ELSEVIER

Journal of Nuclear Materials 274 (1999) 181–188

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Neutronic analysis of U-free inert matrix and thoria fuels for plutonium disposition in pressurised water reactors

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Abstract

Inserting reactor-grade (RG) or weapons-grade (WG) plutonium in uranium-free matrices and burning it in light water reactors (LWRs) is an option gaining a wider consensus in the nuclear community. The main results of our neutronic studies performed in the last few years on this subject are reported. Our attention was mainly concentrated on two kinds of matrices: inert matrix in the form of calcia-stabilised zirconia, and thoria. Both materials are likely to exhibit excellent behaviour under irradiation (already demonstrated for thoria fuels) and high chemical stability. Direct disposal of spent fuel should be made feasible and attractive. A preliminary neutronic analysis was performed on these U-free fuels, imposing the constraint of maintaining the same assembly design and cycle length of a standard enriched-uranium fuel. In particular inert matrix fuel (IMF) showed a high plutonium burning capability, but associated with unacceptable feedback coefficients. Therefore, a whole IMF core results unfeasible, and only a partial core loading is possible. The solution then studied consists in replacing ≈21% of the pins of a standard enriched-U subassembly with IMF pins. Detailed assembly and core calculations were performed. A crucial aspect is the choice of a suitable burnable poison, which has to dampen the power peaks in the different fuel pin types without life penalisation. Among the considered poisons, a thin boron coating on the IMF pellets resulted the only effective one. Preliminary IMF pin cell calculations and the detailed ones gave similar results in terms of burnt plutonium fractions: 90% of fissile and 73% of total plutonium is burnt when RG plutonium is used. The main drawbacks of this fuel are the limited core loading capability and the lack of in-pile technological validation. In the case of Pu–Th fuels, pin cell calculations showed that increasing the plutonia content, decreasing the thoria content, and decreasing the pellet diameter are all possible ways to reach a longer fuel cycle and a higher percentage of burnt plutonium. Attained values for RG–Pu are $\geqslant$80% and >60% for the fissile and total plutonium, respectively. The use of IMF is an effective solution to proliferation concerns, while some concerns remain for thoria fuels because of the production of ^{233}U. This, however, can be eliminated by a small addition of ^{238}U. Long time radiotoxicity is scarcely affected by these fuels with respect to conventional MOX.

1. Introduction

Since the disarmament agreements resulted in large amounts of weapon grade plutonium to be disposed of, the option of burning plutonium has been considered under a novel standpoint [1–16].

The use of MOX fuel is the current reference solution. Such option does not aim at substantially destroying the plutonium stocks, but rather at reducing the proliferation concerns [17] down to the same level of the concerns regarding spent fuel, while producing electricity from those stocks. This technically sound solution cannot be considered fully satisfactory; in fact, the net plutonium consumption is limited, and the discharged Pu still gives rise to proliferation concerns, even if denatured from weapons-grade (WG) down to usual reactor-grade (RG) plutonium.

In Italy, we started in 1992 to consider alternative solutions, based on the absence of ^{238}U from the fuel [18].

In order of time, we studied the following uranium-free options for an efficient burning of WG- and RG-plutonium:

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0022-3115/99/$ – see front matter
PII: S0022-3115(99)00083-5

- IMF, based on stabilised zirconia;
- IMF in which part of zirconia is substituted with thoria;
- thoria fuel.

For all these solutions a once-through cycle has been considered, with direct disposal of the burnt fuel.

The present article is devoted to the neutronic analysis of these different solutions, while a companion article outlines the national activity on the technological aspects.

2. Pin cell calculations

The calculations were performed assuming that a reactor having the AP600 design characteristics was fully loaded with four different fuel types: $(U,Pu)O_2$, IMF, thorium-doped IMF, and $(Th,Pu)O_2$. The fuel compositions and their acronyms here adopted are shown in Table 1. For each fuel type, two different plutonium qualities, i.e. WG and RG, were considered; the corresponding isotopic composition vectors (^{239}Pu–^{240}Pu–^{241}Pu–^{242}Pu) were 94%–6%–0%–0% and 58%–24%–13%–5%.

The thoria content in the thorium-doped IMF was fixed to 30% mol. in order to achieve an acceptable Doppler coefficient (DC) value. For all ZrO_2 fuels, the constituting pellets were assumed to have a central hole with a volume equal to 25% of the total, in order to cope with the low thermal conductivity of the zirconia matrix. The plutonium inventory at the beginning of life (BOL) was determined such as to achieve a discharge burnup of 1185 equivalent full power days (EFPDs) for all fuels, considering a 3-batch fuel management scheme; this burnup value corresponds to 33 MWd kg^{-1} for a 3.2% enriched UO_2 fuel irradiated at a power rate of 13.5 kW m^{-1}. The depletion analysis was performed via the WIMSD-5 program [19]. This code, particularly suited for modelling cluster elements of RBMK, CANDU, SGHWR and AGR kind, is adequate for survey analysis also for LWR lattices. It uses a 69-group cross section library derived from the old UKAEA Nuclear Data Library and revised with the JEF-1 data files in 1986. The self shielding for the main resonance elements is obtained interpolating pre-calculated tables of effective energy-group resonance integrals as functions of the dilution cross section and temperature. After the cross sections are collapsed in few groups, the flux is computed via collision probability or discrete ordinate method in 1D geometry. Free, lattice, multicell o cluster boundary conditions can be taken into account.

In the past, computed results were successfully compared to experimental ones for fuels with large quantities of fertile nuclides, but the same level of confidence cannot be expected for IMF calculations. For this reason we participated in the definition and execution of a pertinent benchmark [20] which pointed out some discrepancies for the reactivity coefficients calculated with different codes.

2.1. Reactivity swing and reactivity coefficients

Fig. 1(a) and (b) report the reactivity swings for fuels bearing RG–Pu and WG–Pu, respectively. Among the four types of fuel, $(U,Pu)O_2$ and $(Th,Pu)O_2$ show almost coincident criticality rundown curves; in general, the curves show that the lower the fertile isotope content is, the larger is the criticality swing. In this case burnable poisons (BPs) should be added in order to dampen power peaking effects.

The values obtained for the reactivity coefficients, namely the DC, the void coefficient (VC) and the boron coefficient (BC), are detailed in Table 2. The DC has the lowest value for IMF, due to the lack of any fertile material, while for thoria-doped fuels it has a value close to that of UO_2, because of the selected composition. The same effect can be noticed for the VC (calculated for a 10% reduction of the water density); in general, for all the fuel types, the values are similar to that of UO_2, except in the case of IMFs, for which the coefficient can even become positive at end of life (EOL). The BC appears to be strongly influenced by the presence of plutonium: its high thermal cross section induces hardening of the neutron spectrum and causes partial shielding of the boron capture. The reduced boron worth and delayed neutron fraction render more difficult the control of a whole core plutonium-loaded reactor, thus requiring a different control rod design and the adoption of

Table 1
Composition of the analysed fuels (mol%) for obtaining 1185 equivalent full power days

Fuel	Description	PuO_2	UO_2	ZrO_2	ThO_2
PU–RG	$(U,Pu)O_2$ with RG-Pu	6.60	93.40	–	–
PU–WG	$(U,Pu)O_2$ with WG-Pu	3.65	96.35	–	–
IM–RG	IMF with RG-Pu	5.09	–	94.91	–
IM–WG	IMF with WG-Pu	4.14	–	95.86	–
PT–RG	$(Th,Pu)O_2$ with RG-Pu	8.12	–	–	91.88
PT–WG	$(Th,Pu)O_2$ with WG-Pu	4.92	–	–	95.08
TD–RG	Thoria-doped IMF with RG-Pu	5.94	–	64.06	30.00
TD–WG	Thoria-doped IMF with WG-Pu	4.32	–	65.68	30.00

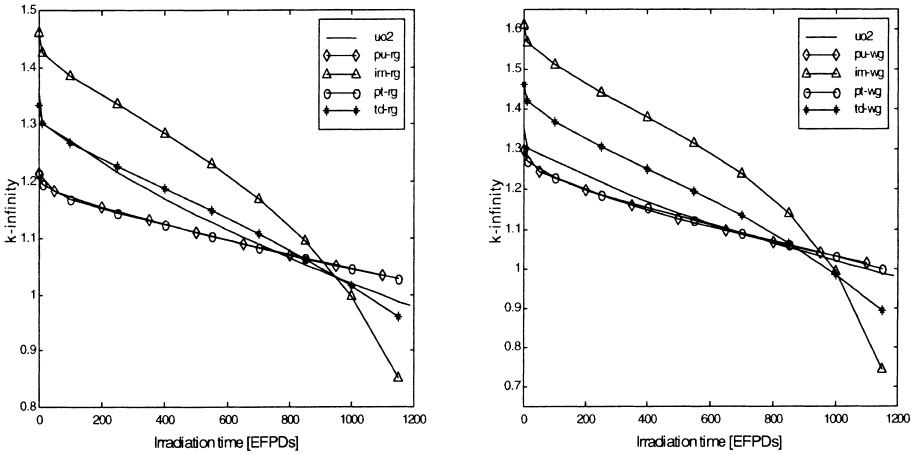

Fig. 1. (a) Reactivity swings for various reactor-grade plutonium fuels. EFPDs: equivalent full power days. b) Reactivity swings for various weapons-grade plutonium fuels. EFPDs: equivalent full power days.

Table 2
Doppler coefficients, void coefficients and boron coefficients for various fuels at beginning of life and end of life.

	DC (pcm K^{-1})		VC (pcm %void^{-1})		BC (pcm ppm^{-1})
	BOL	EOL	BOL	EOL	BOL
UO_2	−2.64	−2.96	−132	−184	−9.99
PU–RG	−3.00	−2.79	−174	−203	−3.08
PU–WG	−3.19	−3.01	−177	−211	−4.36
IM–RG	−0.99	−0.36	−87	−67	−4.61
IM–WG	−0.98	−0.01	−49	+58	−4.92
PT–RG	−3.29	−3.15	−167	−175	−2.98
PT–WG	−3.55	−3.36	−160	−155	−3.99
TD–RG	−2.32	−2.08	−138	−153	−4.28
TD–WG	−2.33	−2.03	−105	−82	−4.95

enriched boron as a soluble poison. The same concerns apply also for thoria fuels.

The low values of the reactivity coefficients make the IMF not suited for a full core loading; only a partial loading seems to be feasible with the help of BPs.

The plutonium balance calculated based on cell calculations for the various fuel types is reported in Table 3. The highest plutonium elimination potential is shown by the IMF: 92/96% of the fissile and 75/85% of total RG–Pu/WG–Pu is burnt during the irradiation. The thoria-doped IMF show interesting plutonium elimination capabilities, associated to acceptable reactivity coefficients. Fairly good results are obtained also for $(Th,Pu)O_2$. The performance of $(U,Pu)O_2$ are definitely worst because of the breeding of new plutonium from ^{238}U.

3. Assembly and core calculations

Because the IMFs are to be evaluated within a partial loading scheme, a simple pin cell calculation is not enough to assess their behaviour, but more detailed neutronic calculations are needed.

To this aim we considered a reference and a burner reactor. The reference reactor is an AP600 type, feeded with 3.2% ^{235}U-enriched fuel. The burner reactor is similar to the reference reactor, except for the fuel assembly in which 56 rods out of 264 (−21%) are replaced with IMF pins (see Fig. 2).

Complete calculations were performed for RG–Pu fuels. The content of fissile plutonium was set equal to 294 kg m^{-3}; this figure ensures the fissile plutonium content to be equivalent to the ^{235}U mass contained in the 3.2% enriched reference fuel [21].

The presence of a limited number of IMF pins is such that both reactivity swings and reactivity coefficients are only slightly worse than those of the reference reactor [22].

The CASMO-3 lattice program [23] was used to study the fuel assembly and to provide the neutron parameters and cross sections to be fed into the 3-D core simulator ABARTH [24].

With ABARTH, each fuel cycle was simulated up to the equilibrium one. As far as the fuel management is

Table 3
Plutonium balance (kg TWh_{th}^{-1})

	Pu_{fiss}				Pu_{tot}			
	BOL	EOL	Balance	Burnt %	BOL	EOL	Balance	Burnt %
PU–RG	60.6	39.5	−21.0	34.8	84.1	64.5	−19.6	23.3
PU–WG	43.6	21.8	−21.7	49.9	46.4	33.2	−13.1	28.3
IM–RG	42.9	3.4	−39.5	92.0	59.6	15.0	−44.6	74.8
IM–WG	45.5	1.6	−43.9	96.4	48.4	7.5	−41.0	84.6
PT–RG	69.0	28.5	−40.5	58.7	95.9	54.4	−41.5	43.3
PT–WG	54.2	12.6	−41.7	76.8	57.7	22.6	−35.1	60.8
TD–RG	45.3	7.5	−37.8	83.5	62.9	21.7	−41.1	65.4
TD–WG	43.0	3.1	−39.9	92.8	45.8	9.5	−36.3	79.2

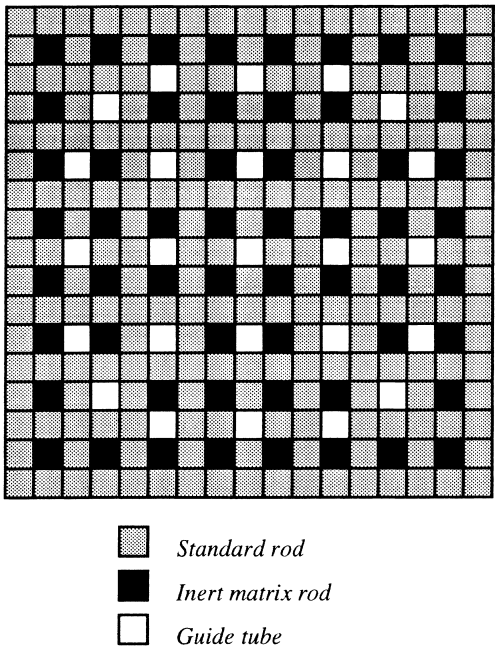

Fig. 2. Fuel assembly layout for the burner reactor.

concerned, a 3-batches out-in reloading strategy was adopted. The objectives of the fuel cycle in the burner reactor were: (a) to obtain a length of the equilibrium cycle comparable to that of the reference reactor, thus allowing the standard UO_2 rods in the burner and reference reactor to reach the same burn-up; (b) to keep as low as possible the maximum linear heat rate; (c) to achieve a high rate of plutonium consumption in the IMF rods.

The length of the equilibrium cycle turned out to be 409 EFPDs for the reference reactor and 393 EFPDs for the burner reactor. The concentration of soluble boron at BOL in the moderator turned out to be 945 and 1046 ppm, respectively.

Due to the high reactivity of the IMF rods, the radial core peaking factor of the burner reactor turned out to be 1.55, that of the reference reactor being 1.47.

Since ABARTH is capable of providing a detailed 3-D description of the core, it is worthwhile considering the rod maximum linear heat rate in the whole core during the equilibrium cycle. The core was then discretised in 13 axial segments and the linear heat rate of each fuel rod segment was evaluated. The maximum values for inert matrix and standard fuel are shown in Fig. 3(a). For the burner reactor the maximum values are of 40 kW m^{-1} at beginning of cycle (BOC) for the IMF and 19 kW m^{-1} at end of cycle (EOC) for the UO_2 fuel. The corresponding figures for the reference reactor are 25 and 18 kW m^{-1}.

In order to dampen the peaking factors and thus reduce the rod maximum linear heat rate, the utilization of BPs was considered. The shuffling strategy was accordingly modified.

As BPs we considered erbia (Er_2O_3), gadolinia (Gd_2O_3) and zirconium diboride coating in the form of integral fuel burnable absorber (IFBA). The study requirements were: (i) to reduce the peaking factors both inside the assembly and across the core; (ii) to achieve the complete consumption of the BPs during the cycle in order to avoid a reduction of the fuel cycle length for the same initial fissile material inventory; (iii) to lower, when possible, the content of soluble boron in the core for improving the reactor dynamic and safety.

Erbia did not appear to be an appropriate solution. When inserted in the IMF pins, erbia gave a significant reduction of the rod peaking factors, provided that its content was rather high, but also an important reduction of the fuel life, because of its relatively low absorption cross section; on the contrary, gadolinia was very efficient at BOC but its consumption was too fast, thus restoring the power peaks at EOC. Too high gadolinia loadings were not allowable for reactor start-up concerns.

The best performing BP was IFBA. In the chosen configuration, a 0.06 mm layer of ZrB_2 (equivalent to a density of $^{10}B = 223$ kg m^{-3}) was smeared on the fuel pellet.

Concerning the radial power distribution, the assembly peak ranges from 1.55 to 1.41 and moves back to

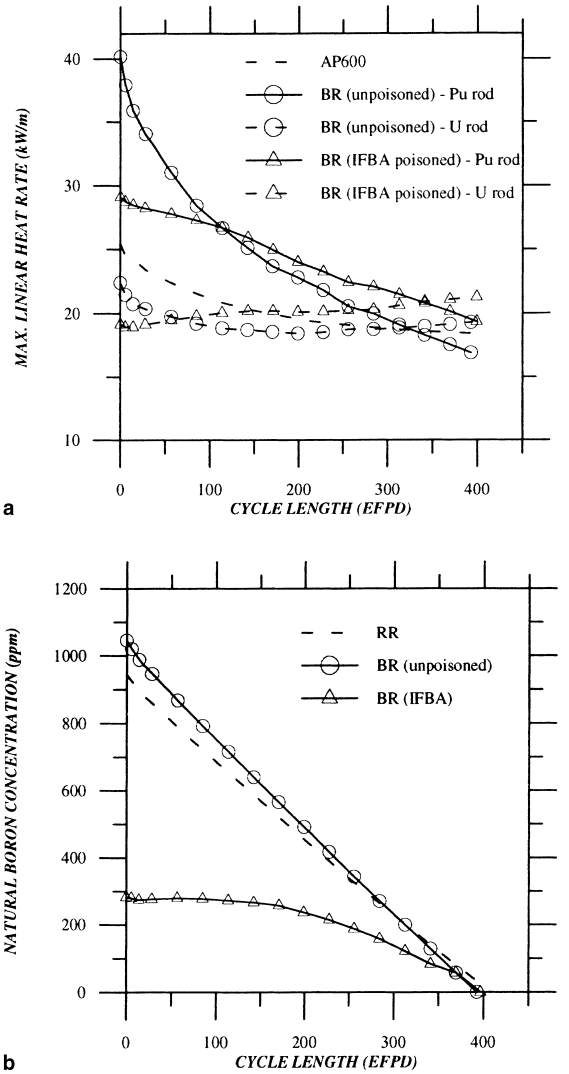

Fig. 3. (a) Max. linear heat rate. AP600 = reference reactor; BR = burner reactor. (b) Boron concentration. RR = reference reactor (AP600); BR = burner reactor.

the central region of the core, thus reducing the core leakage. The cycle length is 399 EFPDs, thus slightly increased with respect to the unpoisoned case; this is due to the reduced leakage. The concentration of soluble boron is lowered to 283 ppm of natural boron (Fig. 3(b)), with a positive impact on the reactor safety and operation. In spite of a slightly higher maximum linear heat rate at EOC in the uranium rods (21 kW m^{-1} against 19 kW m^{-1} for the unpoisoned case), the maximum value for the IMF rods at BOC is reduced from 40 to 29 kW m^{-1} (see Fig. 3(a)). These improvements, even if substantial, are not fully satisfactory. Worse results are expected for WG plutonium. Therefore technological specifications about the IMF rods' limiting heat flux, and maybe a further BP optimisation, are needed.

Table 4
Plutonium mass balance (wt%) at BOL and EOL for IMF rods (reactor-grade Pu). (Fuel assemblies with 56 IMF rods)

	BOL	EOL	Burnt
Pu^{239}	58	1.06	98.17
Pu^{240}	24	10.26	57.25
Pu^{241}	13	6.17	52.56
Pu^{242}	5	9.27	
Fissile Pu	71	7.23	89.82
Fertile Pu	29	19.53	32.64
Total	100	26.76	73.24
Fiss./fert. Pu	2.45	0.37	–

3.1. Plutonium consumption

The plutonium isotopic balance is shown in Table 4 for the IMF.

The fraction of fissile plutonium is 71% at BOL and 27% at EOL: plutonium in the discharged fuel is quality-poor, thus greatly reducing its proliferation potential. The burnt fraction of fissile plutonium is 90%, while 73% of the whole loaded plutonium mass is annihilated. It is worth noticing that these figures are close to those obtained in the pin cell calculations, 92% and 75% respectively (Table 3).

Despite the high fraction of burnt plutonium, its annual amount can be reduced only proportionally to the fraction of IMF rods over the total number of rods. For this reason we considered again the utilisation of (Pu,Th) fuels, seeking for enhanced burning performances.

4. Variants on (Pu,Th) fuel for enhancing its burning capabilities

It seems reasonable to attempt to reduce the ThO_2/PuO_2 fraction so as to lower the initial loss of reactivity and then obtain an increment in the Pu incinerated mass.

Various cases are here presented for WG–Pu and for RG–Pu, and namely: reduction of pellet diameter, adoption of hollow pellets (both with the same plutonium content), increase in Pu content, and a combination of them.

The results, detailed in Table 5, refer to pin cell calculations performed with the WIMSD-5 code. Having fixed the equivalent pitch value to 13.17 mm, a pellet diameter reduction from 8.2 to 7.2 mm results in a better moderated lattice, so limiting the excessive hardening of the reactor spectrum which results when large amounts of plutonium are inserted in the fuel. Therefore, the fuel

Table 5
Combination of several modifications on the $(Pu,Th)O_2$ pellet design

∅ (mm)	EFPD (days)		Pu_{fiss} BOL (kg TWh_{th}^{-1})		Pu_{fiss} burnt (%)		Pu_{tot} BOL (kg TWh_{th}^{-1})		Pu_{tot} burnt (%)	
	RG	WG	RG	WG	RG	WG	RG	WG	RG	WG
8.2 [a]	1185	1185	69.0	54.2	58.7	76.8	95.9	57.7	43.3	60.8
8.2, var. 1	1499	1387	54.6	46.3	71.4	85.8	75.8	49.3	54.5	70.8
8.2, var. 2	2026	2082	60.6	46.3	66.2	84.9	84.1	49.3	50.3	70.4
8.2, var. 3	2316	2243	53.0	43.0	74.4	90.0	73.6	45.7	58.0	77.0
7.2	1652	1427	49.5	45.0	77.8	88.2	68.8	47.9	59.3	72.6
7.2, var. 1	1791	1509	45.7	42.6	83.2	91.9	63.4	45.3	65.0	77.6
7.2, var. 2	2513	2298	48.8	42.0	80.0	92.0	67.8	44.6	62.4	78.7
7.2, var. 3	2645	2344	46.4	41.1	84.0	94.2	64.5	43.8	66.8	82.1
8.2, IM	1185	1185	42.9	45.5	92.0	96.4	59.6	48.4	74.8	84.6

[a] Reference case.
Note: var. 1: Hollow pellet (volume reduction 25%); var. 2: Increase in the Pu content (+50%); var. 3: Combination var. 1 + var. 2; 7.2: Pellet diameter reduced to 7.2 mm; IM: inert matrix fuel.

is able to sustain the reactor's reactivity for a longer time, reaching better burning performances. In fact, for both plutonium types, the 7.2 var. 3 case yields almost the same plutonium elimination capability as the standard IMF. Therefore, it seems that thorium fuel can be viewed as a viable alternative to IMF.

During irradiation, thoria-bearing fuels generate the highly fissile ^{233}U, a major concern from a proliferation standpoint. However, a potential obstacle to diversion lies in the gamma activity of daughters of ^{232}U which emit highly penetrating photons, thus rendering the spent fuel difficult to handle and easy to safeguard. Another conceivable countermeasure to cope with the proliferation risk, could be the addition of small amounts of ^{238}U in order to denature the uranium mixture.

The effect of adding some ^{238}U is displayed in Fig. 4 for reactivity and burnup, respectively. The reactivity is decreased for lack of self shielding of the ^{238}U. This penalty is translated in terms of burnup reduction, which turns out to be less important with increasing initial plutonium content. The reported reactivity and burn-up values are normalised to those of a fuel without ^{238}U. In both figures the two curves are relative to the 8.2 ref. case and 8.2 var. 2 case.

5. Radiotoxicity issues

It is commonly acknowledged that the thorium fuel cycle has a lower production of minor actinides than the standard fuel cycles, but this is not the case when plu-

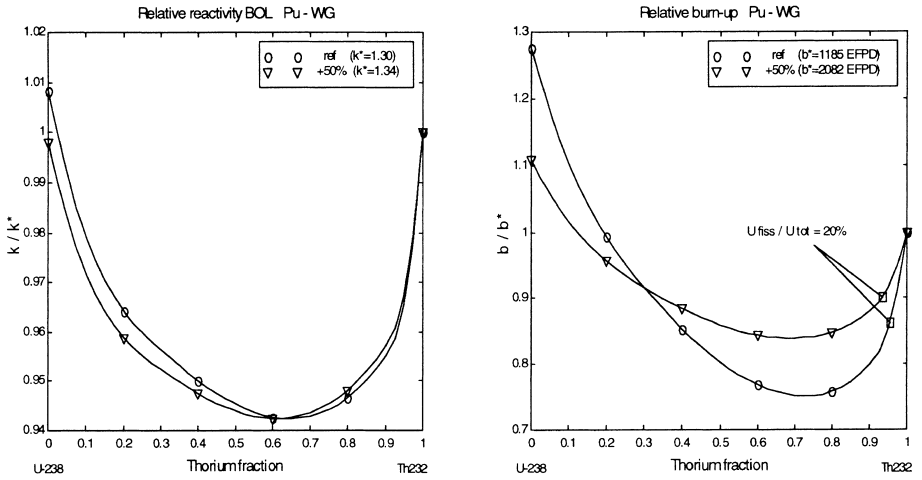

Fig. 4. (a) Reactivity vs. ^{232}Th fraction. Thorium fraction: 0 = pure Pu–U fuel, 1 = pure Pu–Th fuel. (b) Burn-up vs. ^{232}Th fraction. Thorium fraction: 0 = pure Pu–U fuel, 1 = pure Pu–Th fuel.

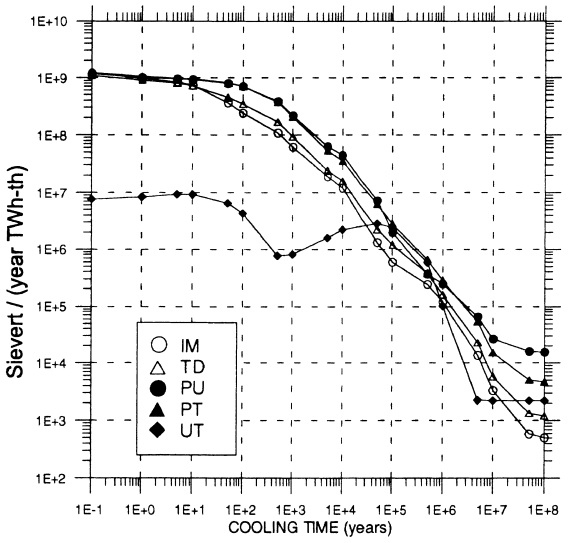

Fig. 5. Radiotoxicity for various reactor-grade plutonium fuels: inert matrix (IM), thorium doped inert matrix (TD), plutonium–uranium (PU), plutonium–thorium (PT), and ^{233}U–thorium (UT)

tonium is present. In Fig. 5 the radiotoxicity curves (Sv y^{-1} TWh_{th}^{-1}) of various fuels are compared: they show a roughly similar trend. In the discharged inert matrix type fuels, the toxicity is lower than for the plutonium–uranium and plutonium–thorium fuels because of a lower initial Pu inventory. A fifth curve, pertaining to a pure thorium-^{233}U cycle (UT) is reported in addition to the curves relative to the four kinds of fuel considered. The UT curve shows a much lower radiotoxicity up to about 10^4 yr, and then merges with that of the other fuels.

6. Concluding remarks

In this work, several nuclear fuel cycles were analysed and discussed, placing the main emphasis on plutonium consumption, reactivity coefficients and proliferation issues. The utilisation of a (Th,Pu) fuel cycle appears advantageous due to the following reasons: (a) the lack of ^{238}U prevents the production of new plutonium thus increasing the fraction of total plutonium burnt; (b) the reactivity coefficients are comparable to those of the standard UO_2 fuel; (c) the fuel shows a good reactivity behaviour as a function of burnup. Moreover, the percentage of burnt plutonium is substantially increased by the reduction of the pellet diameter, the adoption of hollow pellets and the increase of the plutonium content in order to better exploit in situ the produced ^{233}U. Notwithstanding these attractive factors, some drawbacks are also evident in the (Pu,Th) fuel cycle: (i) the high thermal cross sections of plutonium isotopes significantly reduce the worth of control mechanisms such as control rods or soluble boron, thus requiring a different control rod design or the adoption of enriched boron; (ii) the highly fissile ^{233}U is generated during the fuel irradiation and this might pose proliferation problems, although mitigated by the presence of ^{232}U. For this latter problem, the addition of a limited amount of natural uranium (4–7%) eliminates the proliferation concern at the expenses of a modest reduction in fuel burnup.

A great proliferation resistance is achieved with IMF. This fuel consists of PuO_2 dispersed within a carrier matrix composed of inert oxides. In the neutronic simulations standard UO_2 rods and IMF rods were inserted together in fuel assemblies which were uniformly loaded in the core.

An important drawback of this solution is the irregular distribution of the assembly power between IMF pins and UO_2 pins. Due to the fast decrease of the power generation inside the plutonium rods, power peaks are evident in the IMF at BOL while the power requirements from the standard fuel rods increase at EOL. The high reactivity of plutonium bearing fuel also causes peaking factors across the core.

The addition of BPs might be a viable solution to be considered for dampening these two effects. In this particular configuration, the best performing burnable poison is IFBA, which allows one to face the two problems and, at the same time, provides a significant reduction of the content of soluble boron in the core.

The IMF showed very good plutonium annihilation capabilities: more than 98% of the loaded ^{239}Pu was burnt and 73% of the total loaded RG plutonium was consumed. The residual plutonium was quality-poor and thus unattractive for any attempt of recovery and improper use.

The appealing advantage of the thorium fuel is that the existing experience, although limited, indicates an excellent behaviour under irradiation, even better than standard fuel, and a very stable behaviour in deep disposal conditions. For ZrO_2-based IMF there are convincing indications of a similar excellent behaviour, but up to now not substantiated by a thorough in-pile experimental program.

In general, thorium fuel cycle was thoroughly studied in the past, but the strong reduction in nuclear programmes all around the world resulted in a practical stop of the development of the thorium cycle, except in India. Perhaps it is worth recalling Lung opinion that "in view of its potential advantages, the thorium fuel cycle has to be considered again as a promising energy source in, and after, the next century. (...) Burning some of the weapons plutonium could be one interesting way to enter the thorium fuel cycle" [25].

These fuels are also of interest for other applications, namely those involving accelerator-driven systems which carry a strong interest in the present debate about nuclear power. In this context a common program, managed by ENEA, has been recently accepted in Italy.

Acknowledgements

Special thanks are due to Dr Guandalini and Dr Vimercati (CISE) for the continuous assistance in working with ABARTH and SCALE. The CASMO-3 program was used at the ENEL-ATN office; the cooperation of this organization is greatly acknowledged.

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