PROLIFERATION HARDENING OF THORIUM FUSION BREEDERS WITH CANDU SPENT FUEL

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ABSTRACT

The denaturation of the ^{233}U fuel can be realised with a homogenous mixture of ThO_2 and CANDU spent nuclear fuel. The new ^{233}U component will be successfully hardened against proliferation with the help of ^{238}U component in the spent fuel. The plutonium component remains non-prolific through the ^{240}Pu isotope in the spent CANDU fuel.

In this work, a thorium fusion breeder is presented. A (D,T) fusion reactor acts as an external high energetic (14.1 MeV) neutron source. The fissile fuel zone, containing 10 fuel rod rows in radial direction, covers the cylindrical fusion plasma chamber. The rods contain a mixed oxide (MOX) fuel consisting of 25 % spent nuclear fuel of CANDU reactors and 75 % ThO_2. The fissile zone is cooled with pressurised helium gas with volume ratios of $V_{coolant}/V_{fuel}$ = 1 and 2 in the fissile zone.

The fissile fuel breeding occurs through the neutron capture reaction in the ^{232}Th (ThO_2) and in the ^{238}U (CANDU spent fuel) isotopes. The fusion breeder increases the nuclear quality of the spent fuel. It can be defined with the help of the cumulative fissile fuel enrichment (CFFE) grade of the nuclear fuel which is the sum of the isotopic ratios of all fissile material ($^{233}U+^{235}U+^{239}Pu+^{241}Pu$) in the MOX fuel.

Under a first-wall fusion neutron current load of 10^{14} (14.1-MeV n/cm^2.s), corresponding to 2.25 MW/m^2 and by a plant factor of 100 %, the MOX fuel can achieve an enrichment degree of ~ 1 % after ~ 12 to 15 months. A longer rejuvenation period (~ 30 months) increases the fissile fuel enrichment levels of the MOX to much higher degrees (~ 2 %), opening new possibilities for diverse purposes, such as utilization in advanced CANDU thorium breeders.

1. INTRODUCTION

Presently, light water reactors (LWRs) are supplying nuclear energy, followed by Canada deuterium uranium (CANDU) reactors. The former can exploit the nuclear fuel charge only to a moderate burn-up grade of up to around 30000 MWd/Ton, but they require enriched fuel. The latter are able to operate with natural uranium, but they can achieve even lower burn-up grades in the range of <10000 MW·d/Ton. Whereas, it is possible to fabricate mixed oxide fuel rods to withstand burn-up levels up to 200000 MW·d/Ton [1,2].

The main fissile fuel under utilisation is ^{235}U, which constitutes 0.7% of natural uranium. Some ^{239}Pu is also produced in LWRs and CANDU reactors. Still 99% of the fissionable natural resources are not being used for energy

production. The very long doubling time (10-30 years) of a fast breeder is not very promising in supplying the great number of LWRs under operation and construction with nuclear fuel.

Only non-fissile (external) neutron sources can enable the full exploitation of the naturally available nuclear fuel, which include both natural uranium as well as thorium reserves. The idea of the production of abundant fissile fuel through fusion breeders or electro-nuclear breeders is quite old [3-10]. Studies show that a fusion breeder can produce up to 30 times more fissile fuel than a FB per unit of energy. Typically for a hybrid reactor with suppressed fission [7],

$$\frac{\left(\frac{BR-1}{E}\right)_{HR}}{\left(\frac{BR-1}{E}\right)_{FB}} = \frac{\left(\frac{1.8-1}{27}\right)}{\left(\frac{1.2-1}{200}\right)} = 30$$

will be obtained.

On the other hand, world thorium reserves are estimated to be about three times more abundant than the natural uranium reserves. Early work has investigated the possibility of the production of ^{233}U in a fusion-fission (hybrid) reactor [5-11]. However, nuclear fuel, produced in fusion breeders can become of nuclear weapon quality with ^{239}Pu or ^{233}U in the fissile components. Hence, considerations for the denaturation of these new nuclear fuel sources become very important.

Fusion-fission (hybrid) reactors turn out to be major potential candidates for spent nuclear fuel rejuvenation [11-16]. They can ① produce electricity *in situ*; ② rejuvenate the spent nuclear fuel; and furthermore ③ burn all nuclear waste actinides, such as, ^{237}Np, ^{238}Pu, ^{240}Pu, ^{241}Am, ^{243}Am, ^{244}Cm through fast nuclear fission in these isotopes to a great degree, making use of them as nuclear fuel.

The subject of the present work is to analyse the breeding potential of a thorium fusion breeder with inherently enhanced protection properties against nuclear weapon proliferation. For that purpose, thorium fuel is mixed with spent CANDU fuel. Calculations are conducted, using a (D,T) fusion neutron driver for the hybrid reactor.

The behaviour of spent-fuel regeneration in a hybrid blanket is investigated for the following qualities of the cumulative fissile fuel enrichment (CFFE) grades:

1. Enrichment to 0.9 to 1.0 % for recycling in conventional CANDU reactors because the reactivity effect of ^{239}Pu is smaller than that of ^{235}U in a soft CANDU reactor spectrum.
2. Enrichment to 1.5 % for use in an advanced commercial HWR. Recent studies indicate that the burn-up rate in a CANDU reactor can be increased substantially if the fuel charge is slightly enriched [17], namely up to 1 to 1.5 %.
3. Enrichment of > 2.0 % for use in an advanced CANDU breeder concept with thorium fuelling. This reactor would require an average enrichment level of 1.5 to 2.0 % at start-up [18] in order to realise a commercially reasonable breeding capability of ^{233}U from ^{232}Th .

A nuclear spent fuel rejuvenation scenario would have the following economic potential's for an energy producing system:

1. Higher nuclear fuel exploitation (up to 100 % of total natural uranium, including ^{238}U, with reprocessing at necessary stages)
2. Reduction of the fuel rod fabrication costs
3. Drastic reduction of nuclear fuel reprocessing costs per unit of total energy production, notably in the initial critical reactor, during the rejuvenation in the hybrid reactor, and in the recycling phases up to the utilisation limit dictated by material damage
4. Lower nuclear fuel waste output per unit of total energy production.

2. BLANKET GEOMETRY

The neutronic analysis is performed on an experimental hybrid blanket geometry, which was presented to the international scientific community on different occasions [19,20].

Figure 1 shows the basic structure of the hybrid blanket adopted in this work. In this concept, a line neutron source in a cylindrical cavity simulates the fusion plasma chamber. A first wall made of stainless steel of type SS-304 surrounds the latter. Recent work has shown that a first wall made of SS-304 without Mo and Nb components would give in fusion reactors a C-class nuclear waste material after a plant

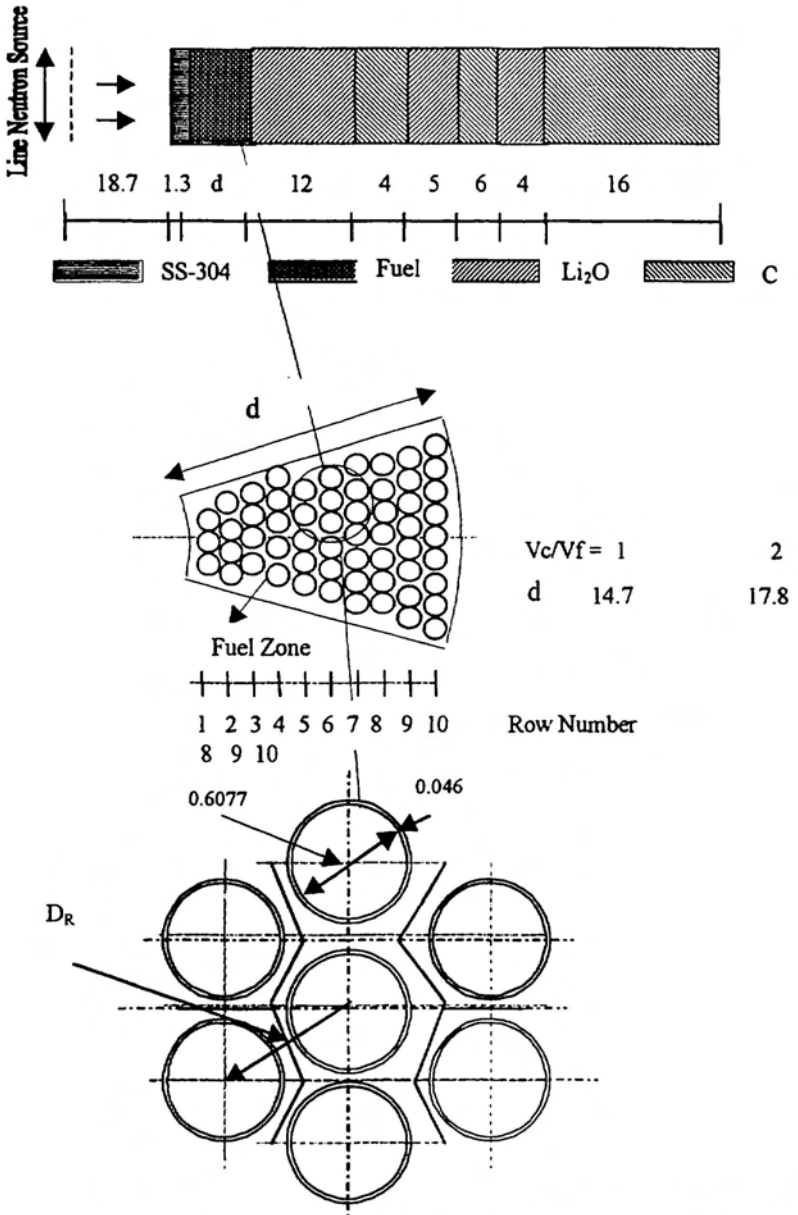

Figure 1. Cross-sectional view of the investigated blanket (dimensions are given in centimetre)

life time of 30 years which will be suitable for shallow burial after the decommissioning of the fusion reactor [21,22]. A first wall made of SS-304 can be selected only if water is not used as coolant material. Because Mo and Nb is needed in stainless steel mainly for corrosion resistance against of water. As the blankets, selected in this work are assumed to be cooled with pressurised helium gas, SS-304 is to be preferred instead of SS-316. A fast thorium fusion breeder would produce nearly 100 % ^{233}U, which would be highly prolific. For the purpose of proliferation highly prolific. For the purpose of proliferation hardening, fresh ThO_2 is mixed with spent CANDU fuel. The latter is the discharged fuel after a burn-up of 12252 MW.d/Ton [23]. The

isotopic composition of the CANDU spent nuclear fuel is given in table V of reference [23] and will not be repeated here. In figure 1, the fuel rods in the fissile zone contain a mixed oxide (MOX) fuel, composed of 75 % ThO_2 and 25 % spent CANDU fuel. The ^{238}U and the even plutonium isotopes(^{240}Pu) in the spent fuel can denaturate the ^{239}Pu and the ^{233}U components, respectively.

The fuel zone is cooled with pressurised helium gas coolant. The volumetric ratio of coolant-to-fuel (V_c/V_f) is selected as 2:1 and 1:1. The cladding of the fuel rods is made of zircaloy for the purpose of a direct reutilization in a CANDU reactor without reprocessing. It will be compatible with helium coolant. However, we remember at this point that it is not compatible with natural lithium nor with lithium derivatives, which are major candidate coolant materials in fusion technology.

The radial reflector is made of Li_2O and graphite in sandwich structure. This measure reduces the neutron leakage drastically and leads to a better neutron economy [11,19,20].

3. NUMERICAL RESULTS

3. 1. Calculational Procedure

The neutronic calculations have been carried out with the neutron transport code ANISN [24] using the neutron transport and activity cross section data libraries TRANSX-2 [25] and CLAW-IV [26]. In order to study major temporal effects in the blanket, it is assumed that the fuel zone is irradiated with a first wall neutron flux of 10^{14} (14.1-MeV) fusion neutrons/(cm^2.sec). This corresponds to a first wall neutron load of 2.25 MW/m^2. The plant

factor (PF) is assumed 100 %. For neutronic calculations, the fuel zone is divided into equidistant subzones which correspond to the fuel rod rows in the fissile zone, as shown in figure 1, in order to follow the space and time dependent nuclide density variations more accurately.

The temporal change of the fuel composition during hybrid reactor plant operation is evaluated for discrete time intervals Δt, as follows:

For breeding reactions (indices b),

$$+\Delta N_2 = PF \cdot \Delta t \cdot N_1 \cdot \int \sigma_{b1}(E) \cdot \Phi(E) \cdot dE + \Delta t \cdot \lambda_{b1} \cdot N_1 \quad (1)$$

Indices 1 and 2 denote mother and daughter isotopes, respectively.

For depletion reactions (indices dep),

$$+\Delta N = PF \cdot \Delta t \cdot N \cdot \int \sigma_{dep}(E) \cdot \Phi(E) \cdot dE + \Delta t \cdot \lambda \cdot N \quad (2)$$

E: Neutron energy
N: Isotope atomic density
σ: Microscopic cross section
λ: Radioactive decay constant
Φ: Neutron flux

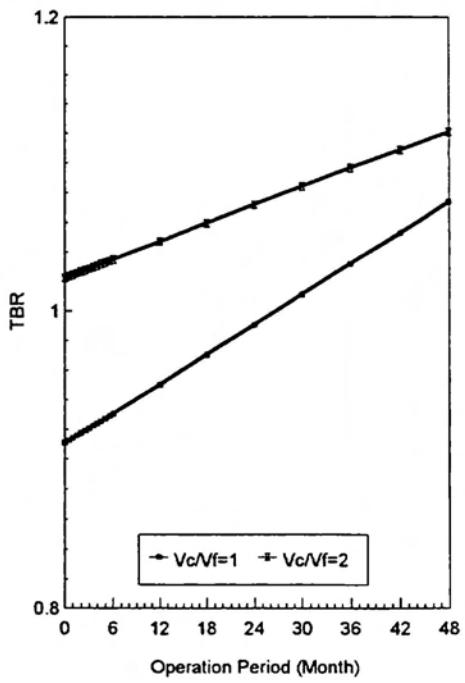

Figure 2. Temporal variation of tritium breeding ratio in the blankets

Equations (1) and (2) consider the variations of the atomic densities of the fissionable isotopes through both nuclear transmutations and as well as radioactive decay. The process of spent fuel

regeneration has been followed in each fuel rod row individually while considering variations in neutron spectrum over the radial coordinate within the fissile zone for discrete time intervals $\Delta t = 15$ days.

3.2. Integral Neutronic Data

The most pertinent integral neutronic data include tritium breeding ratio, blanket energy multiplication and peak-to-average fission power density in the fissile zone. An observation of these data leads over the plant operation time after at start-up one to the following conclusions:

3.2.1. Tritium Breeding Ratio

A sufficient tritium breeding has paramount importance for a self-sustaining fusion driver which would need a tritium breeding rate (TBR) > 1.05. Figure 2 shows the temporal variation of tritium breeding ratio in the blankets for a V_c/V_f = 1 and 2. In the blanket with V_c/V_f = 1, the TBR is not sufficient. But, it becomes amply sufficient in that one with V_c/V_f = 2, especially after an operation time of few months. In the course of the plant operation, TBR increases steadily due to the higher neutron multiplication rate along with the spent fuel rejuvenation.

3.2.2. Blanket Energy Multiplication

The fusion energy multiplication (M) is defined as the ratio of the nuclear heat release in the blanket to the incident fusion neutron energy and is plotted in figure 3. It increases with higher fission rate in the blanket. Due to the higher fissile fuel density, the increase becomes very steep for V_c/V_f = 1. The growths in TBR and M values indicate a temporal enhancement of the blanket neutronic performance. A quasi-invariable energy production over the operation period is essential to exploit the installed non-nuclear island of the plant at an optimal level

3.2.3. Peak-to-Average Fission Power Density

The peak-to-average fission power density ratio Γ in the blanket decreases steadily due to the

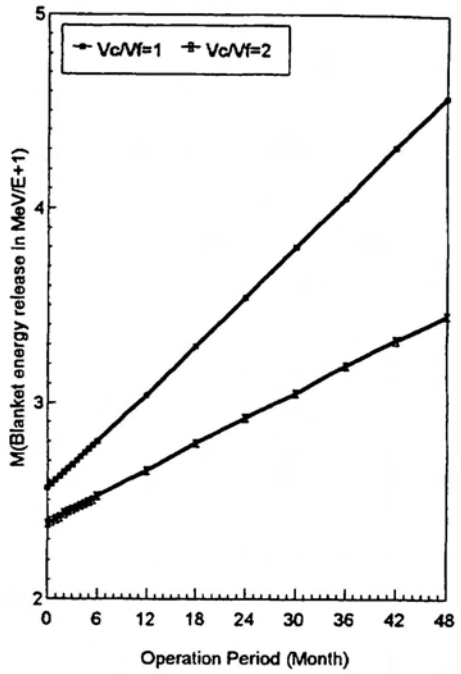

Figure 3. Temporal variation of the blanket energy multiplication

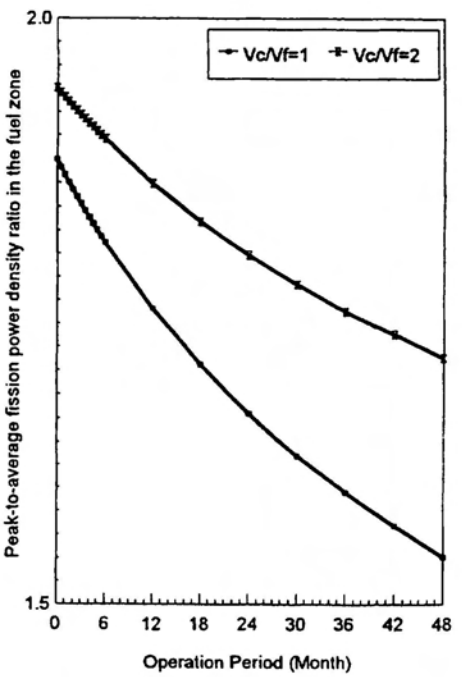

Figure 4. Temporal variation of the peak-to-average fission power density ratio in the blankets

increased fraction of the fission neutrons in the blanket. A low level of Γ allows a more uniform power generation in the fissile zone and consequently a better exploitation of the spent nuclear fuel in different rows in the fissile zone.

3. 3. Temporal Variation of the Cumulative Fissile Fuel Enrichment (CFFE)

Figures 5 and 6 show the steady increase of the CFFE in the blanket for discrete time intervals Δt = 15 days over a total operation period of t = 48 months for different fuel rods in the blankets with V_c/V_f = 1 and 2, respectively.

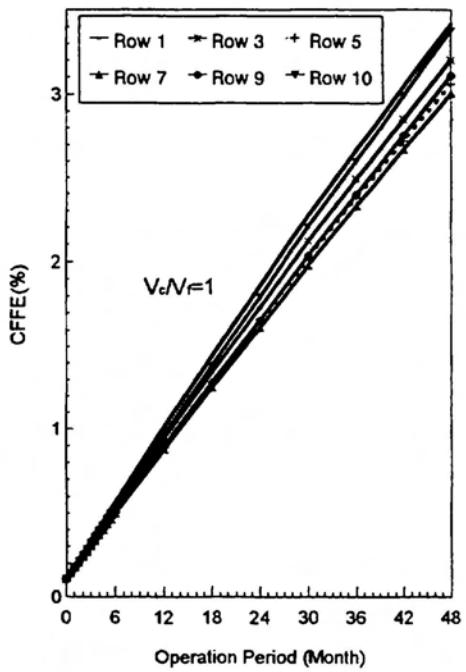

Figure 5. Temporal increase of the fissile fuel enrichment in the blanket (V_c/V_f = 1)

One can observe that the higher fuel density V_c/V_f = 1 causes only a slightly accelerated increase in CFFE, which can reach the levels of 1 %, 1.5 % and 2 % after a rejuvenation period of 12, 19 to 22 and 26 to 30 months, respectively, for the purposes described in chapter 1. For the blanket with V_c/V_f = 2, irradiation periods become slightly longer for the respective levels, namely 15, 23 to 26 and 32 to 36 months, respectively.

The softening of the neutron spectrum at the vicinity of the Li_2O zone will increase the (n,γ) absorption rates in the resonance's of the fertile isotope ^{238}U and accelerate the fuel rejuvenation towards the outer periphery of the fissile zone (row # 10).

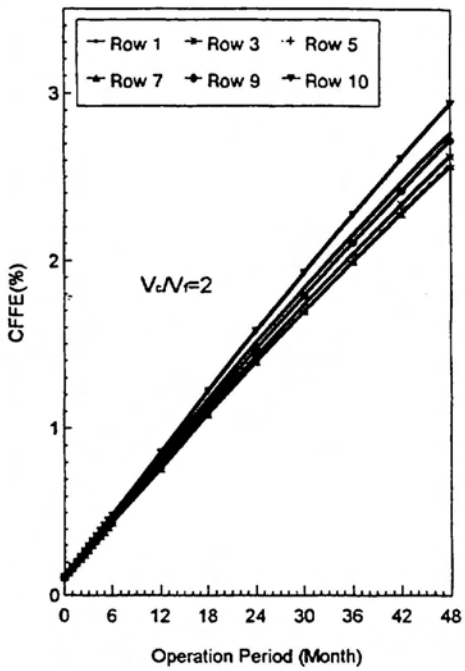

Figure 6. Temporal increase of the fissile fuel enrichment in the blanket (V_c/V_f = 2)

During all stages of the rejuvenation process, all fissionable isotopes in the spent fuel will contribute to nuclear heat generation and consequently to the electricity production of the hybrid reactor.

3.4. Fissile Fuel Burn-up

It would be of interest to estimate the grade of fissile fuel utilisation in the course the rejuvenation process in the hybrid blanket *in situ*. This can be measured easily with the help of fissile fuel burn-up (BU). It can be calculated as follows:

$$BU = PF \cdot \Delta t \cdot E_f \cdot 10^{14} \cdot F_w \cdot C_f \iint \Sigma_f(E) \cdot \Phi(E) \cdot dE \cdot dV / m_{fu} \quad (4)$$

where

E_f = energy per fission, 200 MeV

C_f = conversion factor, $1.6021.10^{-19}$ MW.s/MeV

F_w = Area of the first wall
m_{fu} = nuclear spent fuel charge

Figure 7 shows the average spent fuel burn-up in the hybrid blanket for a plant factor of 100 % as a function of plant operation period. These burn-up values are quite modest. The average burn-up of in a CANDU reactor will be 10000 MW.d/Ton. On the other hand, the full economic exploitation of the mixed oxide (MOX) fuel system requires burn-ups well above 200000 MWd/t [1,2]. Keeping that in mind, one can recognise that the same spent fuel bundles could be used in several cycles without the need for any fuel reprocessing and fuel fabrication, provided that the fuel elements could be designed to withstand the necessary burn-ups. This is a very significant aspect of the spent fuel regeneration in a hybrid reactor. It provides a very high level of safeguarding. The amount of nuclear waste fuel is reduced significantly, in terms of nuclear waste mass (in kilograms per unit energy output) by counting all steps of energy production in the critical reactor and in the phase regeneration in the hybrid reactor over all cycles before fuel rod destruction occurs.

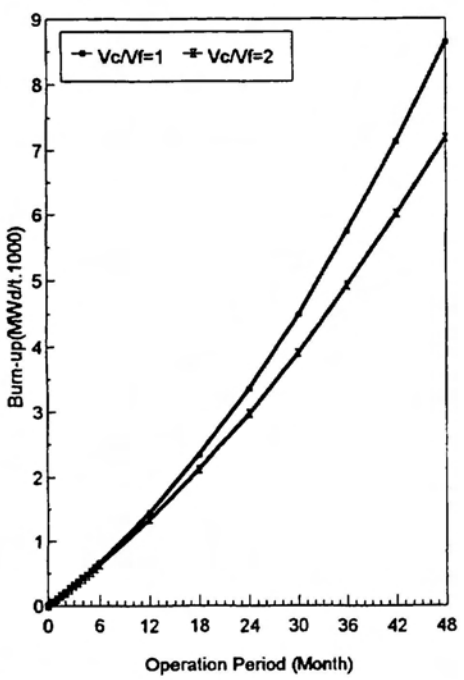

Figure 7. Temporal variation of fissile fuel burn-up in the blankets

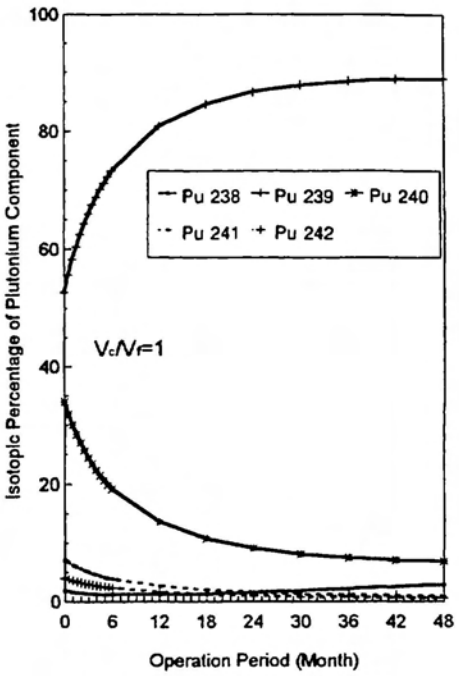

Figure 8. Temporal variation of the percentages of the plutonium isotopes in the blankets ($V_c/V_f = 1$)

3.5. Safeguard Aspects of the Plutonium Component

It would be of a primary interest to follow the non-prolific level of the MOX fuel during rejuvenation. The time evolution of the plutonium and uranium components must be investigated individually.

The intensity of spontaneous fission neutrons in ^{240}Pu (spontaneous fission half-life $T_{1/2}$ = $1.2*10^{11}$ years [27]) is about 50000 times higher than in ^{239}Pu ($T_{1/2}$ = $5.5*10^{15}$ years) so that only a few percent of ^{240}Pu would already denaturate the generated plutonium to a non-prolific level [28]. Previous analyses has indicated that the ^{240}Pu content must be <5 % in weapon grades plutonium fuel [29,30]. Therefore, a ^{240}Pu content of >10 % can be considered amply sufficient to denaturate the plutonium safely. In the CANDU spent fuel, the

isotopic percentages of ^{239}Pu and ^{240}Pu in the plutonium component at start-up are 52.8 % and 34.1 %, respectively. Hence, the initial spent fuel is fully denaturated.

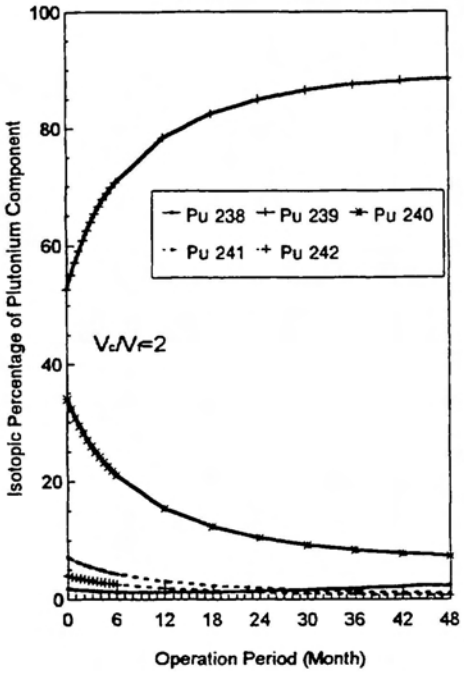

Figure 9. Temporal variation of the percentages of the plutonium isotopes in the blankets ($V_c/V_f = 2$)

Figures 8 and 9 show the variation of the percentages of all plutonium isotopes in the MOX over the plant operation. It is recognised that a rejuvenation period of even > 4 years will not be a critical issue in respect to safeguarding.

Although the nuclear quality of the plutonium increases steadily during plant operation, the isotopic percentages of ^{239}Pu and ^{240}Pu remain below 90 % and above 5 %, respectively, so that the plutonium component can never reach a nuclear weapon grade quality during spent fuel rejuvenation.

Figures 10 and 11 show the temporal variation of the percentages of the uranium isotopes in the MOX. The depletion of ^{238}U proceeds along with the accumulation of ^{233}U very slowly. Although it is difficult to define a concrete isotopic percentage for the prolific quality of ^{233}U, one can assume that a nuclear fuel with a

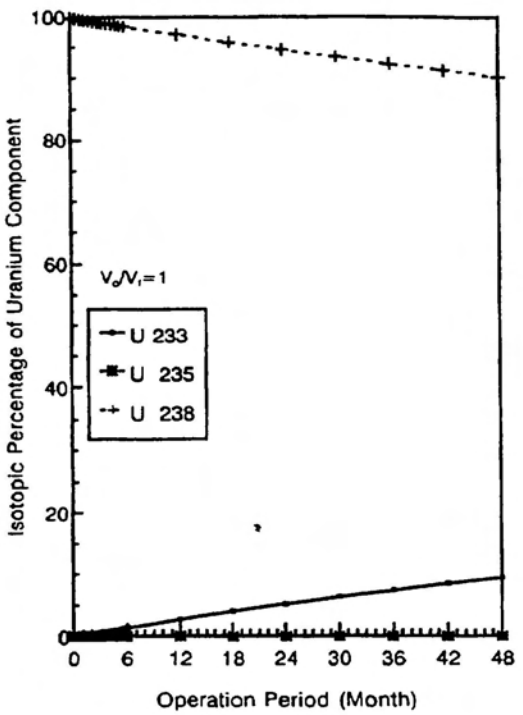

Figure 10. Temporal variation of the percentages of the uranium isotopes in the blankets ($V_c/V_f = 1$)

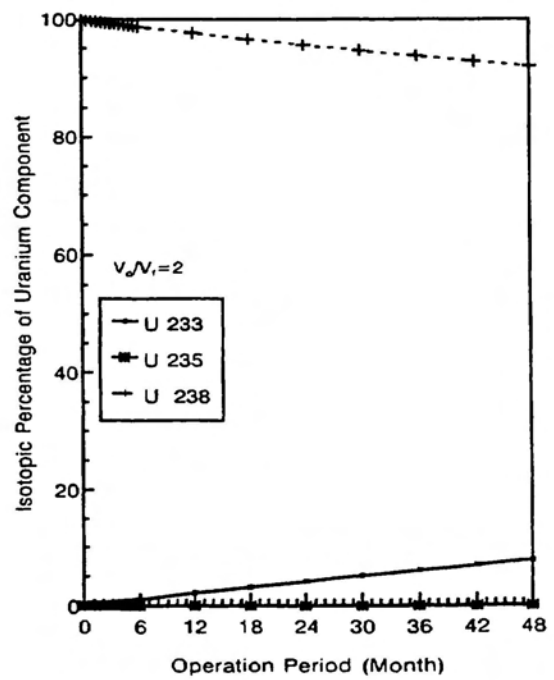

Figure 11. Temporal variation of the percentages of the uranium isotopes in the blankets ($V_c/V_f = 2$)

^{233}U fraction < 20 % becomes proliferation hardened. Under that assumption, ^{233}U becomes non-prolific even after 48 months. This is a very important and positive factor with regard to safeguarding.

4. CONCLUSIONS

In this study, a possibility of denaturing the ^{233}U fuel produced in a fusion breeder has been investigated. The main conclusions are as follows:

- A fusion breeder containing ThO_2 would produce weapon grade ^{233}U. It must be denaturated for commercial utilisation [1].
- The denaturation of the fuel in a fusion breeder can be performed by mixing ThO_2 with CANDU spent nuclear fuel in sealed fuel rods for reutilization in critical reactors without fuel rod reprocessing.
- ThO_2 mixed with CANDU spent nuclear fuel content of 25 % can produce fully denaturated fissile fuel.

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