are design values for liquid metal cooled reactors. This leads to a value of $\Sigma_f\varphi = 500/\varepsilon_f = 1.5\times10^{13}$ fissions/cm^2 s^{-1}. Very high values of the flux can be obtained if Σ_f is very small. However Σ_f cannot be arbitrarily small since the reactor has to be critical,[28] thus $k_\infty = \nu\Sigma_f/(\Sigma_f(1+\alpha)+\Sigma_{cap}) > 1$ where Σ_{cap} is the macroscopic capture cross-section of components other than the fissile part. Thus, one should have $\Sigma_f/\Sigma_{cap} > 1/(\nu-1-\infty) \simeq 1$. As an example of a thermal system we consider pipes containing a molten salt immersed in a heavy water tank. The lower limit of Σ_{cap} is given by that if heavy water $\Sigma_{cap} = 0.000044$. With a fission cross-section of 500 b this corresponds to a fissile nuclei density of 0.8×10^{17}/cm^3. The maximum maximorum of the neutron flux in a thermal reactor is thus 3.4×10^{17}. Of course, due to the components of the salt and of the pipes, such a value will probably never be reached. However, fluxes 10 times smaller have been considered, for example, by Bowman [2]. For such high flux the lifetime of fissile nuclei would be extremely short: 5 h for a 10^{17} n/cm^2 s^{-1} flux. The inventory in fissile material of the reactor would, also be extremely small: for the density of fissile nuclei of 3.0×10^{17}/cm^3, corresponding to the flux of 10^{17} n/cm^2 s^{-1}, the total fissile mass necessary for producing 3 GW would be only 700 g!. The total volume of the reactor would be 6 m^3.

For fast reactors we consider a dilute fissile species in molten lead with $\Sigma_{cap} = 3\times10^{-4}$ and, thus, a maximum maximorum flux of 5×10^{16} n/cm^2 s^{-1}. For such a flux the lifetime of the fissile species would be around 3.000 h. The minimum inventory for a 3 GW reactor would be 350 kg. This shows that thermal reactors have a higher potential for small inventories and fast burn-up. Of course, the actual realization of these potentialities could be very difficult. In fact, the more or less fissile nature of the fuel for thermal neutrons has a deep influence on the achievable incineration rate. This can be seen in the following more quantitative, although schematic, analysis.

We consider a schematic homogeneous infinite reactor with only two components:

1. the fuel, characterized by its atomic density n_{fuel}, absorption cross-section $\sigma_a^{(fuel)}$, and its neutron multiplication coefficient $k_{fuel} > 1$.
2. The coolant characterized by its atomic density n_{cool}, and its absorption cross-section $\sigma_a^{(cool)}$.

The aggregate reactor is characterized by its atomic density $n_{reac} = n_{fuel} + n_{cool}$, its absorption cross-section $\sigma_a^{(reac)} = (n_{fuel}/n_{reac})\sigma_a^{(fuel)} + (n_{cool}/n_{reac})\sigma_a^{(cool)}$, and an effective multiplication

[28] For a moderately subcritical system, the same condition is valid, in practice.

coefficient $k_{reac} = k_{fuel} n_{fuel} \sigma_a^{(fuel)} / (n_{fuel} \sigma_a^{(fuel)} + n_{cool} \sigma_a^{(cool)})$. We define the atomic fraction of the fuel $x = n_{fuel}/n_{reac}$. The criticality condition $k_{reac} = 1$ allows to express x as

$$x = \frac{\sigma_a^{(cool)}}{\sigma_a^{(fuel)}(k_{fuel} - 1) + \sigma_a^{(cool)}}. \quad (53)$$

Aside from the criticality condition it seems reasonable to assume that the fission density is limited to a specific value w. Thus,

$$w = x \frac{\sigma_a^{(fuel)}}{(1+\sigma)} n_{reac} \varphi \quad (54)$$

with $\alpha = (\sigma_a^{(fuel)} - \sigma_f^{(fuel)})/\sigma_f^{(fuel)}$. Thus, the incineration rate reads:

$$\lambda_{inc} = \frac{\sigma_a^{(fuel)}}{(1+\alpha)} \varphi = \frac{w}{x n_{reac}} = \frac{w}{n_{reac}} \left(1 + \frac{\sigma_a^{(fuel)}(k_{fuel} - 1)}{\sigma_a^{(cool)}} \right). \quad (55)$$

In the case of fissile mixtures, it appears that the main difference between fast and thermal reactors stays in the ratio $\sigma_a^{(fuel)}/\sigma_a^{(cool)}$. There is a clear advantage to use coolants with small absorption cross-sections. As examples, for heavy water $n_{cool} \sigma_a^{(cool)} = 4 \times 10^{-5}$ for thermal reactors and $n_{cool} \sigma_a^{(cool)} = 3 \times 10^{-4}$ for lead and fast spectra. Thermal neutrons fuel absorption cross-sections exceed 500 b while they range around 2 b only for fast neutrons. It follows that, for fissile mixtures, incineration rates with thermal neutrons could, in principle, be three orders of magnitude larger than those with fast neutrons.

The situation is different for non-fissile (Minor Actinides, for example) mixtures. In this case the major difference between thermal and fast neutrons incineration is that of the corresponding fuel multiplication factors. The subcritical nature of the MA fuel with thermal neutrons makes dilution of the fuel counterproductive since it would decrease the reactor multiplication coefficient k_{reac} below k_{fuel}, and thus require higher accelerator current to keep the neutron flux constant. The incineration rate reduces to the first term of the l.h.s. of Eq. (55), i.e.

$$\lambda_{inc} = \frac{w}{n_{reac}} \quad (56)$$

which means that it depends, essentially, on the fission density. Indeed, because of the condensed nature of the components of all practical reactors' designs it is not possible to vary very much the value of n_{reac}. For example, for water, the atomic density is $10^{23}/cm^3$, while, for lead, it is $0.3 \times 10^{23}/cm^3$ and for uranium $0.6 \times 10^{23}/cm^3$.

5.2. The homogeneous spherical subcritical reactor

Because of the localized character of the neutron source, one expects that the size of subcritical reactors will be limited. Optimization of the reactor has to be made with respect to several key quantities:

1. The value of the source multiplication factor k_s, which relates the beam power to the total power of the reactor, and, thus, to the energy gain. The possibility of innovative designs in this respect is discussed in Section 5.4.
2. The value of the effective multiplication factor k_{eff} which commands the safety of the reactor. Because of the general positive correlation between k_{eff} and k_s, the highest values of k_{eff}, compatible with safety, are looked for. In Section 3.3.1, we have seen that, for fast systems a limit of $k_{eff} = 0.98$ seems reasonable.
3. A maximum value of the specific power which is imposed by the heat removal system. Practically, maximum specific powers of the order of 500 W/cm^3 are possible with standard liquid metal cooling.
4. The fuel volume which needs to be minimized at the same time as the spatial variation of the specific power.

In order to give the reader a feeling of the size of subcritical reactors we have studied a simple spherical model of a reactor. The reactor is made of three concentric zones:

- The central zone (1), where the spallation reaction takes place and where we neglect neutron absorptions. This spherical zone has radius R_1.
- The fuel zone (2) between radius R_1 and radius R_2.
- A reflector zone (3) between R_2 and infinity.

The details of the treatment, based on the solution of the one-group diffusion equation, can be found in Section 6.2 of PPNP. We obtained the fuel volume necessary to obtain $k_s = 0.98$ as a function of R_1, for a reactor using a uranium–plutonium fuel with the following characteristics:

- The relative volumic fractions were 0.5 for lead, 0.08 for iron, 0.39 for the fuel and 0.03 for vacuum.
- The fuel was 88% uranium 238 and 12% plutonium, both in the dioxide form (see the next section for a justification of these proportions).
- The relative amounts of plutonium isotopes were 62.7% ^{239}Pu, 24.3% ^{240}Pu, and 13% ^{241}Pu, corresponding to the concentration of used PWR fuel.
- The cross-sections were one-group cross-sections extracted from the MCNP Monte-Carlo calculation which is described in the next section.

The results are shown in Fig. 11. The values shown in the figure correspond to a 1 MW proton beam, each proton is assumed to produce 30 neutrons. For an internal radius $R_1 = 0.15$ m, the maximum neutron flux reached is 3.6×10^{15} n/cm^2 s^{-1}, corresponding to a maximum specific power of 280 W/cm^3. The total thermal power is 120 MW. The volume of the fuel zone is around 0.7 m^3 for a fuel weight of 3.5 tons. This configuration can be considered as the smallest possible demonstration design that is able to:

- reach a multiplication factor of 0.98;
- approach the maximum acceptable specific power;
- reach representative neuron fluxes, so that fuel evolution can be studied in realistic conditions.

The 1 m internal radius could be representative of an energy producing reactor. The maximum specific power is 50 W/cm^3 for the 1 mA beam. A 10 mA beam would lead to an acceptable 500 W/cm^3 specific power and a 1200 MWth reactor, for a fuel zone volume of 2.7 m^3, and a fuel weight of 14 tons. In this configuration, the ratio of maximum to minimum flux is only 1.25, a very reasonable value. The thickness of the fuel zone is less than 20 cm.

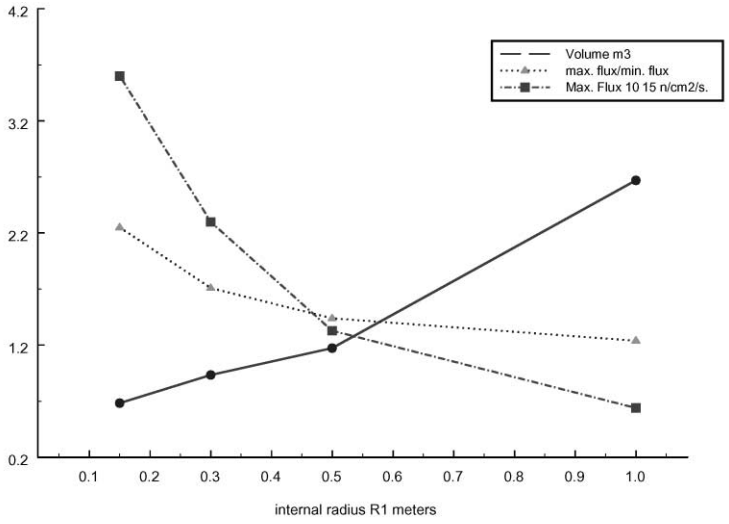

Fig. 11. Variations of the volume, maximum flux, and ratio of maximum-to-minimum flux, for a three-zones spherical reactor, as a function of the internal radius of the fuel zone. The multiplication factor is $k_s = 0.98$. The neutron source is a 1 mA/1 GeV proton beam producing 30 neutrons per proton.

A realistic reactor could neither be homogeneous nor spherical. The necessity to let the proton beam penetrate the reactor leads, rather than to spherical shape, to truncated cylindrical shapes which are not apt to a simple analytical treatment, even in the one group diffusion approach, More realistic calculations are necessary, and we give an example of one of these.

5.3. Realistic systems

5.3.1. Practical calculation methods

Practical calculations of reactors use either deterministic or Monte-Carlo codes. The deterministic codes are the most used for critical reactors calculations, while, on the other hand, Monte-Carlo methods are almost exclusively used for hybrid reactors. Since our emphasis is on the latter, we only give a short reminder of the deterministic methods.

5.3.1.1. Deterministic methods. These methods are essentially more or less elaborate approximations of the Boltzmann equation. The most widely used approximation is the multi-group diffusion theory which we outline here, as an example. The different groups correspond to energy bands $E_i < E < E_{i+1}$. The set of multi-group equations reads

$$D_i \Delta \varphi_i(\mathbf{r}) - \Sigma_{t,i}\varphi_i(\mathbf{r}) + \sum_j \Sigma_{r,j\to i}\varphi_j(\mathbf{r}) + \chi_i \sum_j \nu\Sigma_{f,j}\varphi_j(\mathbf{r}) \quad (57)$$

where $i(j)$ denotes the $i(j)$th group. $\Sigma_{r,j\to i}$ is the cross-section for a jump from group j to group i. $\Sigma_{t,i} = \Sigma_{a,i} + \sum_j \Sigma_{r,j\to i}$ is the cross-section for removing neutrons from group i. $\Sigma_{f,j}$ is the fission cross-section in group j and χ_i is the fraction of the fission neutrons which have energies within group i. The diffusion constant $D_i = \Sigma_{s,d}/3\Sigma^2_{T,i}$. The cross-sections should be computed as averages over the group energy domain by

$$\Sigma_i = \frac{\int_{E_i}^{E_{i+1}} \Sigma_i(E)\varphi_i(E)\,\mathrm{d}E}{\int_{E_i}^{E_{i+1}} \varphi_i(E)\,\mathrm{d}E} \quad (58)$$

which means that Eq. (57) are, in fact, a set of complicated integro-differential equations. In particular, in the resonance regions the flux has a complicated structure due to its depletion at energy in the vicinity of resonance energy. Thus, approximations are made on the calculation of the group cross-sections Eq. (58). In particular, in heterogeneous reactors one, first, computes the cross-sections, with a large number of groups, for the cells, with simplifying assumptions on the shape of the flux, and, possibly, correction factors. In a second step one computes the flux on the cell network. In practice, experiments are needed to validate the group cross-sections for each type of reactors.

5.3.1.2. Monte-Carlo methods. Monte-Carlo calculations follow the history of individual neutrons. The most used codes are MORSE [26] and MCNP [25]. The CERN group has written its own code, MC2 [36], which is, however, not in the public domain. The physics involved is basically the same in all these codes. Neutrons are propagated on straight paths in a medium, until they escape it or suffer a nuclear interaction which occurs with probability $\Sigma^{(i)}_T(E)$ characteristic of medium i. If the neutron exits the medium without interaction, it is, then, followed on the same trajectory, but with the new medium cross-sections. If the neutron interacts,

1. first, the struck nucleus (l) is chosen randomly, with a weight proportional to the partial macroscopic total cross-section of this nucleus $p(l) = n_{(l)}\sigma_{\mathrm{T},(l)}(E)/\Sigma_{\mathrm{T}}(E)$;
2. second the type of interaction α is chosen randomly according to its partial weight $\sigma_{\alpha,(l)}/\sigma_{\mathrm{T},(l)}$.

The cross-sections are evaluated from experimental data. They are, usually, found in nuclear data evaluated files like ENDF-B6, JEF 2.2, JENDL or BROND. These files, as well as the experimental files (*.EXFOR files in the CSFRS library), can be found on the National Nuclear Data Center (NXDO) site[29] at Brookhaven National Laboratory. It is important to note

[29] http://www.nndc.bnl.gov.

Another important site is that of the French Nuclear Agency: http://www.nea.fr.

that all resonances appearing on the evaluated files have not necessarily been experimentally observed.

In the resonance region the evaluation process proceeds in the following way:

1. Extract the resonances parameters from the experimental data. These are the resonance energy E_R, the resonance width Γ_T and the partial widths: neutron Γ_n, gamma Γ_γ and fission Γ_f. In the evaluation process the widths are corrected for the broadening due to the thermal Doppler effect, and for the experimental broadening.
2. Compute the average values of the widths $\langle\Gamma^{(\alpha)}\rangle$, of the level spacing $\langle D\rangle$ and of the strength functions $\langle\Gamma^{(\alpha)}/D\rangle$.
3. Reconstruct the cross-sections with the corrected resonance parameters, in the region where the experimental data show well-separated resonances.
4. In regions where resonances are not well separated on the experimental data, simulated cross-sections are built with resonance parameters chosen randomly. Individual partial widths are chosen following, the Porter and Thomas [56][30] distribution which reads $P_n \times (x) = n/2\Gamma(n/2)(nx/2)^{n/2-1}e^{-(n/2)x}$ with $x = \Gamma^{(\alpha)} / \langle\Gamma^{(\alpha)}\rangle$.[31] The average values $\langle\Gamma^{(\alpha)}\rangle$ are extrapolated from the region of well separated resonances or from nuclear model estimates. In the Porter and Thomas distribution, n is the number of degrees of freedom. For neutron elastic widths, there is only one final state, so that $n = 1$. For gamma rays, there are many available levels for the primary gamma decays, $n \simeq 30$–40. For fission, the relevant degrees of freedom are the Bohr and Wheeler transition states, and one finds, typically $n \simeq 3$–4. Note that large values of n correspond to small fluctuations around the average. Resonance energies are chosen according to the Wigner interval distribution [57] between next-neighbor levels with same spin and parity which reads: $P(S) = (\pi/2)Se^{-(\pi/4)S^2}$ with $S = D/\langle D\rangle$ and D the distance between two nearest neighbors.[32] Families of resonances with different spins and (or) parties are treated independently. In the continuum region, where experimental cross-sections are not available, the Optical Model is used to obtain cross-sections. This approach is limited to energies below 20 MeV. Efforts are presently being made to extend the Optical Model calculations and experimental data between 20 and 100 MeV [58].
5. The average neutron flux in a volume V is obtained by $\varphi = L/V$ where L is the total length traveled by all neutrons in volume V.

Monte-Carlo methods allow exact treatment of the most complicated geometries, the only limitation being the statistics. They also allow a quasi-continuous treatment of neutron energies. However, for reactors close to critically or, even more, for superficial reactors a special difficulty comes from the fact that more and more chains becomes infinitely long. To overcome this difficulty, one stops the calculation after a fixed time t_{step}, or number of generations n_{step}, and resumed it at that point with a limited sample of the results. The time over which the calculation step is carried out should be long compared to the generation time, but small compared to the evolution time: $\tau_D \ll l_{step} < \tau_D/(k-1)$ or $1 \ll n_{step} < 1/(k-1)$. This condition is not always easily fulfilled when the system becomes very super-critical.

The influence of very long multiplication chains on the accuracy of Monte-Carlo simulations have been recently discussed by the CERN group [59]. $M = 1/(1-k)$ being the total number of neutrons originating from one initial neutron, these authors give the number N of cascades to be generated to obtain a relative error ε on M: $N = 2.56M/\varepsilon^2 \times (v/2.55)$ with v the neutron number per fission. Equivalently, the precision for N cascades is $\varepsilon = 1.6\sqrt{M/N\, v/2.55}$.

[30] This law is also known as a chi-square distribution with n degrees of freedom.

[31] Note that widths $\Gamma^{(\alpha)}$ should not be confused with the Γ function.

[32] The Wigner law shows that levels with same spin and parity repulse each other. They do not follow the random Poisson distribution, and they reflect quantum chaos.

5.3.2. Optimization of the size of a possible demonstration subcritical reactor

In the process of designing a possible demonstration facility, a realistic study was made, using a MCNP calculation [60]. The fuel composition was the same as that described in the preceding section, except that the total concentration of the industrial plutonium could be varied, and the lead fraction was reduced to 30%. In addition to the constrainsts given there, it was required that k_∞ should not vary by more than one per cent during the first year. Such value is certainly more than would be acceptable for an industrial reactor, but it allows one to start with a higher plutonium concentration (no breeding), and, thus, to decrease the size of the reactor.

The geometry of the simulated reactor is shown in Fig. 12. The inner radius of the fuel zone was 15 cm throughout the calculations, while the external radius and height were such as to minimize the surface of the fuel zone for a given volume. Fig. 13 shows how k_s depends on the total initial plutonium concentration and on the volume of the fuel.

It was found that the evolution of k_s with time (dk_s/dt) was essentially independent of the fuel volume, as expected from the fact that neutron leakage should not vary much with time. The choice of an initial plutonium concentration of 12% lead to an acceptable value of $dk_s/dt = -0.007/\text{yr}$. In this case a fuel zone volume of 1.5 m^3 yielded the required initial value of $k_s = 0.98$, together with an average specific power of 150 W/cm^3 for 1 MW beam power.

It appears that the realistic simulations lead to about twice larger fuel volumes than the analytic calculations. The main reason for such a discrepancy is the existence of large neutron losses through the open ends of the fuel zone cylinder.

5.4. Two-stage neutron multipliers for subcritical reactor systems

We have seen that, if control rods are to be avoided, the multiplication factor k_{eff} should be limited to about 0.98 for standard fast-neutron subcritical reactors and 0.95 for the slow-neutron ones. This limitation on k_{eff} also limits the energy gain $G_0/(1-k_{\text{eff}})$ accordingly. An interesting suggestion was made already in 1958 by Avery [61], in order to increase the energy gain of a subcritical system, and revitalized by Abalin et al. [62] and Daniel and Petrov [63]. It consists in coupling two multiplying systems in such a way that neutrons produced in the first one can penetrate the second while those produced in the second cannot penetrate the first. We quantify the possible gain which can be obtained in this way.

Let one neutron be created in a multiplying medium. If absorbed, it produces k_∞ new neutrons. However, in a finite system it only produces

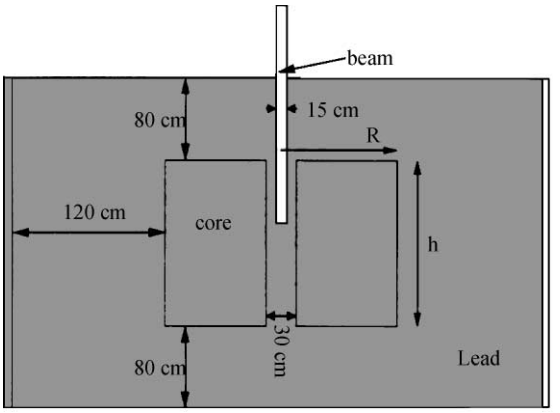

Fig. 12. Schematic view of the demonstration reactor.

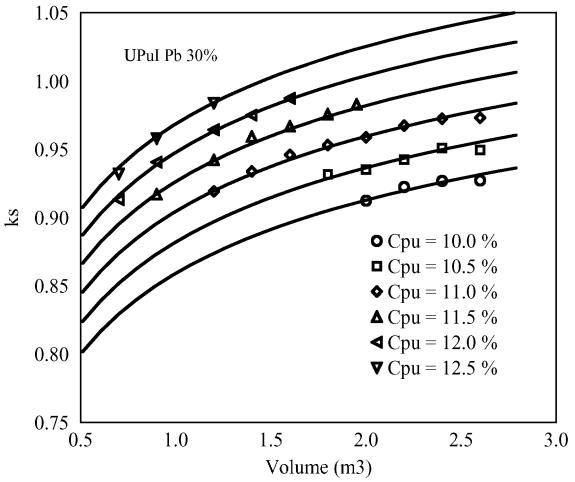

Fig. 13. Variations of k_s as function of the "industrial" plutonium concentration in the fuel and of the volume of the fuel zone of the reactor.

k_{eff} neutrons.[33] Since $k_{\text{eff}} = P_{\text{cap}} k_\infty$, the escape probability is

$$P_{\text{esc}} = 1 - \frac{d_{\text{eff}}}{k_\infty}. \tag{59}$$

If we consider a system with N_0 injected neutrons and multiplication k_{eff}, the number of escaping neutrons will be $N_0/(1-k_{\text{eff}})(k_\infty - k_{\text{eff}})/k_\infty$.

Now, we consider two multiplying media which communicate. Let

$$\bar{\bar{K}} = \begin{matrix} k_1 & \omega_{21} \\ \omega_{12} & k_2 \end{matrix}$$

be the matrix of efficiencies: a neutron born in medium 1 gives k_1 progeny in medium 1, and ω_{12} in medium 2. Let $n_i^{(1)}$ and $n_i^{(2)}$ be the number of neutrons in media 1 and 2 at generation i. The numbers of neutrons of the next generation are

$$n_{i+1}^{(1)} = k_1 n_i^{(1)} + \omega_{21} n_i^{(2)} \tag{60}$$

$$n_{i+1}^{(2)} = k_2 n_i^{(2)} + \omega_{12} n_i^{(1)}, \tag{61}$$

i.e.

$$\boldsymbol{n}_{i+1} = \bar{\bar{K}} \cdot \boldsymbol{n}_i$$

and the final number of neutrons as a function of the initial one:

$$\boldsymbol{n}^{(\text{F})} = \frac{\boldsymbol{n}^{(\text{Ini})}}{\bar{\bar{I}} - \bar{\bar{K}}}$$

which yields, for

$$\boldsymbol{n}^{(\text{Ini})} = \begin{matrix} N_0 \\ 0 \end{matrix} \tag{62}$$

$$n_1^{(\text{F})} = N_0 \frac{1-k_2}{(1-k_1)(1-k_2) - \omega_{21}\omega_{12}} \tag{63}$$

$$n_2^{(\text{F})} = N_0 \frac{\omega_{12}}{(1-k_1)(1-k_2) - \omega_{21}\omega_{12}} \tag{64}$$

while, if one neutron is created in medium 2, the final numbers are

$$n_1 = \frac{\omega_{21}}{(1-k_1)(1-k_2) - \omega_{12}\omega_{21}} \tag{65}$$

$$n_2 = \frac{1}{(1-k_2) - \omega_{12}\omega_{21}/(1-k_1)}. \tag{66}$$

[33] This is only true on the average for a neutron chosen randomly according to the flux distribution of the adjoint reactor.

If one could define a system where $\omega_{12} \neq 0$ and $\omega_{21} = 0$, one would get

$$n_2^{(\text{F})} = \frac{\omega_{12} N_0}{(1-k_1)(1-k_2)} \tag{67}$$

$$n_1^{(\text{F})} = \frac{N_0}{1-k_1}. \tag{68}$$

Abalin et al. [62] propose that the first medium could be a fast neutron multiplier but a strong thermal neutron absorber while the second medium would be both a slowing down and thermal neutron multiplier. The fast neutrons created in medium 1 could, eventually, reach medium 2 and be slowed down and multiplied. On the other hand, slow neutrons from medium 2 could not reach medium 1 without being, immediately, absorbed in the strong thermal neutron absorber (for example a Gadolinium nucleus). As suggested by Eq. (59), an estimate of ω_{12} is

$$\omega_{12} = \frac{k_{1\infty} - k_1}{k_{1\infty}} \tag{69}$$

which shows that it is interesting to maximize $k_{1\infty}$, and thus to use as pure-fissile material as possible. Then ω_{12} will, in general, be of order unity. It is mandatory that $(1-k_1)(1-k_2) > \omega_{21}\omega_{12} \simeq \omega_{21}$. In order for the system to be of interest as compared to standard ones, one should have k_1 and k_2 close to 0.95. This means that the coupling term ω_{21} should be less than 2×10^{-3}. A serious safety problem might arise from an unwanted decrease of the amount of absorber in medium 1.

Rather than using a difference between the neutronic properties of medium 1 and 2, it is possible to play on the relative geometrical arrangement of the two media. For example, consider that the first medium is a sphere of radius R_1 surrounded by a spherical shell at radius R_2 which is medium 2. A neutron exiting medium 1 has, evidently, a unit probability of entering medium 2, so that ω_{12} is given by Eq. (69). A neutron emitted from the inner surface of the shell has probability $1 - \sqrt{1 - (R_1^2/R_2^2)}$ to enter medium 1. In the absence of medium 1, neutrons lost by medium 2 exit by the external surface of the shell. If the shell is not too thick, the number of neutrons crossing the inner surface of the shell should be approximately equal to this last

quantity. It follows that

$$\omega_{21} \simeq \frac{k_{2\infty} - k_2}{k_{2\infty}} \left(1 - \sqrt{1 - \left(\frac{R_1^2}{R_2^2} \right)} \right). \quad (70)$$

The minimization of ω_{21} implies a minimization of $(k_{2\infty} - k_2)/k_{2\infty}$ in agreement with other constraints. Typically, for breeder reactors, $(k_{2\infty} - k_2)/k_{2\infty}$ is close to 0.1. It follows that the condition on the product $\omega_{12}\omega_{21}$ implies that $R_1/R_2 < 0.2$. With $R_1/R_2 = 0.1$, $\omega_{12}\omega_{21} \simeq 5.\times 10^{-4}$ and

$$n_2^{(F)} = 500 N_0 \quad (71)$$

for $k_1 = k_2 = 0.95$ and $\omega_{12} = 1$. Under these conditions, if a neutron is created in medium 2, the final number of neutrons will be $n_1 = 0.25$ and $n_2 = 25$ and $n_1 + n_2 = 25.25$. The large difference between the amplification when neutrons are created in medium 1 and when they are created in medium 2 shows that very high neutron multiplication can be obtained in the former case, the system remaining, however, safely far from criticality. This could give the possibility to reduce by almost one order of magnitude the necessary power of the accelerator.

Fig. 1 shows the result of a very simple Monte-Carlo calculation which illustrates the preceding discussion, and shows how a very high multiplication can be obtained, while keeping very far from criticality. The model reactor is made of a central plutonium sphere with a radius of 4.62 cm surrounded by a plutonium shell with an inner radius of 10 m and a thickness of 1.54 cm. Each single component is characterized by $k_\infty = 0.95$. The very high values of k_i for small i reflects the high value of η for pure plutonium. The sharp decrease is due to the large escape probability of neutrons created in the inner sphere. After 20 generations, the multiplication process takes essentially place in the outer shell. The simulated value of $k_s = 0.997$ is to be compared to the analytically calculated value of $k_s = 0.9964$.

Finally Daniel and Petrov [63] have proposed to use the difference in fissile concentration in zone 1(booster) and zone 2 to obtain a high value of k_s while keeping a reasonably small value of k_{eff}. They made a one-group diffusion calculation for a two-zone fast reactor with subcriticality $\rho_2 = 1 - k_{eff2} = 0.03$, for the external zone and $k_{1\infty} = 1.2$, corresponding to $k_{eff1} = 0.98$. For the spherical geometry and a volume ratio between the two zones of 10^3, they obtained a booster gain of 3.6, allowing a corresponding decrease in the beam power.

6. Practical proposals and projects for subcritical reactors

6.1. Lead cooled reactors

As typical examples of solid fuel subcritical reactors one can consider lead or lead–bismuth cooled reactors such as that proposed by Rubbia et al. [36] and presented in detail in the paper by Revol et al. in this issue. One may also cite those under consideration in the US [64,70] which are discussed in the paper by Beller et al. in this issue and in Japan [71]. The proposal of Rubbia et al. has the widest scope since it aims both at the establishment of a whole nuclear power system based on the thorium–uranium cycle and at the incineration of Transuranics produced in a classical PWR pool, with the associated production of ^{233}U which could be used as fuel for the PWRs on as seed for the establishment of the new ADSR-based thorium–uranium cycle. The US proposal has the more specific goal to incinerate plutonium and transplutonics produced by the existing PWR and BWR reactors, without specific extraction of the plutonium because of anti-proliferation policy. Finally, the Japanese project restricts itself to the incineration of transplutonic elements, plutonium being incinerated either in thermal or fast critical reactors. It is worth noting that, while the CERN and U.S. proposals use a lead or lead–bismuth spallation target, the Japanese propose to use a solid tungsten spallation target.

6.2. Molten salt reactors

Molten salt fuels give the possibility of quasi online treatment and purification which allows a very good control of the reactivity, as well as optimization of the neutron economy by preventing neutron losses by capture by fission products.

Some of the earliest proposals of subcritical reactors were based on molten salt fuels [1]. The Furukawa proposal is close in spirit to that of Rubbia et al., since it aims at the implementation of a thorium–uranium cycle. The main difference is the existence of two types of reactors: converters which produce almost as much ^{233}U as they consume and which need not be accelerator driven, and breeders which are powerful ^{233}U producers and need very high intensity accelerators. With the aim of incinerating Minor Actinides in the frame of the Omega project [71], the option of using molten salt fuels has been considered by Katsuta et al. [15].

As examples of molten salts systems, we describe two proposals made by Bowman on the basis of earlier work [69]. His original proposal [2] is based on a thorium–uranium cycle.

The main objectives are

1. to incinerate transuranics;
2. to transmute a number of fission products.

The very high proposed thermal neutron flux reaches $10^{16}/cm^2\ s^{-1}$. Neutron multiplication is obtained either by fission of uranium 233, or by fission of the actinides one wants to incinerate: plutonium, americium and curium. uranium 233 is obtained via neutron irradiation of a thorium 232 blanket, followed by an on-line extraction of protactinium 233 which is allowed to decay into uranium 233. This is made possible by the use of a molten salt fuel (a mix of fluorides), similar to that which was used in the Oak Ridge pilot reactor. The liquid fuel circulates continuously through the protactinium extraction facility. In order to limit the amount of neutron capture in protactinium, the thorium blanket is positioned in a neutron flux limited to a few 10^{14} neutrons/$cm^2\ s^{-1}$. The region of maximum thermal flux is where the actinides are incinerated. Indeed, very high fluxes have the following advantages:

- reduced lifetime of the actinides in the reactor; the lifetime of ^{239}Pu in a thermal neutron flux of 10^{16} neutrons/$cm^2\ s^{-1}$ is less than 2 days;
- improved neutron balance of the incineration process;
- small inventory of fissile matter in the system; the quantity of plutonium necessary to produce 3 GW, in a flux of 10^{16} neutrons/$cm^2\ s^{-1}$ is as small as 8 kg, with a daily burn out of 3.5 kg.

Transmutation of fission products would be optimal in the epithermal flux region since it is in the resonances that the absorption cross-sections are a maximum. Fission products with a capture cross-section of 1 b would live 3 years in a 10^{16} neutrons/$cm^2\ s^{-1}$ flux. In order to prevent stable fission products to become radioactive by neutron capture, an on-line separation of fission products to be transmuted is necessary.

The cited advantages are, of course, counterbalanced by the great complexity of the system:

- An accelerator able to accelerate protons to at least 1 GeV. with currents larger than 100 mA.
- A subcritical assembly using molten salt fuel. Although tested at a small scale at Oak Ridge, this technique has yet to demonstrate its resistance to very high fluxes. Corrosion problems may be serious, even if the use of hastalloy (a special nickel alloy) seemed to be satisfactory in the Oak Ridge conditions. One should also note that since the fuel itself circulates in the primary heat exchangers, any intervention on these often delicate components would be very difficult if not impossible.
- A very complex on-line chemistry for separation of protactinium, fission products and continuous injection of the fuel.

More recently, Bowman [68] has proposed to use a molten salt subcritical reactor for plutonium incineration with the principal aim to prevent its use for nuclear proliferation. The reactor would have the following characteristics:

- Thermal power: 750 MWth.
- Molten salt fuel with $NaF-ZrF_4$ carrier, fission fragments and plutonium fluorides.
- Thermal flux: 2×10^{14} n/$cm^2\ s^{-1}$
- Moderator: graphite.
- $k_s = 0.96$.

The reactor is fed with a mixture of fission fragments, zirconium and plutonium fluorides

obtained through fluorization of the spent fuels and extraction by sublimation of the uranium hexafluoride. The yearly input would be 300 kg of plutonium and minor actinides, 1200 kg of Fission Products and zirconium cladding.[34] The output would be 65 kg of plutonium and minor actinides, 1435 kg of Fission Products and carrier salt.

The advantages of such a system would be:

- no weapons plutonium or other weapons materials in repository;
- possibility of underground criticality in repository eliminated;
- 80% of fission energy recovered before waste emplacement;
- instant irreversible elimination of weapons potential upon entry into transmuter.

The emphasis is clearly put on the prevention of uncontrolled military use of the plutonium in spent fuels. One TIER reactor would be associated to every 3000 MWth reactor, thereby eliminating needs of radioactive material transportation.

Further incineration of the remaining plutonium and minor actinides would require more elaborate chemical processing in order to separate fission fragments. Special reactors would be devoted to the second stage of incineration. In this case, however, transportation would again be necessary, but with no risk of weapons materials smuggling.

6.3. Discussion of the basic ADSR components

Only a few Accelerator Driven Subcritical Reactors have been designed to some degree of details. These are, essentially, those described by Rubbia et al. [36], by Bowman [68] and by Furukawa [1]. However, significant and growing efforts are going on in the USA, Japan, Western Europe and Russia. These efforts aim at exploring the rather large space of possible ADSR concepts and designs. In a sense, the situation resembles that of the fifties and sixties when many concepts of critical reactors were examined and only a few survived. It may be that some decisions have been made too hastily, at that time, like the choice of Sodium Cooled Fast Breeder Reactors against Molten Salt Reactors. One should try to avoid too hasty a freezing of the ADSR design and take time to explore several of them. Choices have to be made concerning:

1. The type of neutron spectrum: fast or thermal.
2. The type of fuel: solid (metallic, oxides, nitrides, carbides, etc.) or liquid (fluorides, chlorides).
3. The type of spallation target: lead, lead–bismuth, tungsten, molten salt, etc.
4. The nature of the cooling agent: gas, molten metal, molten salt.
5. The accelerator system: cyclotrons or LINACs.

The difficulty to find an optimum design can be illustrated by a short discussion of each of these parts.

6.3.1. The neutron spectrum

Thermal neutron reaction cross-sections are, generally, much higher than those for fast neutrons. This gives the potential of higher incineration rates with thermal spectra, as stressed by Bowman [68]. However, as shown in Section 5.1, this is only true for fissile mixtures, like plutonium, but not in the case of non-thermally fissile ones like Minor Actinides [72]. In this case fast neutron spectra allow easier incineration due to their larger fission cross-sections.

As shown in Section 3.4.1, the protactinium effect, which limits the achievable values of k_∞, is less severe for fast spectra. In general reactor control is easier with fast spectra, especially for the thorium based cycle. For solid fuels, due to smaller capture cross-sections of fission products, the variations of k_∞ are less severe for fast than for thermal spectra. However, the inventory of ^{233}U is much larger in fast reactors (about 7 times), with the associated larger breeding times and inventory radiotoxicity.

6.3.2. The fuel

Solid fuels, especially oxides, have the advantage to be very well known and documented. A large experience with their reprocessing is available, mostly with wet processes, but also with pyro-chemistry. Due to progressive poisoning by

[34] The fact that fission products are not seperated is due to the extremely simple way to extract uranium by sublimation of UF_6.

fission products, the neutronics of solid fuels are not optimized. On the other hand, liquid fuels like molten salts allow a continuous monitoring and optimization of the neutronics. However, in spite of the very successful Molten Salt Reactor Experiment [37] at Oak Ridge, the reliability and safety of the on-line processing of the salt for large reactors has to be demonstrated. Similarly, although the MSRE has shown that hastalloy-n had good properties against corrosion by the salt, this has to be verified also for the very high irradiation doses expected with ADSRs. Fluorides are less corrosive than chlorides and appear to be the choice fuel. Their small atomic weight slows down neutrons and may be incompatible with fast spectra. However, a recent study concludes to the feasibility of a fast reactor with fluoride fuel [73].

The modern tendency to consider metallic fuels as the most promising when associated to pyrochemistry reprocessing involves a fluorization step. It would, then, be tempting to stop the process at this stage and use molten fluoride fuels.

Notwithstanding these technological challenges, molten salt fuels appear as very promising option for a new generation of nuclear reactors, either critical or subcritical.

6.3.3. The spallation target

Due to their higher neutron yields only heavy targets are considered practical.[35] Lead [36], or more often, lead–bismuth [70], are proposed as liquid targets. Lead has a rather high fusion temperature of 327°C and it might be difficult and costly to keep it in a fused state at all times. Lead–bismuth has a fusion temperature of only 123.5°C, bismuth leads to ample production of the very radiotoxic and volatile ^{210}Po. It is also produced, but at a very much lower rate, by lead. However, since it is possible for the lead–bismuth target to work at much lower temperature than pure lead the evaporation rates of ^{210}Po can be similar in both cases. Both lead and lead–bismuth corrode metals, the more so at higher temperatures. In this respect the lower working temperature of lead–bismuth is a further advantage.

Tungsten has been chosen as a solid target in several projects [16,71]. Very high energy depositions by the proton beam have to be disposed off. This is done with molten metal coolants, either sodium, lead or lead–bismuth. Sodium leads to the well-known safety problems related to the high chemical reactivity of sodium. Lead and lead–bismuth lead to the same solidification and corrosion problems as in the case of all liquid targets. Furthermore, the possibility of embrittlement of tungsten has to be considered.

Finally, building upon the Russian experience with lead–bismuth cooling of the reactors of nuclear submarines, lead–bismuth spallation targets seem to be especially attractive.

6.3.4. The cooling agent

Gas cooling. Some recent designs of ADSR [75] are inspired by High Temperature Gas Reactors. Such HTGR have some very appealing features:

- The high temperatures allow very high thermodynamical efficiencies with the possible implementation of combined cycles.
- For not too big reactors, radiation cooling is able to prevent fusion of the very refractory fuel (uranium or thorium carbides).
- Very high burn-ups of the rugged fuel could be obtained.

However, some limitations do exist: possible difficulties for a reliable fuel fabrication, difficulties for reprocessing the refractory and chemically inert fuel, low power densities due to the small thermal capacity of the gas, significant probability for a loss of coolant accident.

Present experience of HTGR fuels is with carbides. The large quantity of carbon in the reactor leads, naturally, to thermal reactors and would limit the possibility of HTGR for MA incineration. For that matter Framatome[36] is studying a new type of fuel based on Nickel alloys which might allow fast neutron spectra. However, it is not clear that such new fuels would allow one to reach as high temperatures as carbides would.

[35] An exception is the proposition by Mittig et al. [74] to use a deuteron beam and a light (Be, Li or C) converter producing fast neutrons which interact in a downstream heavy target.

[36] The French reactor building company.

Lead cooling. The Energy Amplifier proposed by Rubbia et al. makes an extensive use of molten lead both as spallation target and as cooling agent. The beam tube as well as the fuel elements lie in a swimming pool of 10 000 tons of molten lead. The design offers many advantages like convective cooling, passive safety and apparent simplicity. The simple design may help in keeping the lead molten and controlling the corrosion, although this remains a difficult challenge. One of the most delicate points of the design is the long beam tube which might be difficult to position and change. Furthermore, due to the high irradiation damages by the proton beam, this tube will have to be changed rather frequently. Finally, long-lived radiotoxic spallation products of lead like ^{194}Hg would be diffused in the whole 10 000 tons of lead and might cause serious decommissioning problems.

Lead–bismuth cooling. Because of the high melting temperature of lead it has been proposed to use eutectic lead–bismuth as coolant [70]. However, due to the high working temperature of the coolant necessary to obtain a good thermodynamical efficiency, the ^{210}Po evaporation may become a severe problem. The cost of bismuth is much higher than that of lead, and it is not clear that the bismuth reserves will be abundant enough to provide a large pool of reactors with the required quantities.

Molten salt cooling. Molten salt fuels are used simultaneously as coolant, with the possible problems of contamination of the secondary coolant loop. Even with solid fuels molten salts might be considered as an interesting option for cooling, provided corrosion can be managed. One of the advantages of molten salts over molten metals is that they are transparent to visible light, and thus allow visual inspections.

6.3.5. The accelerator

For acceleration either cyclotrons [36] or LINACs [1,2,70] are considered. Record intensities of more than 1 mA have been obtained for both types of accelerators at PSI for a cyclotron, and at LAMPF for a LINAC. Cyclotrons are more compact and thus require less space and are more economical. Due to the continuous nature of the beam structure and the compactness of the cyclotron center, it appears that the space charges and HF loadings obtained at PSI are already close to the limits. It seems difficult for cyclotrons to provide beam intensities larger than 5–10 mA. In the LINAC case, mA beam intensities have been obtained at LAMPF with 1% duty cycle. Space charge and instantaneous HF power are no limitations for reaching much higher beam intensities. Intensities in the 100 mA range are considered to be feasible. Since intensities between 5 and 10 mA are required for most ADSR projects, LINACs are usually preferred. However, if k_s values larger than 0.99 together with $k_{eff} < 0.98$, as described in Section 5.4 could be demonstrated, cyclotrons might become a good possibility again.

7. Conclusion

If one assumes that a massive use of nuclear power remains a possible option, it is clear that any production system should be fuel breeding. Hybrid systems have very good characteristics in this respect. They would allow switching from a plutonium economy to a much less polluting thorium economy. They could, in principle, allow the realization of intrinsically safe reactors. They are also an attractive option for nuclear waste incineration, including minor actinides (transplutonium elements) which would be difficult fuels for critical reactors. In their molten salt version they could also allow fast incineration of plutonium.

As mentioned in Section 6.3, numerous questions have to be studied: among them, reliability, safety and economical competitiveness. High intensity accelerators have to be built. A first demonstration prototype of several tense of MW could be built within 5–7 years. An industrial realization would probably require at least 20 years.

Hybrid systems require non-conventional technologies for the neutron multiplying assembly: molten salts, molten lead, natural convection, thorium–uranium cycle. In principle, such technologies could be used with critical reactors. The neutron surplus obtained from spallation is

relatively small, especially for fast systems. The main interest of subcritical systems is their subcriticality which would allow one to build reactors with a deterministic safety, and use fuels with unfavorable safety characteristics when used in critical reactors. They give a unique opportunity to improve the social acceptability of fission energy. In particular, given the well-known problems of the sodium cooled fast reactors, they are a credible alternative to reach breeding conditions, a must for any large extension of nuclear energy production.

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References

[1] K. Furukawa et al., The combined system of accelerator molten salt breeder (AMSB) and molten salt converter reactor (MSCR), Japan–US Seminar on Thorium Fuel Reactors, Naora, Japan, October 1982.
[2] C.D. Bowman et al., Nucl. Instr. and Meth. A 320 (1992) 336;
F. Venneri et al., Los Alamos Report LA-UR-93-752, 1993.
[3] F. Carminati et al., Report CERN-AT-93-47(ET), 1993.
[4] W.B. Lewis, Report AECL-968, 1952.
[5] AEC Research and Development Report, Facilities for Electro-nuclear (MTA) Program. Report LWS-24736, 1953;
Status of the MTA Process, Livermore Research Laboratory Report LRL-102, 1954.
[6] E.O. Lawrence, E.M. MacMillan, Luis W. Alvarez, Electro-nuclear Reactor, US Patent No. 933, 442 April 19, 1960.
[7] W.A. Gibson et al., Low-energy neutron production by high energy bombardment of thick targets, in ORNL-3940, p110-111, Electro-nuclear Division, Annual Progress Report, 1965.
[8] G.A. Bartholomew, P.R. Tunnicliffe (Eds.), The AECL Study for an Intense Neutron-Generator (ING) (Technical Details), Atomic Energy of Canada Limited Report AECL 2600, 1966.
[9] C.M. Van Atta, J.D. Lee, W. Heckrotte, The electro-nuclear conversion of fertile to fissile material, Lawrence Livermore Laboratory Report UCRL-52144, 1976.
[10] ABACS Study Group, Accelerator breeder and converter reactor symbiosis preliminary report on the promise of accelerator breeding and converter reactor symbiosis (ABACS) as an alternative energy system, Oak Ridge National Laboratory, Report ORNL/TM-5750, 1977.
[11] G.A. Bartholomew, J.S. Fraser, P.M. Garvey, Accelerator Breeder Concept, Atomic Energy of Canada Limited Report AECL-6363, 1978.
[12] T.J. Burns, D.E. Bartine, J.P. Renier, Concept evaluation of a nuclear design for electro-nuclear fuel production: evaluation of ORNLs proposed TME-ENFP (Tennary Metal Fueled Electro-nuclear Fuel Producer), Oak Ridge National Laboratory Report ORNL/TM-6828, 1979.
[13] J.S. Fraser, C.R. Hoffmann, S.O. Schriber, P.M. Garvey, B.M. Townes, Report AECL-7260, Chalk River Nuclear Laboratories, 1981.
[14] J.S. Fraser, G.J. Russell, H. Robinson, R.E. Prael, Nucl. Sci. Eng. 99 (1988) 41.
[15] H. Katsuta et al., Proceedings of the Second OECD/NEA Information Exchange Meeting on Actinide and Fission Product Separation and Transmutation, Argonne, November 11–13, 1993.
[16] G.J. Van Tuyle et al., Nucl. Technol. 101 (1993) 1.
[17] H. Nifenecker et al., Progr. Part. Nucl. Phys. 43 (1999) 683.
[18] Nuclear Wastes, Technologies for Separation and Transmutation, National Research Council, Nat. Academy Press, Washington DC, 1996.
[19] J.P. Schapira, in: R. Turlay (Ed.), Les Déchets Nucléaires, les Editions de Physique, 1997.
[20] Nuclear Waste Technical Review Board. Sixth Report to the U.S. Congress and the U.S. Secretary of Energy, NWTRB, Arlington, VA, December 1992.
[21] D. Bodansky, Nuclear Energy, AIP Press, Woodbury, New York, 1996.
[22] M. Salvatores, A. Zaetta, in: R. Turlay (Ed.), Les Déchets Nucléaires, les Editions de Physique, 1997;
M. Salvatores et al., Nucl. Sci. Eng. 116 (1994) 1.
[23] H.W. Bertini, Phys. Rev. 188 (1969) 171.
[24] L. Dresner, EVAP-a fortran program for calculating the evaporation of various particles from excited compound nuclei, Report ORNL-TM-7882, July 1981.
[25] W.L. Thomson et al., MCNP—a general Monte Carlo code for neutron and photon transport, LA-87545-M, April 13, 1981.
[26] M.B. Emmett, The MORSE Monte Carlo radiation transport code system, Report ORNL-4972, 1975.
[27] O. Bersillon, Second International Conference on Accelerator Driven Transmutation Technologies, Kalmar, Sweden, June 3–7 1996, p. 520.
[28] C. Rubbia et al., Report CERN/AT/95-44 (ET), 1995.
[29] D. Hilscher et al., Nucl. Instr. and Meth. A 414 (1998) 100.
[30] Y. Yariv, Z. Fraenkel, Phys. Rev. C 20 (1979) 2227.
[31] Y. Yariv, Z. Fraenkel, Phys. Rev. C 24 (1981) 488.

[32] J. Cugnon, Nucl. Phys. A 462 (1987) 751;
J. Cugnon, C. Volant, S. Vuillier, Nucl. Phys. A 620 (1997) 475.
[33] J.J. Gaimard, K.H. Schmidt, Nucl. Phys. A 531 (1991) 709;
A.R. Junghans et al., Nucl. Phys. A 629 (1998) 635.
[34] M.B. Chadwick et al., Cross section evaluations to 150 MeV for accelerator-driven systems and implementation in MCNPX, Nucl. Sci. Eng., Los Alamos Report LA-UR-98-1825, 1998.
[35] S. Andriamonge et al., Phys. Lett. B 348 (1995) 697;
F. Attale, Thèse Université Joseph Fourier Grenoble, May 1997, ISN 97-40.
[36] C. Rubbia et al., Report CERN/AT/95–44 (ET), 1995.
[37] E.S. Bettis, R.C. Robertson, Nucl. Appl. Technol. 8 (1970) 190.
[38] G. Ritter, Mémoire de DEA, Université Joseph Fourier Grenoble, 1995.
[40] C. Rubbia et al., Report CERN/AT/95-53 (ET), 1995.
[41] J.R. Lamarsh, Introduction to Nuclear Reactor Theory, Addison-Wesley, Reading, MA, 1966.
[42] J. Bussac, P. Reuss, Traité de neutronique, Hermann, Paris, 1985.
[43] Y. Kadi, Thèse, Université d'Aix-Marseille, 1995.
[44] H.S. Plendl (Ed.), Nucl. Instr. and Meth. A 414 (1998) 1.
[45] R. Turlay (Ed.), Les Déchets Nucléaires, les Editions de Physique, 1997.
[46] Special issue on nuclear wastes, Phys. Today, June (1997).
[47] International Commission on Radiation Protection, ICRP Publication 68, Pergamon Press, Amsterdam, 1995.
[48] J.P. Schapira, Transmutation des radionucléides à vie longue, principes, méthodes, réacteurs, accélérateurs, DEA lectures Université Paris XI, 1997.
[49] M. Viala, M. Salvatores, Proceedings of GLOBAL 95 International Conference On Evaluation of Emerging Nuclear Fuel Cycle Systems, Versailles September, 1995 p. 118.
[50] H. Nifenecker, O. Meplan, Workshop GEDEON at Jouy-en-Josas, 6–7 May 1996.
[51] M. Spiro, Workshop GEDEON at Jouy-en-Josas, 6–7 May 1996.
[52] A. Puill, J. Bergeron, Nucl. Technol. 119 (1997) 123.
[53] S. David, Thèse Université Joseph Fourier, Grenoble, 1999.
[54] N. Messaoudi, Thèse Université of Marseille, 1996.
[55] S. Andriamonge et al., Proposal: experimental study of the phenomenology of spallation neutrons in a large lead block, Report CERN/SPSLC-95-17, May 1995;
S. Andriamonge et al., in: G. Fioni et al. (Eds.), Nuclear Fission and Fission-Product Spectroscopy, Seyssins, France, April 1998, p. 26, American Institute of Physics, ATP Conference Proceedings 447;
A. Abanades et al., in: G. Fioni et al. (Eds.), Nuclear Fission and Fission-Product Spectroscopy, Seyssins, France, April 1998, p. 35, American Institute of Physics, ATP Conference Proceedings 447;
E. Belle, Thèse Université Joseph Fourier, Grenoble; 1998; Report ISN 98-71, 1998; P. Revol et al., Nucl. Instr. and Meth. 463 (2001) 586, this issue.
[56] C.E. Porter, R.G. Thomas, Phys. Rev. 104 (1956) 483.
[57] E.P. Wigner, Conference on Neutron Physics by Time-of-flight, Gatlinburg, Report ORNL-2309, 1959, p. 59;
C.E. Porter (Ed.), Statistical Theories of Spectra: Fluctuations, New York, 1965;
Weinberg, Perry, The collected works of E.P. Wigner, Part a, Vol. 5.
[58] A. Koning et al., Intermediate energy nuclear data: models and codes, Proceedings of a Specialists Meeting. Issy-les-Moulineaux, May 1994, OECD Documents; A. Koning et al., Nucl. Instr. and Meth. A 414 (1998) 48.
[59] S. Atzeni, Y. Kadi, C. Rubbia, Statistical fluctuations in Monte Carlo simulations of subcritical system, CERN/LHC/97-12(ET), 1997.
[60] D. Heuer et al., in: Proceedings of International Conference on Future Nuclear Systems (GLOBAL 97), Vol. 1, Yokohama, Japan, 1997, 440.
[61] R. Avery, Coupled reactors with suppressed feed back, Proceedings of the Second U.N. International Conference, Geneva, Vol. 12, 1958, p. 12.
[62] S.S. Abalin et al., Conception of electron beam-driven subcritical molten salt ultimate safety reactor, Preprint "Dubna", "Kurtchatov", "Novosibirsk" and "Kharkhov", 1997.
[63] H. Daniel, Y. Petrov, Nucl. Instr. and Meth. A 373 (1996) 131.
[64] G.J. Van Tuyle et al., Factoring Potential ATW Demonstration into APT Design Planning, APT-118-1998-67, RO, 1998.
[68] C.D. Bowman, Once-through thermal-spectrum accelerator driven system for LWR waste destruction without reprocessing: tier-1 description, Report ADNA/98-04, August 1998.
[69] A.M. Weinberg (Ed.), Nucl. Appl. Technol. 8 (1970) 102.
[70] A Roadmap for developing ATW Technology. ANL-99/16, 1999.
[71] T. Takizuka et al., A study of Incineration Target System, Proceedings of the Fifth International Conference on Emerging Nuclear Systems, Karlsruhe, 1989; M. Mizumoto et al., High intensity proton accelerator for nuclear waste transmutation, 16th International Linear Accelerator Conference LINAC-92, Ottawa, 1992.
[72] H. Nifenecker, O. Meplan, S. David, Comptes-rendus à l'Académie des Sciences, Paris, t.2, Sériе IV, 2000, pp. 163–184.
[73] M. Salvatores, I. Slessarev, private communication; GEDEON workshop Avignon, April 2000.
[74] D. Ridikas, W. Mittig, Nucl. Instr. and Meth. A 418 (1998) 449.
[75] B. Carluec, Proceedings of the 3rd International Conference on Accelerator Driven Transmutation Technologies and Application, Prague, September 1999.