

# ASSESSMENT OF THE TRANSMUTATION CAPABILITY OF AN ACCELERATOR DRIVEN SYSTEM COOLED BY LEAD BISMUTH EUTECTIC ALLOY

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## ABSTRACT

Any nuclear fast reactor is able to burn and transmute minor actinides (MA), but the amount of MA content has to be limited to a few percent to avoid unfavourable consequences on the coolant void reactivity, Doppler effect, and delayed neutron fraction, and therefore on the dynamic behaviour and control. Accelerator driven system is instead able to safely burn and/or transmute a large quantity of actinides and Long-Lived Fission Products (LLFP), as it does not rely on delayed neutrons for reactor control in normal or accident conditions.

This paper summarizes and compares the results of neutronic calculations aimed at evaluating the transmutation capability of subcritical cores cooled by Lead-Bismuth Eutectic alloy and loaded with assemblies based on (Pu, Am, Np, Cm) oxide dispersed in a molybdenum metal or magnesia matrices. It also compares the thermo-mechanical behaviour of these innovative fuels versus some key parameters, namely gas release and pellet swelling.

The detailed neutronic calculations were performed with ERANOS code, whereas the sensitivity analysis was carried out by a special ERANOS Procedure, so-called MECONG. The behaviour of fuels pin during the cycle was investigated by using TRANSURANUS code, appropriately modified.

The performed analysis shows a good compromise for both fuels between transmutation and core performance at increasing of the core power and the use of CERMET fuels may present some advantages.

**Key Words:** Accelerator Driven System, Accelerator Driven Burners, Transmutation, MA-based fuel

## 1 INTRODUCTION

The reduction of LLFP and MA from spent fuel is a key point for the public acceptability of nuclear plants.

The current fuel cycles (“once-through” or “Pu burning in LWRs”) do not resolve this issue. The former, besides being a burden for the future generations mainly due to long-term radio-toxicity of MA to be sent to the Deep Geological Repository, is also unacceptable from the point of view of sustainable nuclear energy development (using resources efficiently) and for the long-term heat source also generated by fission products (cesium and strontium) that contributes to reduce the storage capacity of the repository. The latter allows to increase the storage capacity by a factor of five recycling plutonium and low-enough fraction of higher isotopes and to take partially advantage of energy content, being at present the number of

recycles limited (not more than two) because of the build-up of even-numbered plutonium isotopes and other non-fissile, alpha-emitting actinides that renders the recycled product both harder to work and less useful as fuel, but the content of MA and higher degraded plutonium has to be sent to repository or, in alternative, accumulated and stored until other reactors are available to burn them. These goals can be achieved only with alternative scenarios that foresee the deployment of fast reactors to achieve a fuel cycle closure for all actinides on the long-term goal or to minimize the long-term radio-toxicity of high-level waste as well as to maximize the energy extraction from fuel (double strata). The latter option consists in a first layer comprehending LWRs (and possibly fast reactors), burning U and Pu, and a second layer comprehending MA (and possibly Pu) subcritical burners driven by accelerator (ADT), and/or critical burners. The use of ADT rather than critical systems in the second layer is preferable for safety considerations, even if ADT should introduce extra costs, in particular connected with the accelerator capital, operation, and maintenance, besides the plant efficiency penalty. In fact a high amount of MA content has unfavourable consequences on the coolant void reactivity, Doppler effect and delayed neutron fraction, and consequently on the dynamic behaviour and control of nuclear plants. Thanks to subcriticality ADT can be safely operated with fuel with a high MA content and can facilitate tasks that would be difficult or inefficient in critical systems.

The development of subcritical burner core requires also the development of MA-based fuels. A suitable content of plutonium and MA, a multiple recycling and the possibility of reprocessing are requirements of the key importance for a “double strata” strategy or those that foresee the deployment of fast reactors. Among the various fuel technologies available U-free oxide composite fuels - ceramic-ceramic (CERCER) and ceramic-metal (CERMET) - have been considered at European level, as knowledge on the plutonium and americium-based oxide targets and fuels reprocessing, fabrication and handling was already partially available or/and has been acquired, in particular through the European projects, such as FUTURE [1] and EUROTRANS (AFTRA domain) [2]. The principal innovation of these fuels is related to the high MA (Am, Np and Cm) content and the absence of  $U^{238}$  (to avoid Pu production). Pu is, instead, added to control the core reactivity and the core safety behaviour. Moreover they exhibit matrices that have the double role of giving a fuel stability and enhancing the thermal conductivity of the composites fuels, which otherwise would have led to a high operating temperature given the relatively low thermal conductivity of MA oxide materials. Another issue related to these fuels is the lack of knowledge about the physical and mechanical properties under irradiation, such as thermal conductivity, etc.

As the technical feasibility of a subcritical burner must be demonstrated and the development of innovative fuel for MA-burner reactors is a time consuming process that requires at least ten years between initial laboratories studies and industrial qualification, EC has promoted studies on partitioning and transmutation oriented to design a first-kind of ADT since IV Framework Programme [1, 2, 3, 4]. In particular preliminary scoping analyses aimed at evaluating the transmutation capability of an experimental ADT (XADT-80) charged with CERCER fuel (Pu and MA dispersed in a magnesia matrix) at varying of selected parameters were performed during the PDS-XADS project [5] in order to provide useful information for the EFIT design [2]. This activity continued in domestic programmes and was also extended to CERMET fuels.

This paper summarizes the results of neutronic calculations performed within the above-mentioned project and domestic programmes for evaluating the transmutation capability of subcritical burner cores cooled by Lead-Bismuth Eutectic alloy (LBE) and loaded with assemblies based on (Pu, Am, Np, Cm) oxide dispersed in a molybdenum metal or magnesia matrices. It also compares the thermo-mechanical behaviour of these innovative fuels in

function some parameters, namely gas release and pellet swelling.

## 2 TRANSMUTATION CAPABILITY ASSESSMENT

The evaluation of transmutation capability for a core loaded with MA-based assemblies was partially performed within the PDS-XADS project and domestic programmes. Two types of fuel were considered: U-free CERCER with a magnesia matrix and CERMET with molybdenum matrix enriched in Mo92 (Mo92 =92.7%).

Three kinds of analyses were carried out using the deterministic ERANOS modular system code or its special procedure MECONG: 1) the density power of the reference core (XADT-80) [5] was doubled without varying the core dimensions; 2) the power size was raised of 5 times (400 MWth), modifying the dimensions (radius and height) and enrichment of the two fuel regions so that to maintain the same dimensional ratio and power density of XADT-80; 3) the fuel rows number was increased in order to achieve a power size of 300 MW and a maximum power rating of 250W/cm maintaining the total number of SAs constant (288). The diameters of the two fuel zones were varied consequently and the active length was increased by 12 cm.

The first two cases were analysed by MECONG, as it allows simplified and fast evaluation of neutronic parameters along the cycles; the third one was studied with ERANOS code for investigating also the influence of more detailed calculations, e.g. number of special fission products, neutronic equations.

Furthermore the following constraints derived from the accelerator, MA-dedicated fuels and the core itself were taken into account:

a) the maximum values of beam energy and current has been assumed equal to 1 GeV and 20 mA, as the specific neutron yield for energy higher than 1 GeV reaches an asymptotic trend of about 32-34 neutrons per proton with LBE as target material and the technological feasibility of components needed to obtain these values, such as the injector, elliptical cavities and RF power system, is assured by the significant experience acquired on the fabrication and operation of similar components realized for machines dedicated to physics experiments, even if the performances required are different (high values of energy and current for limited length). Therefore the operating values must be fixed taking into account that they affect the technological feasibility in terms of reliability and availability of HPPA-class machine, the doses related to beam losses during both normal and accidental operation, the beam power release in the target spallation zone and the spallation source;

b) fuel performance during the cycle mainly depends on: **a)** pellet swelling (maximum value assumed 10%); **b)** helium production and gaseous release that may have consequences on the mechanical cladding strength (maximum pressure value equal to 50 bar to limit the creep of cladding material); **c)** allowed fuel pin residence time ( $\leq 40\%$  of heavy atoms in the hottest rod); **d)** fuel cladding damage ( $\leq 100$  dpa) and **e)** degradation of physical and mechanical properties under irradiation.

c) to cope with the general deterioration of safety parameters of these fuels during the cycle, the core has to remain subcritical under any foreseeable occurrence pertaining to DBC (Design Basis Conditions) and DEC (Design Extension Conditions);

d) a limit of 45 kg/TWh<sub>th</sub> exists between the installed power and the optimum transmutation capability, as all fissionable plutonium and MA isotopes produce about 200 MeV per fission.

The reference subcritical burner core (XADT-80) has the same annular configuration around the target system and thermal power (80 MW) of experimental accelerator driven system (XADS-80) [5]. It consists of 120 MA-dedicated assemblies (SAs), whose 42 inner with a lower enrichment and 78 outer with a higher enrichment (average enrichment equal to about 35%) and 168 dummy reflector SAs, Fig. 1. Each SA has the same geometry of XADS-80 MOX SA [6] and consequently it contains 90 pins with an outer diameter of 8.5 mm and an active length of 87 cm. The cladding material is 9Cr1Mo grade 91 (French designation T91).

## 2.1 Hypotheses

Several hypotheses were made in a pragmatic way in order to limit the calculations to be performed.

A multi-recycling scenario of 10 cycles, each lasting 930 days, was considered.

As the source importance does not change significantly at increasing the energy of proton to 800 MeV (multiplication factor for a 800 MeV proton beam is equal to 29.66, whereas for 600 MeV one is 29.65), the neutron external source was assumed equal to XADS-80 one (14.98 neutrons per incident proton).

U-free fuels based on (Pu, Cm, Am)O<sub>2</sub> dispersed in a MgO matrix (CERCER) or a molybdenum matrix enriched in Mo92 (CERMET) was selected and the volumetric fraction was set to the minimum allowed value (40%). The isotopic composition of MA-based fuel at BOC of first cycle is reported in Table I.

In order to ensure a safe behaviour acceptable also from the thermo-hydraulic point of view the power rating was limited to 250 W/cm. That should assure the hottest fuel temperature is lower than the design limits for fuel and cladding ( $T_{\text{fuel}} = 2350$  K for CERCER and 2146 K for CERMET;  $T_{\text{cladding}} = 823$  K).

**Table I. Fuel Isotopic Composition normalized to 100%**

Isotope	%	Isotope	%
U234	0.0040	Pu242	0.4362
U235	0.5973	Am241	27.8434
U236	0.0057	Am242m	0.2278
Np237	0.0067	Am243	25.0035
Pu238	0.0938	Cm243	0.0784
Pu239	23.7489	Cm244	10.9484
Pu240	8.3569	Cm245	1.6345
Pu241	0.8887	Cm246	0.1257

## 2.2 Computer codes and models

The evolution of the neutronic parameters (burn-up rate, flux, core reactivity, etc.) along the cycles was calculated by means of the deterministic ERANOS modular system code version 2.0 using a special procedure called MECONG that allows to study simplified two-dimensional RZ geometries and to analyse multi-recycling scenarios. The cross-sections at 33 energy groups were obtained with a homogenous geometrical cell description using the cell code ECCO version 6 and the ERALIB1 library, an updating of JEF 2.2 point wise library that includes 279 nuclides at 1968 energy groups for different temperatures and orders of scattering anisotropy. The cylindrical approximation, instead of real hexagonal one, has a

small effect on the results, as confirmed by heterogeneous ERANOS calculations.

The detailed neutronic calculations were instead performed with ERANOS code for the first cycle. In this case the cross-sections were calculated as above-mentioned using a detailed heterogeneous geometrical description of the fuel assemblies. Successively each set of heterogeneous cross-sections was homogenized all over the assembly to preserve the total reaction rates and then collapsed to 33 energy groups. The core calculations were performed by BISTRO (Bidimensional Sn Transport Optimisé) code, which solves the transport equations adopting a 2D cylindrical RZ core description by using a discrete ordinates-coarse mesh and finite differences method. The non-isotropic scattering is represented by Legendre polynomials and the discrete treatment of angles is sampled by using the discrete ordinates Sn method ( $n = 4, 8, 16$ ).

The model of XADT for both calculations is shown in Fig. 2.

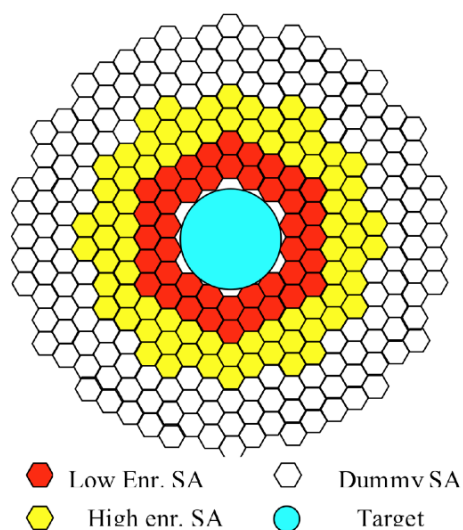


Figure 1. XADT core configuration

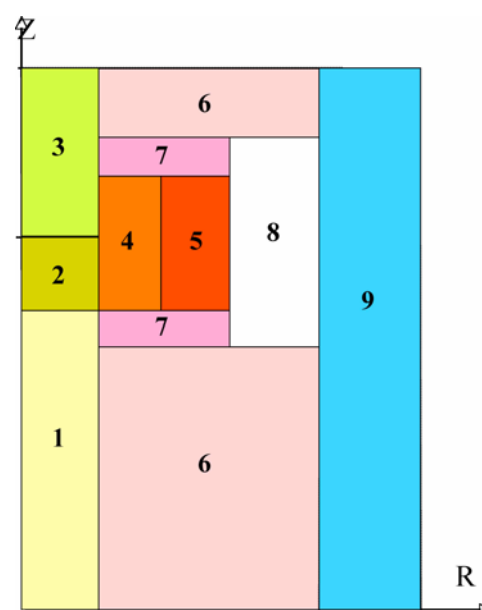


Figure 2. XADT model [1- LBE; 2- Target (source); 3- Beampipe; 4/5- Fuel low/high enrichment; 6- Axial shielding; 7- Plenum; 8- Dummy SAs; 9- LBE]

## 2.3 Neutronic analysis

### 2.3.1 Multi-recycling scenario analysis

Each cycle was subdivided in six evolution time-steps of 155 EFPD (Equivalent Full Power Days) in order to take into account the change of the core composition during the cycles and a subcritical level at BOC (Beginning Of Cycle) of about -3000 pcm in presence of the external neutron source was imposed adjusting the Pu and MA content of the two core zones so that the enrichment concentrations have the same values of the first cycle. The solid fission products accumulating during the cycles were taken into account with six special pseudo nuclides. The cross-sections of core materials were updated for each time step and the main parameters (average and maximum burnup rate, average flux, density power, power rating, etc.) were calculated for the two fuel zones.

The results for XADT-80 loaded with CERCER and CERMET fuel SAs are summarized

in Tables II and III. Table III reports the effective delayed neutron fraction as well as the reactivity parameters related to the Doppler and coolant void effects. The Doppler effect was assessed in the range of 1000-1470 K. Due to the lack of U238 in the fuel composition, it is slightly positive. The coolant void effect that represents the variation of reactivity per unit volume of coolant loss is strongly space-dependent and it was calculated for the whole fuel zone, and the high and low enrichment zones. The values confirm the conclusions of more detailed evaluations, namely that the coolant void in the central zone rows has a greater impact than in the outer rows. The positive coolant void effect may be mitigated by LBE high coolant boiling point and it can be reduced if a CERMET fuel is adopted. It is worth underlining that for both fuels: 1) the reactivity decreases from BOC to EOC and it is practically constant at EOC (End of Cycle) in each cycle, Figs. 3 and 4; 2) the capability to transmute MA decreases with the number of cycles, Fig. 5, whereas the capability to burn plutonium increases, Fig. 6. Moreover Pu is burnt from the first cycle for CERMET fuel, whereas it is produced when the core is loaded with CERCER fuels; 3) the neutron spectra for CERMET fuel is harder than that related to CERCER, Fig. 7. That explains the difference between the two fuels concerning the transmutation rates, core enrichments and reactivity parameters; 4) the reactivity feedbacks do not play an essential role during the normal operation. In any case a suitable subcritical level is imperative due to the small values of Doppler feedback and the delayed neutron fraction.

**Table II. XADT-80 – Main neutronic parameters**

Parameters	MgO		Mo92	
	1 <sup>st</sup> cycle	10 <sup>th</sup> cycle	1 <sup>st</sup> cycle	10 <sup>th</sup> cycle
Inner core enrichment [%]	30.85	30.85	30.87	30.87
Outer core enrichment [%]	37.17	37.17	37.19	37.19
Reactivity loss per day [pcm/d]	1.46	1.46	1.95	2.13
Max density power [W/cm <sup>3</sup> ]	BOC	66.5	66.0	64.0
	EOC	71.1	69.2	71.8
Max power rating [W/cm]	BOC	122.0	121.0	117.4
	EOC	130.4	126.9	131.6
Average enrichment	34.96	34.96	34.98	34.98
Inner Core Flux *E14 [n.cm <sup>-2</sup> .s <sup>-1</sup> ]	BOC	7.55	7.67	8.06
	EOC	8.09	8.49	8.70
Outer Core Flux *E14 [n.cm <sup>-2</sup> .s <sup>-1</sup> ]	BOC	5.56	5.65	6.05
	EOC	5.70	6.11	6.17
Average Burnup [MWd/T]	37125	37086	37119	37085
Max Burnup [MWd/T]	55902	54071	55849	54485

**Table III. XADT-80 Safety parameters at BOC**

Parameter	MgO	Mo92
$\beta$ [pcm]	151.4	150.9
Fuel Doppler (pcm/K)	0.011	0.006
Coolant Void worth [pcm/dm <sup>3</sup> ]		
Fissile	2.0	1.7
LE Fissile	4.0	3.7
HE Fissile	0.7	0.39

Increasing the density power, the trend of reactivity along the cycle is the same of XADT-80 for both fuels, but with a lower swing due to production of plutonium for the CERCER fuel, Fig. 3, whereas for the CERMET the reactivity loss is greater, Fig. 4. Furthermore the increase of density power has an opposite effect on MA transmutation rate

and on plutonium burnup rate, because the former decreases, as shown in Fig. 5, the latter increases, Fig. 6. Except for XADT-80 plutonium is always burnt from the first cycle.

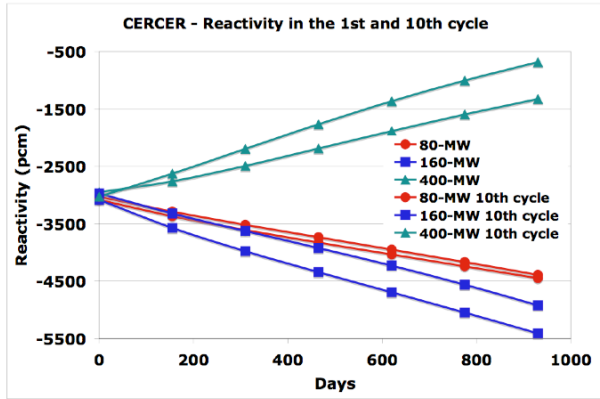


Figure 3. CERCER: Reactivity vs. time

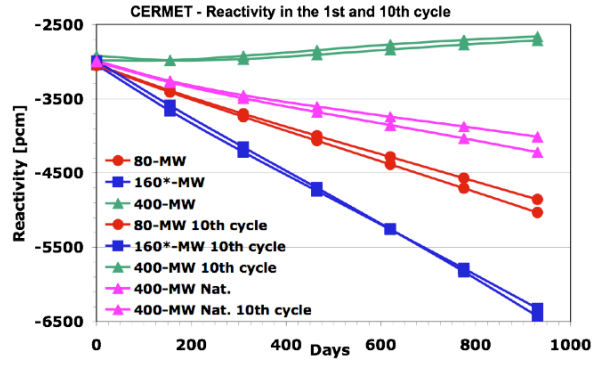


Figure 4. CERMET: Reactivity vs. time

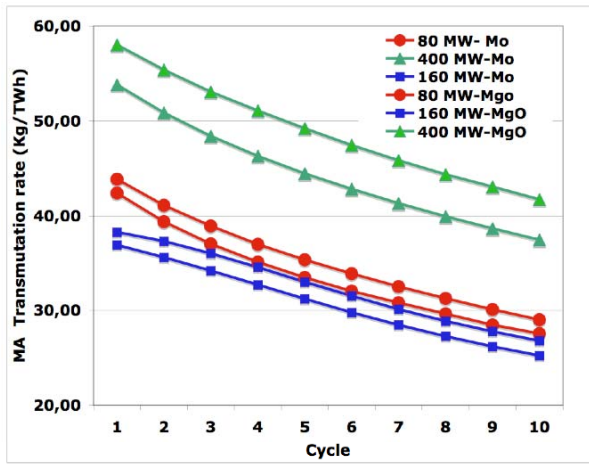


Figure 5. MA transmutation rate vs number of cycles

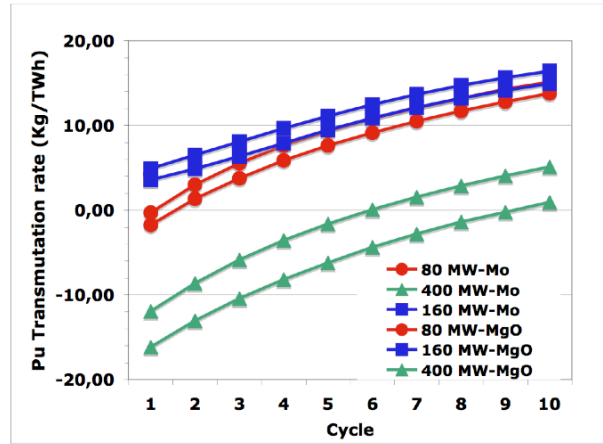


Figure 6. Pu transmutation rate vs number of cycles

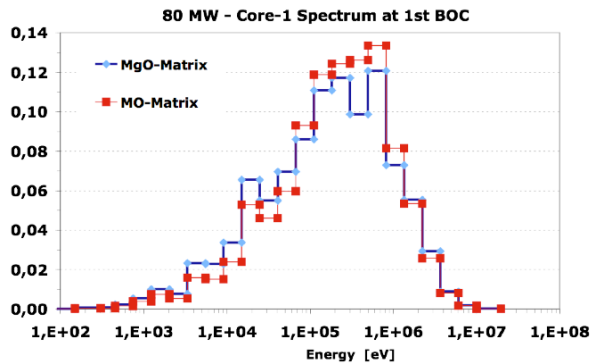


Figure 7. Core low enrichment neutron spectrum

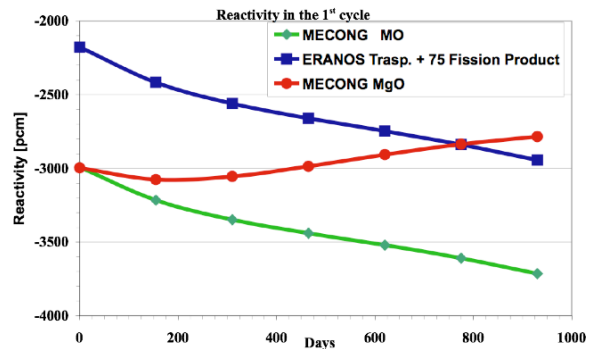


Figure 8 XADT-300 Reactivity vs. time

By increasing the plant size (power equal to 400 MW<sub>th</sub>), the trend of the reactivity and Pu and MA transmutation rates along the cycles is reported in Figs 3, 4, 5, 6 for both fuels. It can be noticed that: 1) the plutonium content needed at the beginning of each cycle decreases, whereas it is necessary to provide more minor actinides and this situation is favourable for the transmutation; 2) unlike XADT-80 core, where the reactivity always decreases between BOC and EOC, the reactivity increases during each cycle, except for a CERMET fuel in

which the component of the matrix is Mo natural, characterized by a small content of Mo92 (14.8%); 3) the MA transmutation capability decreases during the multi-recycling, reducing to about 70 % in comparison with the first cycle, whereas the capability to burn plutonium increases by about 17 Kg/TWh. Except for XADT-80 plutonium is accumulated in the first 6 cycles and then it is burnt if a CERMET fuel is used, whereas for a CERCER fuel Pu is burnt beginning from 9 cycle.

### 2.3.2 Detailed neutronic assessment

This analysis was only performed for a core charged with CERMET fuel and characterized by a thermal power of 300 MWth, a power rating of 250W/cm and a core active height of 0.99 m in order to avoid a core too flattened.

The main differences compared with MECONG calculations concern: 1) the microscopic ( $\sigma$ ) and macroscopic ( $\Sigma$ ) cross-sections that were obtained with a heterogeneous description of fuel assemblies and treated with a fine energy structure (1968 groups) and then collapsed in 33 energy-groups; 2) the fission products (FPs) accumulated along the cycle were considered by means of 75 solid FPs for the heavy nuclides instead of six special nuclides. The gaseous FPs were neglected, because their effect on reactivity is negligible during the cycle; 3) use of transport equations instead of diffusion ones, consequently the neutronic calculations (flux and neutronic balance) were performed by BISTRO code.

It is worth noting that the reactivity swing during the first cycle is -767 pcm against -723 pcm calculated by MECONG. Both values are greater than CERCER reactivity swing (+212 pcm) calculated with MECONG, Fig. 8. Furthermore, due to the combined effect of the three above-mentioned differences, the reactivity calculated by ERANOS is greater than MECONG values, Fig. 8.

The MECONG calculations about the MA and Pu transmutation capability show that the MA transmutation rates are almost equivalent for both fuels (46.58 Kg/TWh for CERCER fuel and 45.40 Kg/TWh for CERMET fuel) and Pu is burnt beginning from 5<sup>th</sup> cycle.

### 2.3.3 Accelerator beam current analysis

The beam currents needed to compensate the reactivity swing along the cycle are reported in Table IV. It is worth noting that the current values are always lower than the design limit.

Table IV. Proton beam current vs reactivity swing

Power [MW]	MgO				Mo92			
	1 <sup>st</sup> cycle		10 <sup>th</sup> cycle		1 <sup>st</sup> cycle		10 <sup>th</sup> cycle	
	BOC	EOC	BOC	EOC	BOC	EOC	BOC	EOC
80	2.45	3.53	4.37	3.44	2.37	3.80	2.39	3.93
160	4.89	8.59	4.69	7.74	4.82	10.0	4.7	10.1
400	11.9	5.36	12.1	2.72	11.7	10.9	11.8	10.5
300	9.02	8.41	9.01	5.53	9.00	11.2	-	-
300*	-	-	-	-	6.55	8.87	-	-

\* values referring to with ERANOS calculations

## 2.4 Fuel analysis

A sensitivity analysis was performed to investigate the CERCER and CERMET fuels performance under assumed conditions (300 MW<sub>th</sub>). The maximum EOC fuel temperature and Damage Function (DF), defined as sum of plastic and thermal creep strain of cladding both normalized on a 0.2 % limit, were investigated for the hot fuel pin in function of the pellet swelling and the gas release.

To this purpose, TRANSURANUS code [7] was modified by introducing basic thermo-physical and mechanical properties (density, specific heat, thermal conductivity, linear thermal expansion, Poisson ratio, Young modulus, yield stress) from literature data and predictive models developed in EU projects [1, 2] for both fuels.

The following hypotheses about the fuel behaviour under irradiation were made: 1) thermal conductivity: the degradation of the composites thermal conductivity with burn-up is only due to a UO<sub>2</sub>-like MA oxide thermal conductivity degradation, as the degradation of matrix thermal conductivity due to alpha and fission products (that may be important for the fuels composition considered) was neglected; 2) swelling: the effect of neutron flux on the voids formation in the matrix was considered only for CERMET, while an annealing effect was assumed for CERCER matrix. A 0.5 %/at% burnup solid fuel swelling was assumed. Moreover a High Burnup Structure formation was supposed for the fuel phase with a modelling similar to the LWR case. The solid component of swelling was changed by using an input factor; 3) gas release: a diffusion modelling was assumed for evaluating the gas release for Xe-Kr and He in the fuel phase, while the effect of matrix was represented by an input factor to increase the grain boundary saturation limit; 4) densification and relocation: standard TRANSURANUS modelling was assumed.

The results are summarized in Figs 9 and 10. Fig. 9 shows the EOC hot fuel temperature and the cladding Damage Function vs. He release for a given swelling (4.71% for CERCER and 5.13% for CERMET). The hot fuel temperature and DF are well below the design limits. Fig. 10 reports the same parameters in function of the fuel swelling for a given He release. In this case CERMET proves a better swelling-tolerant behaviour than CERCER fuel. Even if the CERMET fuel has a negligible open porosity so that the fission and decay products are retained within the matrix, high values of helium release were investigated to take into account possible melting or cracking phenomena.

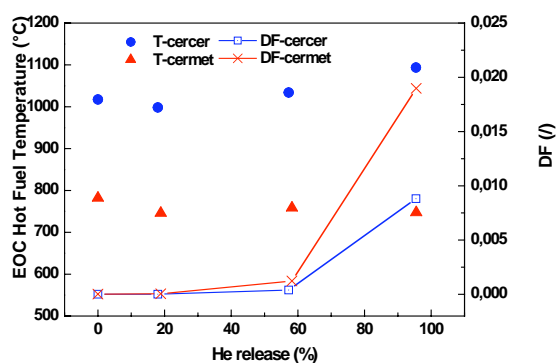


Fig. 9 EOC fuel temperature and DF vs. He release

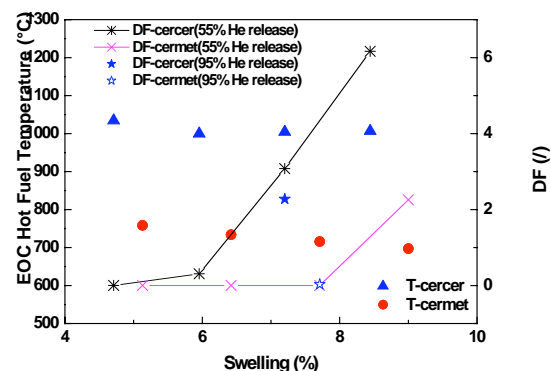


Fig. 10 EOC fuel temperature and DF vs. swelling

### 3 CONCLUSIONS

The performed analysis shows a good compromise for both fuels between transmutation and core performance by increasing of the fuel core dimensions (power size). In particular, the reactivity and transmutation rate show an opposite trend along the cycles for CERCER fuel, as the former increases and the latter decreases, whereas for CERMET fuel that happens beginning from about 400 MWth. To cope with this rise it is necessary to choose a suitable refuelling strategy, otherwise it needs to design the core so that  $K_{\text{eff}}$  is constant or almost constant during the cycle with the advantage to operate the accelerator at a constant beam current.

The use of CERMET fuels with a matrix of Mo92 may present some advantages, among them to reduce the reactivity swing.

### 4 ACKNOWLEDGMENTS

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