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Safety Characteristics of Candidate Oxide Fuels for Accelerator Driven Transmuters

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The FUTURE project (Fuels for the Transmutation of Trans-Uranium Elements), established within the 5th European Framework Programme on Partitioning and Transmutation, has the main aim of designing dedicated fuels for an Accelerator Driven Transmuter (ADT) based on well known oxide fuel technology. This paper presents the work that has been carried out to assess the basic feasibility of ADT cores utilising the candidate fuels selected within the FUTURE programme. Two particular aspects have been addressed in detail, the impact and behaviour of the different inert matrix materials, and an evaluation of their safety characteristics as a function of fuel irradiation and fuel isotopic content. Results for core performance and safety parameters are presented and discussed. These studies have been performed by Serco Assurance within the context of the European FUTURE programme and are sponsored by BNFL.

KEYWORDS : FUTURE, ADT, oxide fuel, inert matrix, CERCER, CERMET.

1. Introduction

The objective of the FUTURE project is to study the feasibility of uranium free oxide based fuels to be irradiated in an accelerator driven transmuter. Dedicated oxide fuels intended for ADT systems, consisting of innovative oxide compounds in a solid solution which is diluted in an inert matrix, have not so far been developed in Europe or in the international scientific community. The principal innovation is related to the high minor actinide content and the absence of a fertile component. These characteristics have a significant impact on the fuel and plant safety and also impose limitations on the choice of fuel, matrix and core design.

Various candidate fuel types for accelerator driven transmuters have been studied in parallel within the FUTURE project :

- a solid solution such as (Pu, Am, Cm, Zr)O_{2-x} or (Pu, Am, Cm, Th)O_{2-x},
- a CERMET fuel (Pu, Am, Cm)O_{2-x} with an inert Mo^{92} (2), W, Cr or V matrix,
- a CERCER fuel (Pu, Am, Cm)O_{2-x} with an inert MgO matrix.

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(2) Depleted Mo is selected instead of Mo to avoid Tc production during irradiation.

To be able to provide a recommendation on ADT fuel design a neutronic investigation has been undertaken to assess the basic feasibility and safety characteristics of ADT cores utilising the candidate fuels. Two particular aspects have been addressed in detail, the impact and behaviour of the different inert matrix materials, and an evaluation of their safety characteristics as a function of fuel irradiation and fuel isotopic content.

2. Fuel Cycle Scenario and ADT Design

The development of oxide fuels intended for ADT systems depends strongly on hypotheses concerning the fuel cycle scenario and the ADT design characteristics. As no European reference fuel cycle scenario nor ADT design are available at present a series of hypotheses, based on published international studies, were defined at the beginning of the FUTURE project. The ADT core design characteristics used during these studies are summarised in Table 1.

Table 1 Reference ADT Core Design Parameters

Core Power	800 MWth
Coolant	Lead Bismuth Eutectic (LBE)
Spallation Target Power	20 MW
Beam power	< 40 MW
Proton Beam Energy	1 GeV
Beam Current	40 mA
Neutron Source Yield	25 neutrons/proton (1 GeV)
Target Radius	20 cm
Core Height	100 cm
Lower Plenum Height	100 cm
Upper Plenum Height	50 cm
K-eigenvalue (start of cycle)	0.970
Proton Beam Variation	0.940 < K-effective < 0.970
Inlet Coolant Temperature	300 °C (nominal power)
Outlet Coolant Temperature	500 °C (nominal power)

ADT systems operate in a dedicated stratum and are fed with plutonium and minor actinides arising from a first stratum devoted to electricity production. Composition of the first stratum, and therefore of the feed fuel for the dedicated stratum, depends on national policy. Since no consensual European scenario is presently available it was decided to use plutonium and minor actinide fuel issued from three different scenarios : a UOX, MOX and Advanced Plutonium (AP) cycle. Each fuel has undergone a once through irradiation in an LWR to reach a discharge irradiation of 50 GWd/tHM after a cycle length of 18 months, followed by seven years of cooling. The isotopic composition of the fissile materials is given in Table 2.

Table 2 Feed Fuel Isotopic Composition at the Start of the 1st Cycle (weight %)

	Feed Fuel Cycle Scenario		
	UOX	MOX	AP
Plutonium			
Pu^{238}	3.49	5.05	5.80
Pu^{239}	51.87	37.91	11.75
Pu^{240}	23.81	30.31	21.69
Pu^{241}	12.90	13.21	9.61
Pu^{242}	7.91	13.51	51.15
Americium			
Am^{241}	57.57	66.67	29.81
Am^{243}	42.43	33.33	70.06
Curium			
Cm^{244}	90.90	90.00	87.39
Cm^{245}	9.10	10.00	10.09
Cm^{246}	0.00	0.00	2.03

3. Calculational Methods

All of the core configurations evaluated during this study have been modelled neutronically using Version 1.2 of the European fast reactor neutronics code scheme ERANOS along with the JEF2.2 nuclear cross section data library [1]. Broad group resonance shielded cross sections have been produced for each core material using the ECCO cell code. A fine group slowing down treatment is combined with the sub group method within each fine group to provide an accurate description of the reaction thresholds and resonances for each type of critical and sub-critical sub-assembly. Whole core flux calculations have been carried out using the TGV/VARIANT variational nodal transport theory method with a P_3 angular expansion of the flux and a P_1 anisotropic scattering approximation. Neutronics calculations have been performed at the start of life, and for the first and subsequent burnup cycles during the approach to equilibrium. Given the significant minor actinide content in the fuel, shielded microscopic cross section data has been recalculated at the beginning of each cycle.

4. Fuel Characterisation

The volume fraction of inert matrix in the fuel is driven by the core neutronic behaviour which is significantly affected by the type of matrix. To obtain a k-effective value of 0.970 at the start of life the volume fraction of inert matrix has to be adjusted, as illustrated in Figure 1 for the MOX feed fuel composition. The inert matrix volume fraction is also dependent on the fuel fabrication process. The sintering temperatures of the inert matrices and the actinide oxides are very different. The inert matrices require a low sintering temperature whereas the actinide oxides require a high temperature. This results in a large decrease in the fuel density as the actinide oxide content increases. Furthermore, a high actinide oxide volume fraction also provides for a significant reduction in the fuel thermal conductivity.

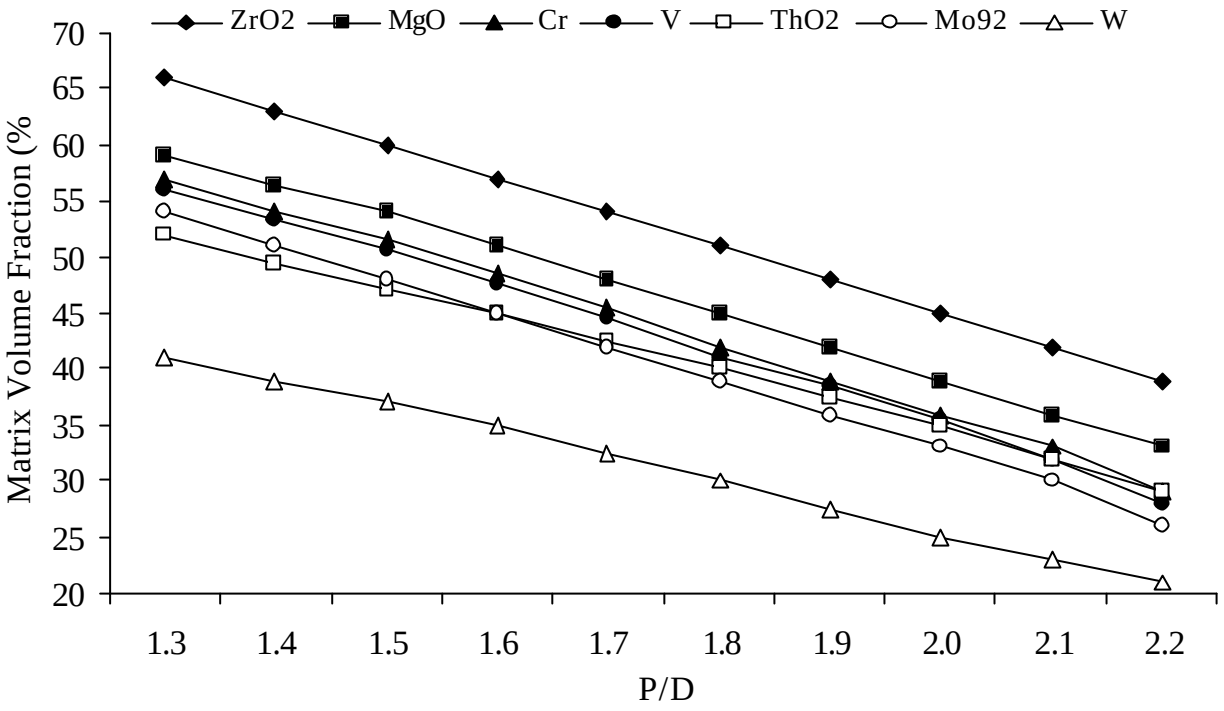

Fig.1 Matrix Volume Fraction as a Function of Pitch-to-Diameter Ratio for D=5.0mm

It is therefore desirable to keep the inert matrix volume fraction as high as possible. Neutronically transparent matrices such as ZrO_2, MgO, V and Cr allow a composite fuel with the highest inert matrix volume fraction (> 50%). However Cr and V matrices are considered undesirable due a possible eutectic reaction with $(Pu, Am, Cm)O_2$ at low temperature. For absorbing matrices such as W the matrix content is very low (< 40%). For these refractory metals the inert matrix volume fraction could be increased provided that the fuel pin diameter (D) increases and/or P/D decreases. However a compromise has then to be found between the fuel performance and the pressure drop or coolant void coefficient which provide significant constraints on the choice of D and P/D. Absorbing metallic matrices such as W are therefore not considered viable for the FUTURE candidate fuels.

Another key parameter affecting the choice and content of the inert matrix in the fuel is the coolant void worth, which is usually a positive reactivity effect for liquid metal cooled ADT cores. In this case the coolant voiding effect is mitigated by the high LBE boiling point. However, core voiding could be triggered by breached pins and the subsequent helium and fission gas blow down from the core. The calculated void worth for the different FUTURE composite fuels, assuming a single zone core with the MOX feed fuel, is shown in Figure 2.

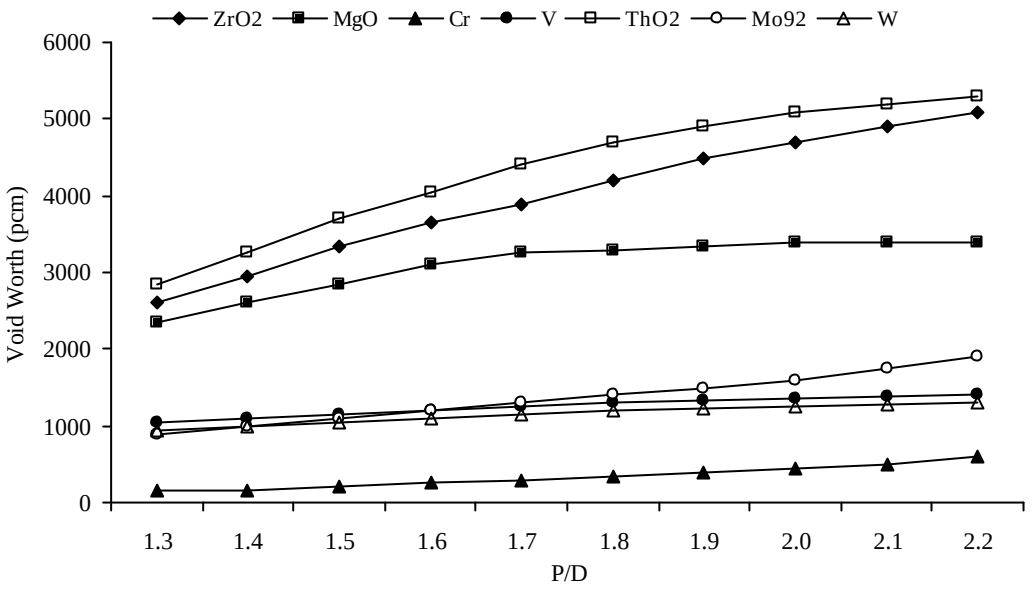

Fig.2 Coolant Void Worth as a Function of Pitch-to-Diameter Ratio for D=5.0mm

The coolant void worth is significantly reduced when metallic matrices (by order of preference : Cr then Mo^{92}, W and lastly V) are utilised as their high thermal conductivity results in a smaller core size with increased neutron leakage. Conversely, CERCER fuels yield the highest void worth due to the larger core size. MgO based fuels provide acceptable void worths whereas ZrO_2 and in particular ThO_2 fuels provide high values due to their low threshold for inelastic scatter.

On the basis of these fuel characterisation studies it is evident that three candidate fuels merit further detailed investigation : a solid solution with a ZrO_2 matrix, a CERCER fuel with a MgO matrix, and a CERMET fuel with a Mo^{92} matrix. Although ZrO_2 based fuels are known to exhibit poor thermodynamic and safety performance, ZrO_2 is currently considered to be a reference benchmark fuel for which a significant amount of knowledge is already available.

5. Fuel Performance

Core configurations have been defined for the three FUTURE composite fuels selected for further study. The core average linear rating was kept below 160 W/cm for the ZrO_2 fuel due to its low thermal conductivity. For the MgO and Mo^{92} fuels, which exhibit a higher thermal conductivity, the average linear rating was restricted to 250 W/cm. These limitations have a direct impact on the core size : the higher the thermal conductivity and linear rating, the smaller the core size. Therefore, cores containing 48048 and 28392 pins were adopted for the ZrO_2 and MgO/Mo^{92} fuels respectively. Fuel zoning is required in ADT cores in order to flatten the power distribution which is perturbed due to the presence of the spallation target and neutron source at the core centre.

To ensure that the radial power peaking factor is acceptable (< 1.5), three radial zones with a differing matrix volume fraction are required occupying approximately 25%, 50% and 25% of the core volume respectively. For the ZrO_2 and MgO composite fuels the fuel pellet diameter is 5.85 mm, and for the Mo^{92} fuel the pellet diameter is 5.80 mm. In all cases the clad inner diameter is 6.0 mm and the clad outer diameter is 6.8 mm. A P/D of 1.6 has been chosen as it provides acceptable characteristics for both thermodynamic and safety behaviour. The inert matrix volume fraction for each of the three candidate fuels, providing a k-effective value of 0.970 at the start of life, with the plutonium/americium/curium ratio fixed at 40/50/10, is given in Table 3.

Table 3 Matrix Volume Fraction for the Selected Candidate Fuels (volume %)

Radial Zone	ZrO_2			MgO			Mo^{92}		
	UOX	MOX	AP	UOX	MOX	AP	UOX	MOX	AP
Inner	68	60	34	64	56	30	63	55	29
Middle	62	58	29	56	52	23	55	51	22
Outer	54	48	17	45	40	9	44	39	8

5.1 Start of Life

These defined conditions have been used to calculate the core performance and safety parameters for the selected candidate fuels at the start of life, as shown in Table 4. The values for the peak linear rating and the radial power form factor satisfy the design criteria for all of the different fuel types. The coolant void worth is generally acceptable although significantly higher values are attained for the ZrO_2 matrix fuel.

Table 4 Core Performance and Safety Parameters for the Selected Candidate Fuels at the Start of Life

	ZrO_2			MgO			Mo^{92}		
	UOX	MOX	AP	UOX	MOX	AP	UOX	MOX	AP
Peak Rating (W/cm)	211	221	236	319	329	335	325	339	346
Average Rating (W/cm)	152	162	168	251	262	269	258	269	273
Radial Power Form Factor	1.25	1.36	1.41	1.22	1.35	1.39	1.26	1.38	1.45
Void Worth (pcm)									
Core	5152	6235	9275	3751	4840	7846	2455	3548	6576
Core + Plenum	4648	5753	8716	3207	4327	7317	1948	3039	5996
Fuel Doppler (pcm/K xE-03)	28.6	3.54	0.63	-0.88	-1.54	-49.3	-8.41	-17.4	-33.5
Delayed Neutron Fraction (pcm)	193	190	187	186	182	179	200	198	194
Prompt Neutron Lifetime (E-07 s)	6.13	5.89	3.33	5.26	4.44	2.19	4.14	3.85	1.76

It can be seen that the void worth is strongly dependent on the fuel isotopic vector. A more degraded fuel quality leads to a more positive void reactivity worth. A loss of coolant shifts the neutron spectrum to energies above the fission threshold of the higher plutonium and minor actinide isotopes, which due to their increased importance, results in a large positive net void worth. These results also illustrate the low Doppler coefficient that exists for all the FUTURE candidate fuels, due to the absence of the significant contribution that usually arises from U^{238} capture. The delayed neutron fraction is less than 200 pcm for all of the FUTURE candidate fuels in the start up core. Again, this is due to the absence of U^{238} and the consequent reduction in the effective yield of delayed neutrons. It can be noted however that for an ADT under operational conditions the effective delayed neutron fraction and the prompt neutron generation time do not play an essential role in reactor control.

5.2 First Cycle

For safety concerns it is essential that the core reactivity swing does not exceed 3000 pcm to ensure that the value of k-effective is maintained below unity, and within the constraints imposed by the accelerator current. Such a requirement is met providing that the plutonium/minor actinide ratio is kept below unity. Addition of plutonium is necessary to balance the reactivity increase that occurs due to americium and curium transmutation. The appropriate fraction is dependent on the neutron spectrum as well as the plutonium and minor actinide quality. Table 5 shows the reactivity swing for the selected FUTURE candidate fuels during the first irradiation cycle for two different plutonium/americium/curium ratios.

Table 5 Reactivity Loss for the Selected Candidate Fuels during the First Irradiation Cycle (pcm)

	ZrO_2			MgO			Mo^{92}		
Pu/Am/Cm Ratio	UOX	MOX	AP	UOX	MOX	AP	UOX	MOX	AP
40/50/10	3162	1567	39	3298	1633	42	3535	1754	44
50/42/8	8455	4188	117	9296	4607	119	9964	4948	125

For a constant plutonium volume fraction, MgO and Mo^{92} yield a harder neutron spectrum and therefore a larger reactivity loss since the probability of americium fission is increased. The plutonium content, and the plutonium and minor actinide quality, also have a significant influence on the reactivity loss. The reactivity swing is increased by almost a factor of three as the plutonium content and quality is increased. The reactivity swing for the most degraded AP feed fuel is negligible. For most of the FUTURE candidate fuels a 40% plutonium content is sufficient to provide an acceptable minimum reactivity swing. However, for the AP feed fuel an increased plutonium content of 50%, or even 60%, is feasible. This brings the benefit of improving the core performance and safety characteristics, in particular by reducing the coolant void coefficient which is large and positive for the AP quality fuel.

The transmutation performance in the first cycle is relatively independent of the matrix type, with an americium and plutonium consumption rate of approximately 120 kg/TWhe and 20 kg/TWhe respectively for a plutonium/americium/curium ratio of 40/50/10. This suggests that ADT cores fuelled with FUTURE composite fuels are capable of significant minor actinide transmutation. There is however, a curium production of the order of 12 kg/TWhe in the first cycle. The curium production increases as the plutonium and minor actinide vector becomes more degraded, approaching 40 kg/TWhe for the AP feed fuel.

5.3 Equilibrium Cycle

An analysis of the equilibrium ADT core is important as it will define the long term costs of fuel fabrication and reprocessing, which are mainly driven by the curium inventory. Calculations have been performed to derive an equilibrium fuel loading for the selected FUTURE composite fuels. It is possible that the reprocessing of ADT fuels may be undertaken using either hydrochemical or pyrochemical techniques. Therefore two different strategies have been followed. Hydrochemical reprocessing assumes five years of storage and reprocessing and two additional years of cooling before core loading, while pyrochemical reprocessing assumes two years of storage and reprocessing followed by two additional years of cooling before core loading. The equilibrium composition of the selected FUTURE composite fuels, assuming hydrochemical reprocessing, is shown in Table 6.

Table 6 Equilibrium Isotopic Compositions of ADT Cores Fuelled with the Selected Candidate Fuels (weight %)

	ZrO_2			MgO			Mo^{92}		
	UOX	MOX	AP	UOX	MOX	AP	UOX	MOX	AP
Pu									
Pu^{238}	11.76	13.24	7.54	10.59	12.17	6.42	9.98	11.44	6.73
Pu^{239}	4.12	4.07	4.65	6.34	6.25	6.89	8.51	8.36	8.88
Pu^{240}	18.44	16.76	21.22	17.23	15.59	20.07	16.29	14.63	19.07
Pu^{241}	1.17	1.23	0.85	1.12	1.18	0.82	1.06	1.13	0.76
Pu^{242}	7.14	6.49	7.67	6.19	5.54	6.73	5.69	5.12	6.31
Total Pu	42.63	41.79	41.93	41.47	40.73	40.93	41.53	40.68	41.75
Am									
Am^{241}	22.54	22.76	18.33	17.41	17.63	13.24	25.43	25.71	21.29
Am^{242m}	1.43	1.54	0.71	1.27	1.33	0.68	1.17	1.24	0.69
Am^{243}	14.41	11.32	21.02	13.86	10.76	20.32	12.04	9.07	19.36
Total Am	38.38	35.62	40.06	32.54	29.72	34.24	38.64	36.02	41.34
Cm									
Cm^{243}	0.23	0.26	0.11	0.20	0.24	0.10	0.18	0.23	0.10
Cm^{244}	9.17	8.85	10.13	13.71	13.39	14.73	7.75	7.39	8.69
Cm^{245}	3.11	2.84	1.17	3.84	3.56	1.92	2.23	1.94	0.99
Cm^{246}	0.96	0.93	0.76	1.31	1.27	1.01	1.37	1.33	1.03
Total Cm	13.47	12.88	12.17	19.06	18.46	17.76	11.53	10.89	10.81

Equilibrium is reached after about 16 cycles or 100-120 years of operation. For a minor actinide fuelled core the approach to equilibrium leads to a significant change in the fuel isotopic composition from one cycle to another. The isotopic change is dependent on the neutron spectrum, the core cooling time, and the initial feed fuel isotopic composition. It can be seen that while the plutonium fraction remains approximately constant during the approach to equilibrium, the isotopic vector degrades significantly. Curium is built up from the irradiation of americium and represents 30-40 % of the minor actinide fraction. The more absorbing matrices, such as Mo^{92}, minimise the curium build up by hardening the neutron spectrum. Conversely, ZrO_2 and MgO tend to contribute to the curium build up due to increased inelastic scatter providing a softer neutron spectrum. The curium content is also significantly affected by the choice of reprocessing scenario. Pyrochemical reprocessing, with its shorter cooling time, reduces the amount of curium decay and results in double the amount of Cm^{244} in the equilibrium fuel load due to its relatively short half life (~18 years).

As for the first irradiation cycle, the transmutation performance in the equilibrium cycle is relatively independent of the matrix type. The plutonium consumption, at 9 kg/TWhe is half of that achieved in the first cycle due to the degradation of the plutonium isotopic vector. Similarly, the americium consumption is reduced to about 80 kg/TWhe due to the reduced americium content in the equilibrium fuel load. At equilibrium, curium is consumed, at a rate of 10 kg/TWhe. This indicates that ADT cores, fuelled with FUTURE composite fuels, are capable of consuming the curium arising from a first stratum of electricity producing reactors. The core performance and safety parameters for the selected candidate fuels at equilibrium are shown in Table 7.

Table 7 Core Performance and Safety Parameters for the Selected Candidate Fuels at Equilibrium

	ZrO_2			MgO			Mo^{92}		
	UOX	MOX	AP	UOX	MOX	AP	UOX	MOX	AP
Peak Rating (W/cm)	217	229	241	324	333	340	331	346	352
Average Rating (W/cm)	153	165	171	255	265	271	261	272	279
Radial Power Form Factor	1.26	1.34	1.44	1.24	1.35	1.43	1.29	1.41	1.47
Void Worth (pcm)									
Core	5563	6650	9693	4134	5223	8231	2829	3917	6945
Core + Plenum	5064	6158	9121	3629	4749	7734	2341	3438	6395
Fuel Doppler (pcm/K xE-03)	-4.47	-16.34	-35.6	-11.3	-21.3	-41.6	-27.3	-41.3	-63.3
Delayed Neutron Fraction (pcm)	158	156	154	156	151	148	163	156	152
Prompt Neutron Lifetime (E-07 s)	6.33	6.01	4.17	6.54	6.45	5.56	6.32	4.17	3.36

The coolant void worth is now between 500-600 pcm more positive than that observed in the first cycle. This is due to the degraded fuel isotopic vector, and the increased proportion of the higher plutonium and curium isotopes in the fuel. Although the Doppler coefficient increases and becomes more negative in magnitude during the approach to equilibrium, due mainly to the softer neutron spectrum, the Doppler feedback is still a negligible effect. Also, at equilibrium the effective delayed neutron fraction has reduced to just over 150 pcm due to the reduction in the plutonium content in the fuel.

6. Conclusion

The FUTURE project, established within the 5^{th} European Framework Programme on Partitioning and Transmutation, has the objective to design dedicated fuels for an Accelerator Driven Transmuter (ADT) based on oxide fuel technology. ADT fuels studies within the FUTURE project include a solid solution such as (Pu, Am, Cm, Zr)O_2 or (Pu, Am, Cm, Th)O_2, a CERMET fuel with an inert Mo^{92}, W, Cr or W matrix, and a CERCER fuel with an inert MgO matrix.

During the first part of the FUTURE project a neutronics analysis has been undertaken on a 800 MWth Pb-Bi cooled ADT core design to evaluate the impact of the fuel composition on core performance and safety. This has allowed the definition of the main fuel characteristics such as the maximum acceptable volume content of the inert matrix and the plutonium to minor actinide ratio in the fuel. The fuel neutronic behaviour has also been determined as a function of fuel irradiation and isotopic content. It is shown that the safety behaviour of these dedicated fuels is strongly dependent on the fuel isotopic composition. This can change significantly according to the fuel cycle scenario and the strategy employed during the approach to equilibrium.

It is evident from the results of this analysis that three fuels merit further investigation : a solid solution with a ZrO_2 matrix, a CERCER fuel with a MgO matrix, and a CERMET fuel with a Mo^{92} matrix. Detailed studies of irradiation and safety performance under design basis and design extension conditions will be undertaken during the second part of the FUTURE project.

Acknowledgements

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