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Neutronic aspects of inert matrix fuels for application in ADS

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Abstract

Accelerator driven systems may operate on uranium or thorium free fuels. In order to guarantee the stability of such fuels at high temperatures, the use of inert matrices is foreseen. In the present study, safety parameters of 800 MWth ADS cores operating on oxide and nitride fuels with high americium content are investigated for a representative range of pin and core geometries. It is shown that among the inert matrices investigated, chromium yields the lowest void worth, hafnium nitride the highest fission probability for americium and magnesia the highest burnup potential.

1. Introduction

In the Double Strata fuel cycle [1,2], the americium and curium produced in critical power reactors is supposed to be multi-recycled in dedicated minor actinide burners. An advantage of this approach is that handling of these highly active elements is constrained to a very small part of the nuclear power park. The poor reactivity coefficients of minor actinide based fuel, in conjunction with a very small effective fraction of delayed neutrons, however makes safe operation of critical dedicated cores questionable. The introduction of accelerator driven systems (ADS) for the purpose of minor actinide burning therefore appears adequate. The ADS core should operate on a fast neutron spectrum, in order to minimise production of strong neutron emitters like curium and californium. While a substantial part of the technology developed for fast breeder reactors is directly applicable to ADS, the fuel composition, form and state remains to be determined. In the present paper, safety parameters like void worths and coolant temperature coefficients of several fuel candidates are investigated. In addition an estimation of the relative burnup potential pertaining to each fuel type is given.

2. Core model

A single zone ductless core model was used, in order to simplify the study of impact of fuel pin diameter and pin pitch on the safety coefficients. Table 1 summarises the parameters that were kept constant. A start-up core was considered, hence no curium in the fuel. The ratio of plutonium to americium was set to 40/60, in order to provide a minimum reactivity swing [3]. Calculations were made for two liquid metal coolants: sodium and lead–bismuth eutectic. Radial steel reflectors were assumed in both cases. Two representative pin diameters were investigated, and the pin pitch was varied from $P/D = 1.25$ to 2.25. The clad thickness was adjusted to allow for a maximum fission and helium gas pressure of 20 MPa, given a plenum height of 100 cm. In all cases, the fraction of inert matrix was adjusted to obtain a k-eigenvalue equal to 0.97. For oxide fuels a porosity of 10% was adopted, for nitride fuels 15%. Table 2 displays the average linear rating assumed for each fuel. A smaller rating corresponds to a larger number of fuel pins in the core.

3. Method of calculation

The continuous energy Monte Carlo code MCNP4C [4] in parallel mode was used to calculate neutron fluxes, k-eigenvalues and cross sections. The cross section data

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Table 1
ADS core parameters kept constant in the present study

Core power	800 MWth
Spallation target	LBE
Target radius	20 cm
Core height	100 cm
Pu/Am	40/60
k-Eigenvalue	0.97

Table 2
The uranium free fuels investigated here

Composition	Matrix	Form	Rating (kW/m)
Oxide	ZrO_2	Solid solution	15
Oxide	MgO	Composite	25
Oxide	W	Composite	35
Oxide	Mo	Composite	35
Oxide	Cr	Composite	35
Nitride	ZrN	Solid solution	35
Nitride	HfN	Solid solution	35

'Rating' denotes average linear power density.

library used for the present work was ENDFB/VI-8. Each fuel pin was modelled explicitly, in order to obtain a correct leakage contribution to the void worth. A standard deviation of less than 40 pcm in the estimated k-eigenvalues was required, corresponding to calculation times of about two hours per core configuration (using eight Athlon 1.5 GHz processors). It should be noted that there is a considerable uncertainty in the inelastic cross section of lead [5], which may lead to errors in the calculated void worths of LBE cooled cores. The magnitude of this uncertainty is of the order of 1000 pcm, but will not be critical for the conclusions of this study. Further, void worths in power flattened multiple zone cores will differ from single zone cores, as the radial leakage contribution to the neutron balance is suppressed in the latter case. Hence, the void worths here reported should be interpreted relative to one another, rather than in terms of absolute magnitude.

4. Inert matrix fraction

In Fig. 1, the volume fraction of inert matrix required to obtain a k-eigenvalue of 0.97 is displayed as function of pin pitch for oxide fuels and LBE coolant. Since erosion concerns limits the velocity of heavy liquid metal coolants to about 2 m/s, the pin pitch must be increased to obtain a heat removal capacity similar to that of sodium. For an inner pin diameter of 5.0 mm and a clad thickness of 0.36 mm, the inert matrix fraction is found to range from 40 to 60 vol.%, with exception for the case of tungsten. Since fabricability may become a concern for smaller fractions of inert matrix, it seems like the

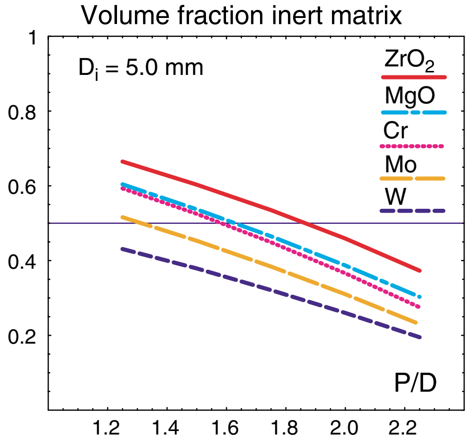

Fig. 1. Volume fraction of inert matrix required to obtain a k-eigenvalue of 0.97 for oxide fuel and LBE coolant. 10% porosity was assumed.

absorption cross section of tungsten is a bit too high for this matrix to be compatible with the adopted Pu/Am ratio.

As the neutron mean free path in sodium is much higher than in LBE, the contribution of absorption in the plenum region to the neutron balance will be larger for identical geometries and fuel compositions. In order to compensate for this the fraction of inert matrix becomes 5–10% lower when adjusting the k-eigenvalue of the core to 0.97.

Increasing the pin diameter will enable a larger fraction of inert matrix to be used. In Fig. 2, the volume fraction of ZrN in nitride fuels is displayed. Increasing the inner clad diameter from 5.0 to 7.0 mm allows for an increase in ZrN fraction by 4–7%. It should be

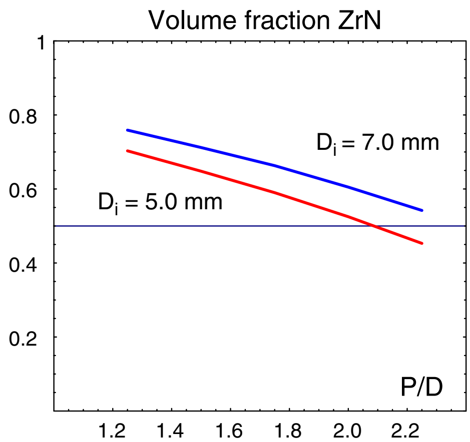

Fig. 2. Volume fraction of ZrN required to obtain a k-eigenvalue of 0.97 for nitride fuel and LBE coolant. 15% porosity was assumed.

emphasized, that in none of the cases investigated, a volume fraction of inert matrix larger than 75% was obtained. If other boundary conditions (fabricability, safety) would require a larger matrix fraction, the Pu/Am ratio would have to be adjusted, with consequenses for reactivity management.

5. Void worths

The lack of a significant Doppler effect in ADS fuels implies that core configurations yielding prompt supercriticality should be avoided. The combination of MA-bearing fuel and liquid metal cooling will typically yield a positive coolant void worth. The choice of coolant, as well as the fuel design however have a significant impact on the magnitude of the void worth. Fig. 3 shows sodium and LBE void worths (for voiding of core and upper plenum) as function of P/D for oxide fuels with zirconia and chromium matrices. As in the case of fast reactors, the void worth is lower for fuels having a high thermal conductivity. The reason for this is that a higher linear rating corresponds to a fewer number of fuel pins in the core and consequently a higher radial leakage in the voided state. The drastic difference in void worth between sodium and LBE is due to the large amount of americium in the fuel. Though the difference is smaller for voiding of the core only, sodium has a clear disadvantage in terms of higher void worth in minor actinide burners.

In Fig. 4, the LBE void worth is compared for the five oxide fuel types investigated. Note that the Cr matrix offers an advantage in terms of a low void worth. The explanation is that chromium has its main inelastic scattering threshold at 1.2 MeV, in contrast to most other nuclides, which have thresholds at around 0.5 MeV. Chromium thus is inelastically transparent for a larger fraction newly born fission neutrons, yielding not only a harder spectrum, but also a smaller shift of spectrum during coolant voiding. Selecting inelastically transparent materials for the fuel matrix hence enables to obtain acceptable void worths for larger cores. The void worth for a nitride fuel in solid solution with ZrN varies with P/D between +1000 and +1500 pcm, much lower than its oxide counterpart. Thus it is mainly the poor thermal conductivity of zirconia that makes it suitable as fuel matrix for ADS (low rating leading to larger number of fuel pins). Substituting ZrN with HfN increases the void worth by a factor of two.

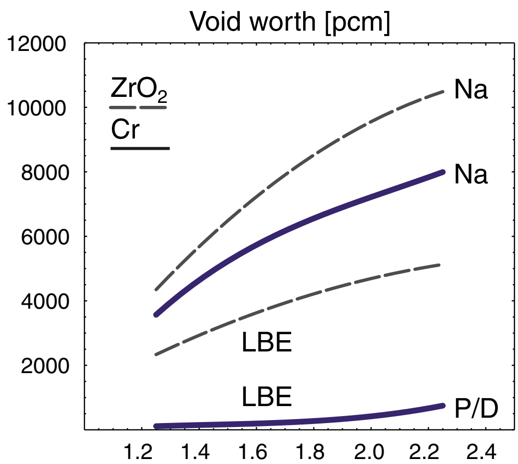

Fig. 3. Void worths pertaining to oxide fuels with zirconia and chromium matrices. An inner clad diameter of 5.0 mm was assumed. The void worth was calculated by voiding the core and upper plenum from coolant. Due to uncertainties in the cross section of lead, the displayed values may be in (absolute) error by up to 1000 pcm.

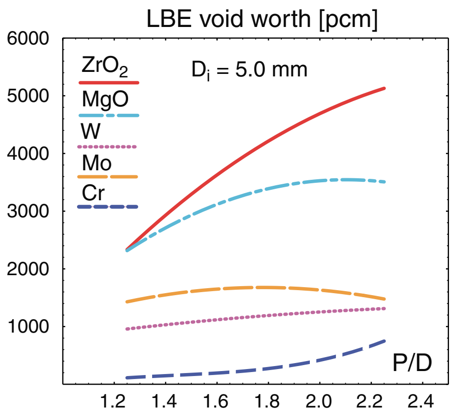

Fig. 4. Void worths pertaining to oxide fuels with ceramic and metallic matrices. Due to uncertainties in the cross section of lead, the displayed values may be in (absolute) error by up to 1000 pcm.

6. Coolant temperature coefficients

While significant temperature feedbacks are not required for normal operation of an ADS, negative feedbacks remain useful in core disruptive accidents, as well as for minimising fluctuations in core power. The Doppler feedback of the ADS fuel is close to negligible, being about −0.05 pcm/K [6]. Among the possible mechanisms to control the core, axial and radial expansion are still active in the ADS. Values of structural material expansion coefficients typically range from −0.2 to −0.5 pcm/K. The coolant temperature coefficient, on the other hand, ranges from +0.2 to +1.0 pcm/K for certain fuels cooled by LBE, as shown in Fig. 5. For sodium cooled ADS cores, the temperature coefficient may be as large as +4 pcm/K, which hardly can be compensated by feedbacks from structural material

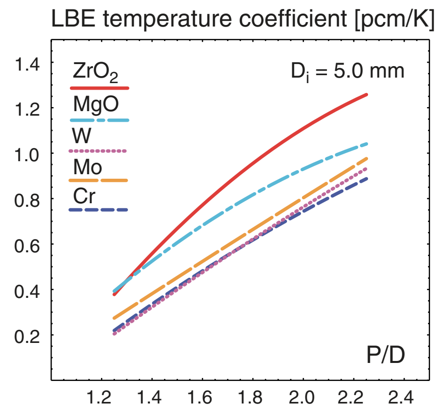

Fig. 5. Lead–bismuth coolant temperature coefficient as function of P/D for oxide fuels.

expansion. Hence introduction of heavy liquid metals or helium gas as coolant for minor actinide burning ADS is well motivated.

7. Cross sections

The neutron spectrum of FBRs varies only marginally from one design to another, due to restrictions set by breeding and neutron economy. In ADS, one has a larger parameter space available for fuel matrix and core design. The less restrictive neutron economy allows for introduction of absorbing matrices, yielding a harder spectrum, or alternatively larger pin pitches, softening the spectrum. Table 3 shows spectrum averaged cross sections and fission probability for Am-241 as function of fuel matrix for the present core model. Note that hafnium nitride gives the highest probability for fission of americium, and consequently, the lowest build-up of curium.

8. Burnup potential

In fast neutron reactors, swelling of fuel cladding and wrapper tubes has been identified as the ultimate limitation to fuel burnup. Steels developed for the purpose of being swelling resistant have enabled to reach doses ranging from 150 DPA (austenitic steels) to 200 DPA (ferritic steels). In order to achieve high burnup of the ADS fuel, the design should provide a high ratio between fission and DPA rates. Since the minor actinides present in the fuel are fissionable by neutrons with energy above 1 MeV, it is of interest to minimise the presence of nuclides with large cross section for inelastic scattering in the fuel while simultaneously suppressing the flux that causes damage (neutrons with $E > 0.1$ MeV).

A simple estimation of the burnup potential B_p of a fuel can be found from the formula:

$$B_p = 1 - \mathrm{Exp}[-\sigma_f \varphi t_{max}], \tag{1}$$

where σ_f is the average fission cross section of fissionable nuclides, φ is the neutron flux, and t_{max} is given by

$$\varphi_{fast} t_{max} = F_{max}. \tag{2}$$

Here, φ_{fast} is the flux of neutrons with energies above 0.1 MeV, i.e. the flux capable of causing radiation damage in the clad, and F_{max} is the fast fluence limit. For ferritic steels 200 DPA roughly corresponds to $F_{max} = 4.0 \times 10^{23}$ n/cm^2 [7]. The fuel averaged fission cross section and the corresponding burnup potential for the ADS fuel candidates here studied are listed in Table 4. These numbers may be compared to the average fission cross section of standard FBR oxide fuel, being 0.33 b, yielding a burnup potential of 20%, if estimated by formula (1). The good agreement with the actual burnup limit may be coincidental, since Eq. (1) contains a bare minimum of physical information. However, the relative burnup potential of different fuels should be possible to predict using this approach.

Table 3
Spectrum averaged cross sections and fission probabilities for Am-241 for the fuels here investigated

Fuel/matrix	σ_f (b)	σ_c (b)	$\sigma_f/(\sigma_f+\sigma_c)$
Oxide/ZrO_2	0.31	1.34	0.19
Oxide/MgO	0.33	1.37	0.20
Oxide/W	0.33	1.10	0.23
Oxide/Mo	0.34	1.09	0.24
Oxide/Cr	0.34	1.17	0.22
Nitride/ZrN	0.35	1.18	0.23
Nitride/HfN	0.38	0.97	0.28

Values are given for LBE coolant, $P/D = 1.75$ and an inner clad diameter of 5.0 mm.

Table 4
Fuel and spectrum averaged fission cross section and burnup potential for the fuels here investigated

Fuel/matrix	σ_f (b)	B_p (%)
Oxide/ZrO_2	0.64	32
Oxide/MgO	0.66	33
Oxide/W	0.63	30
Oxide/Mo	0.64	30
Oxide/Cr	0.65	31
Nitride/ZrN	0.66	31
Nitride/HfN	0.67	30

Values are given for LBE coolant, $P/D = 1.75$ and an inner clad diameter of 5.0 mm.

Due to the comparatively low fission cross section of americium, the burnup potential does not increase in proportion to the fraction of removed U-238. Note that the burnup potential for the fuels with an MgO matrix is higher than for the other fuels. In general, it appears as the inert matrix fuels here studied have a burnup potential ranging from 30% to 33%, which of course only may be realised in full extent if core management succeeds in levelling out power peaking factors.

9. Conclusions

The choice of fuel matrix has a significant impact on safety parameters in accelerator driven systems dedicated to minor actinide transmutation. An oxide fuel with chromium matrix appears to yield the lowest void worth among the fuel types studied in the present work. Strong neutron absorbers like tungsten and hafnium may be used to minimise production rates of curium, but have the disadvantage of increasing the void worth. Hafnium nitride on the other hand, yields the highest fission probability of americium.

The results of the present investigation indicate that chromium is an interesting inert matrix candidate for oxide fuels. It minimises the void worth, while retaining a high fission probability of americium and a reasonably high burnup potential. Fabricability and chemical compatibility do not appear to be major problems, but swelling at high neutron doses and phase separation at elevated temperatures may possibly be issues of concern.

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