DESIGN AND MODELLING OF OXIDE FUEL FOR INCINERATION OF AMERICIUM AND PLUTONIUM IN AN EXPERIMENTAL ADS

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Abstract

The interest to the accelerator-driven subcritical systems (ADS) devoted to the incineration of plutonium and minor actinides is a subject of study around the world. Since 1997, the Belgian Nuclear Research Centre SCK·CEN is performing the pre-design studies aiming at the development of a small experimental ADS called MYRRHA which could be a step towards a prototype design of an ADS-transmuter. In the present article, some results of the preliminary modelling of the behaviour of the fuel pins, filled with $(Am_{0.5}Pu_{0.5})O_{2-x}$ dissolved in ThO_2 matrix and with the same fuel in YSZ matrix, under typical ADS operation conditions are discussed and compared with the behaviour of MOX. Modelling has been performed with the fuel performance code MACROS-II which is under development and testing at SCK·CEN. A care was taken to model correctly the re-distribution of transuranium elements, fission products and helium within the fuel pellets and to their impact on the fuel performances.

Introduction

The interest to the accelerator-driven subcritical systems (ADS) devoted to the incineration of plutonium (Pu) and minor actinides (MA: Np, Am, Cm) is a subject of study around the world. Since 1997, the Belgian Nuclear Research Centre SCK·CEN has being worked on the pre-design of a small experimental ADS called MYRRHA (**M**ultipurpose h**Y**brid **R**esearch **R**eactor for **H**igh-tech **A**pplication) which could be a step towards a prototype design of an ADS transmuter and will allow to test the prototype fuels for MA transmutation [1, 2].

Various fuel systems (metallic, nitride, oxide, CERCER, CERMET) are considered as promising candidates for MA transmutation in different countries. Mixtures of actinide oxides and inert matrices are considered to be more close to realisation.

In the present article, some results of evaluation of the performances of two experimental rods with $(Am,Pu)O_2$ fuel dissolved in ThO_2 and YSZ matrices during their long term operation in the MYRRHA ADS fast core are presented and compared with the behaviour of the fuel rod with MOX. Typical irradiation conditions (neutron spectrum, fission density) were pre-calculated with the MCNPX code [3]. Preliminary evolution of the fuel composition and heating rates (power history) were assessed with the ORIGEN-2.2 code [4]. Finally, the fuel properties and the thermomechanical behaviour of the fuel rods were calculated with the MACROS-II code which is under development and validation at the SCK·CEN [5]. A care was taken to model correctly the re-distribution of isotopes of the transuranium elements (TRU) and fission products within the fuel pellets and to their impact on the fuel properties. Special attention was paid to the effect of helium, produced due to α-decay of some TRU, and of other rare gases (Xe, Kr).

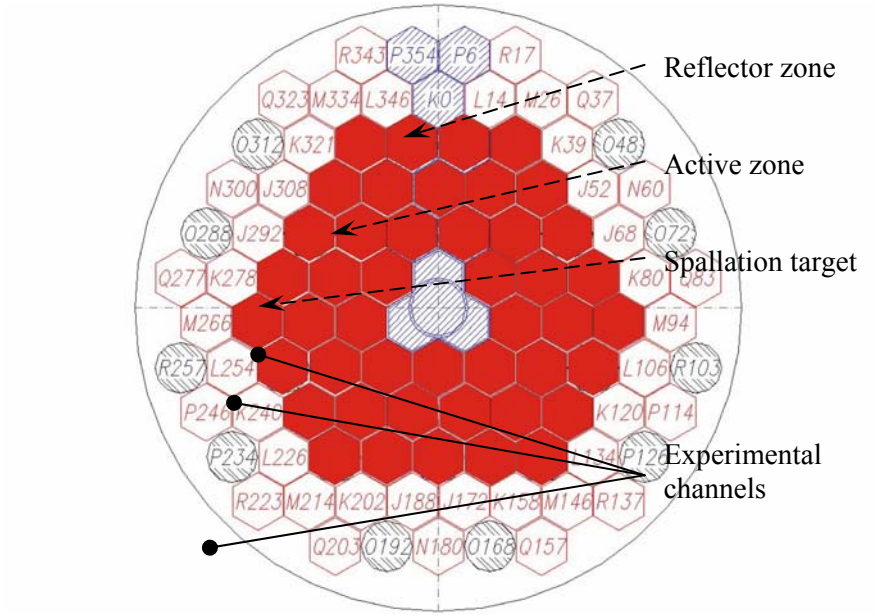
The irradiation conditions in the hottest sub-channel of the hot fuel assembly were used in calculations as reference. The operation period of three irradiation cycles of 90 days with the shutdown periods of 30 days in between was analysed. The obtained results confirm rather good fuel performances during the considered period and indicate a potential to extend the operation for a few cycles more. A high importance of helium, produced in the fuels containing americium, was illustrated.

Brief description of the MYRRHA core

The applications considered in the MYRRHA project focus primarily on the ADS conception demonstration, possibilities of waste transmutation, radioisotope production and safety research on sub-critical systems. The MYRRHA ADS concept, as it is up today, is based on the coupling of a 350 MeV*5mA proton accelerator with a liquid (Pb-Bi eutectic) spallation target surrounded by a subcritical neutron-multiplying core-blanket with a power of about 50MWth. The fast core and the experimental channels are also cooled by the liquid Pb-Bi eutectic. Different types of fuel and designs of a fuel element are under study. In the current design of the MYRRHA core, $(U,Pu)O_2$ MOX fuel with 30 wt.% of the reactor grade plutonium (RG Pu) in heavy metal is considered as a basis of the subcritical core, whereas different sorts of experimental fuels and targets with MA and long-life fission products will be tested in the experimental channels [6]. In a later phase, a part of the driver MOX in the subcritical core could be replaced with non-uranium and MA containing fuels considered as pre-prototypical for an European ADS-transmuter.

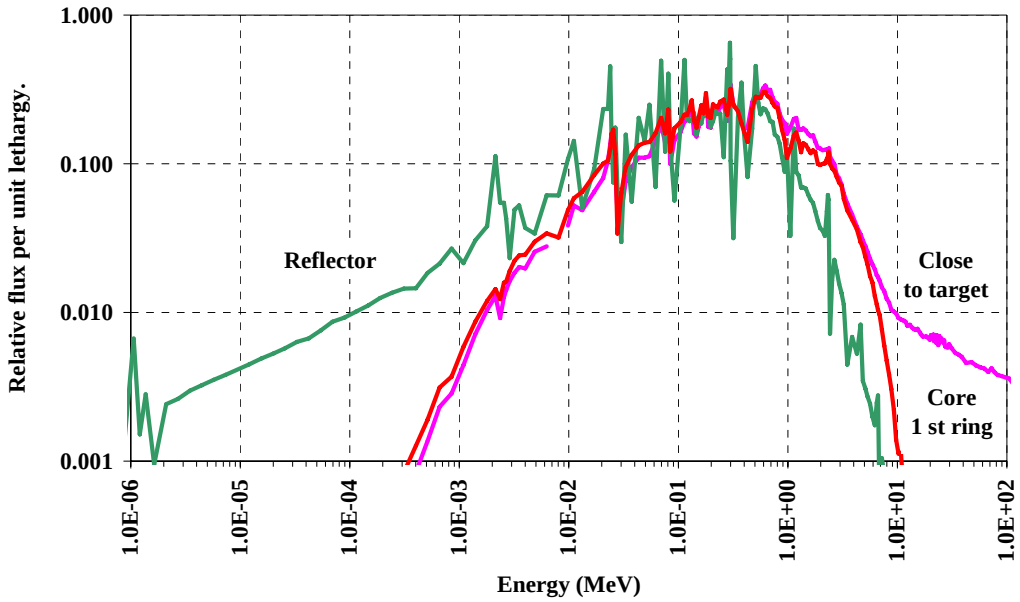
A typical configuration of the MYRRHA subcritical fast core is presented in Figure 1. Three types of channels for irradiation of the MA targets and the experimental fuel rods are foreseen: close to the spallation target, in the active zone and in the reflector.

Figure 1. **A typical configuration of the MYRRHA subcritical core.**



Typical neutron spectra in the reflector, in the fast core and in the channel close to the spallation target are presented in Fig. 2, where the normalized to unity relative neutron flux per unit lethargy is presented in function of neutron energy. The total neutron flux in the experimental channels was calculated to be in the region of $(0.5\text{-}5)\cdot 10^{15}$ n cm^{-2} s^{-1}, depending on the channel position, fuel burnup and core configuration. Other neutronic parameters are given in the report [6].

Figure 2. **Normalised neutron spectra in different experimental channels of MYRRHA ADS**

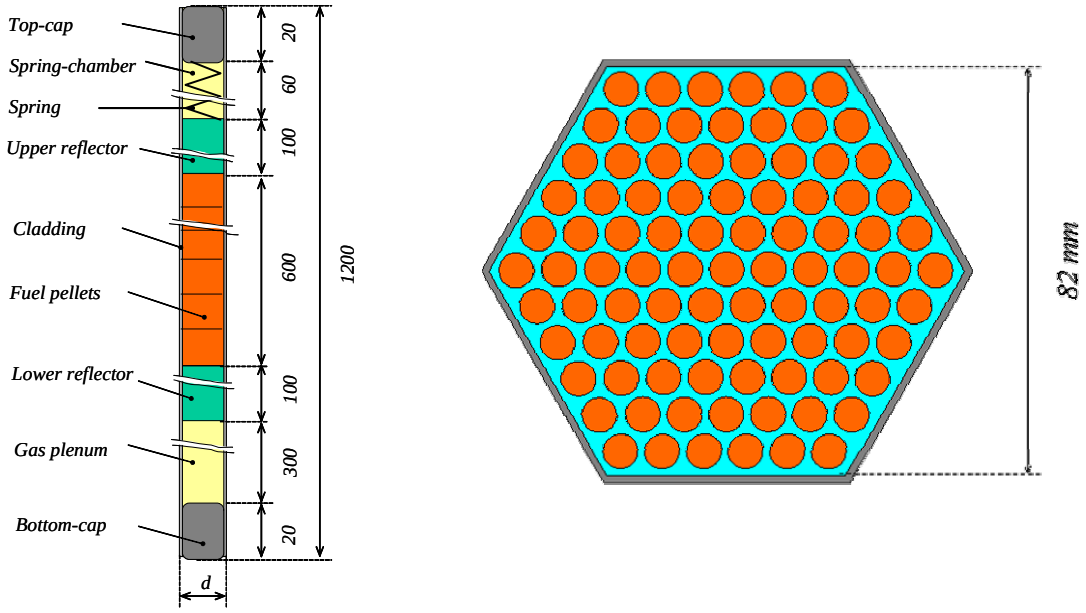


During the normal operation, MYRRHA ADS will work by cycles of 90 days. The basic regime is to keep the proton beam current at constant level, however some experiments could be performed with a constant neutron flux, when a loss in the core reactivity within a cycle is compensated by the proton beam current. Shut-down periods of 30 days between the irradiation cycles are foreseen for the ADS maintenance and for the core reconfiguration, in order to compensate a reactivity loss accumulated during one-two cycles.

Fuel pin and assembly design

Two important requirements for the fuel pin design are non-melting of the fuel pellets and non-damage of the cladding by inner or outer stresses during the total fuel life. Given the maximum desired power density in the core, the first criterion will determine the pellet radial dimensions. The second criterion will determine the clad diameter and thickness and the gas plenum volume. In order to be able to demonstrate effectively the possibility of MA transmutation, a high level of the neutron flux aimed in the design. It resulted in a high peak power density in the MYRRHA fuel: 1.5-1.7 kW cm^{-3} and, consequently, required the fuel pellets of a small diameter: 5.3-5.9 mm in different pre-design options. The MOX fuel pellets were assumed to be of 95 % of theoretical density (TD) and containing 30 wt.% RG Pu in initial heavy metal (iHM). The initial isotopic composition of HM is typical for RG MOX [7] and is given in Table 1. The natural isotopic composition was used for oxygen [8].

Figure 3. **The axial schematics of a fuel rod (a) and the radial schematics of a fuel assembly (b)**



(a) (b)

Martensitic steel T91 was preliminary chosen as cladding material, taking into account its good mechanical parameters, low irradiation induced swelling of the martensitic 7-10 % Cr steels [9] and corrosion resistance in the liquid Pb-Bi eutectic environment at temperatures lower 470 °C [10]. The needed assessments were performed to optimise all parameters of the MYRRHA driver fuel pin (fuel type, pellet density and dimensions, cladding diameter and thickness, gas plenum dimensions, etc.). The axial schematics of the reference pin are presented in Figre 3a. At this stage, the same design was also used for the experimental fuel rods with MA fuel.

The triangular type sub-channel and the hexagonal type shroud were chosen in the fuel assembly design, similar to those widely used in many LMFBR (Fig. 3b). Each fuel assembly contains 91 fuel rods and cooled by Pb-Bi flow entering from below with the inlet temperature of 200 °C. The Pb-Bi mass flow rate is limited by the maximum allowed local velocity that should not exceed 2.0 m/s at normal operation conditions because of possible erosion problems.

Results of modelling

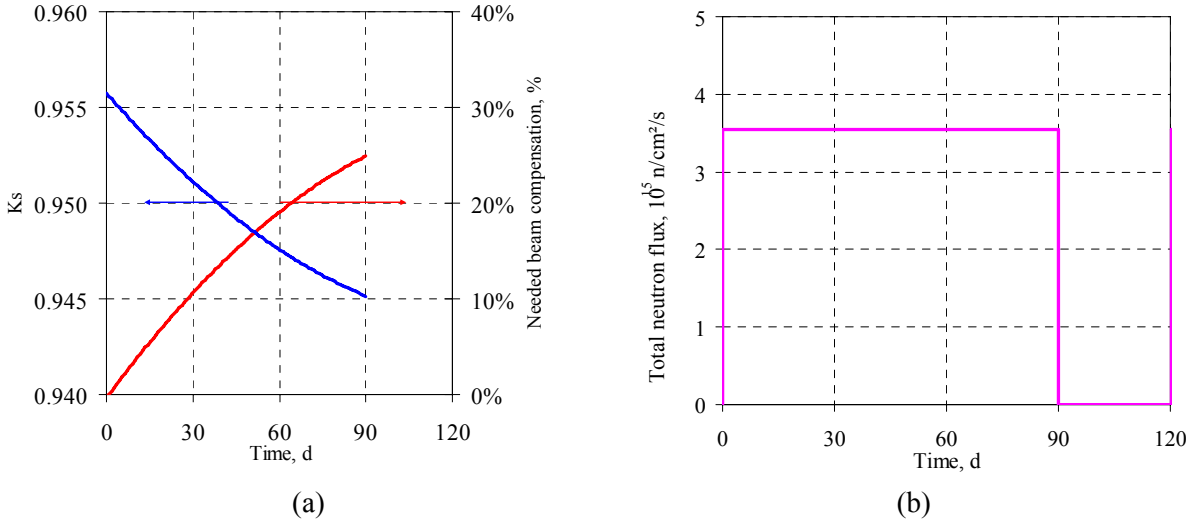
Thermomechanical behaviour of three fuel rods has been analysed: one with the driver MOX, another with $(Am_{0.5}Pu_{0.5})O_{2-x}$ dissolved in the ThO_2 matrix and the third with the same fuel dissolved in the YSZ matrix (called below "inert matrix fuel" - IMF). In two last cases the volume fractions of MA fuel and matrix were equal. The initial isotopic compositions of americium and plutonium used in the calculations were taken from [7] for the spent fuel of a typical PWR (with mean burnup of 33MW d kg^{-1} HM) after ten years of storage; it is given in Table 2. For Th, Zr and O, the natural isotopic compositions were used [8]. All fuels were assumed to be of 95 % TD.

Table 1. **Initial isotopic vectors of uranium, plutonium and americium used in the calculations**

U-isotope	Content wt.%	Pu-isotope	Content wt.%	Am-isotope	Content wt.%
^{234}U	0.003	^{238}Pu	1.27	^{241}Am	84.503
^{235}U	0.404	^{239}Pu	61.88	^{242m}Am	0.247
^{236}U	0.010	^{240}Pu	23.50	^{243}Am	15.25
^{238}U	99.583	^{241}Pu	8.95		
		^{242}Pu	4.40		

The initial irradiation conditions for the studied fuel rods (neutron spectrum, power density and flux level) were calculated with the MCNPX code. The peak value of the total neutron flux $3.5 \cdot 10^{15}$ $cm^{-2}s^{-1}$ was obtained in the hottest channel of the fast subcritical core.

Figure 4. **Reactivity degradation and needed compensation in proton current (a); neutron flux (b)**



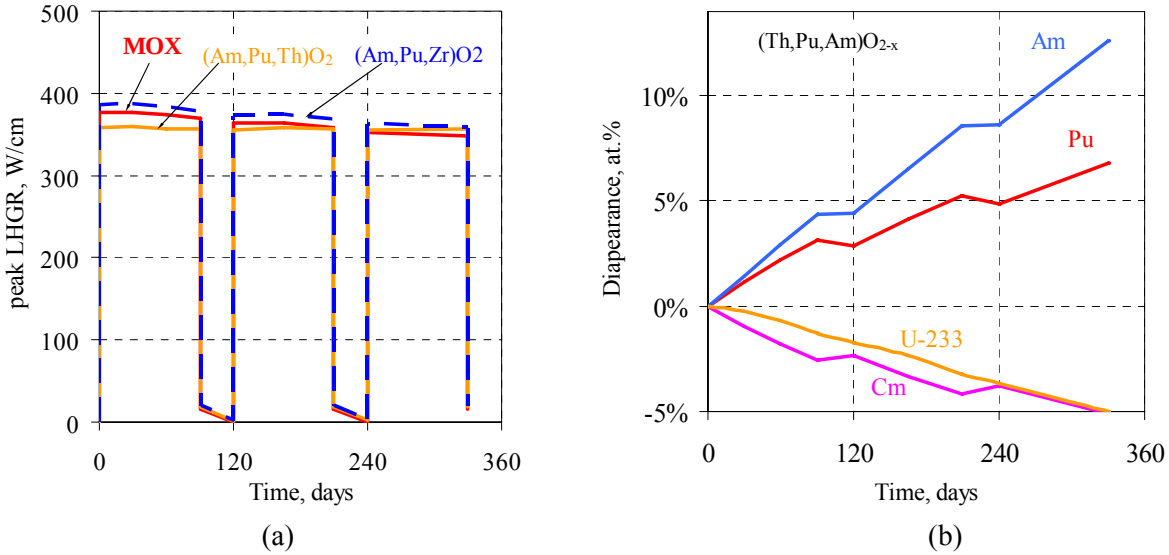
For the following calculations it was assumed that the total neutron flux is kept constant during 3 irradiation cycles (each of 90 days). For this goal, on-line compensation of the reactivity losses is foreseen by the increasing the proton beam current within operation cycle and by the core reconfiguration between the cycles when neutron flux is zero (Fig. 4). This scenario is more conservative (i.e. with a higher fuel power) than that with a constant level of the proton beam currently postulated to be basic in the reference design. The power history for the studied fuel rods was pre-calculated with the ORIGEN 2.2 code.

Normal operation

The modelling of the thermomechanical behaviour of the fuel rods during normal operation was performed with the help of the fuel performance code MACROS-II, which is an advanced version of the code MACROS [5]. The data base of the material properties and burnup modules were modified in order to include new materials and actinide isotopes.

Figure 5a illustrates evolution of the peak linear heat-generation rate (LHGR) with time (history of irradiation) pre-calculated with ORIGEN-2.2 code. The cross sections used in the code libraries were weighted by the neutron spectrum in the hottest channel of the MYRRHA core. During three cycles of operation all three rods have very close LHGR between 350 and 380 W/cm. A tendency to a slow decrease of LHGR with time is observed for MOX and IMF rods (mainly due to plutonium burnout). After three operation cycles this reduction reaches about 7 % of the initial value. About constant value of LHGR is observed for the rod with $(Am,Pu,Th)O_2$ fuel where Pu burning is partially compensated by U-233 (Fig. 5b) produced due to conversion of Th-232. At a long term, this effect can be considered as a positive one which can help to keep the reactivity level in the sub-critical core of an ADS-burner without large compensating reserve in the accelerator current.

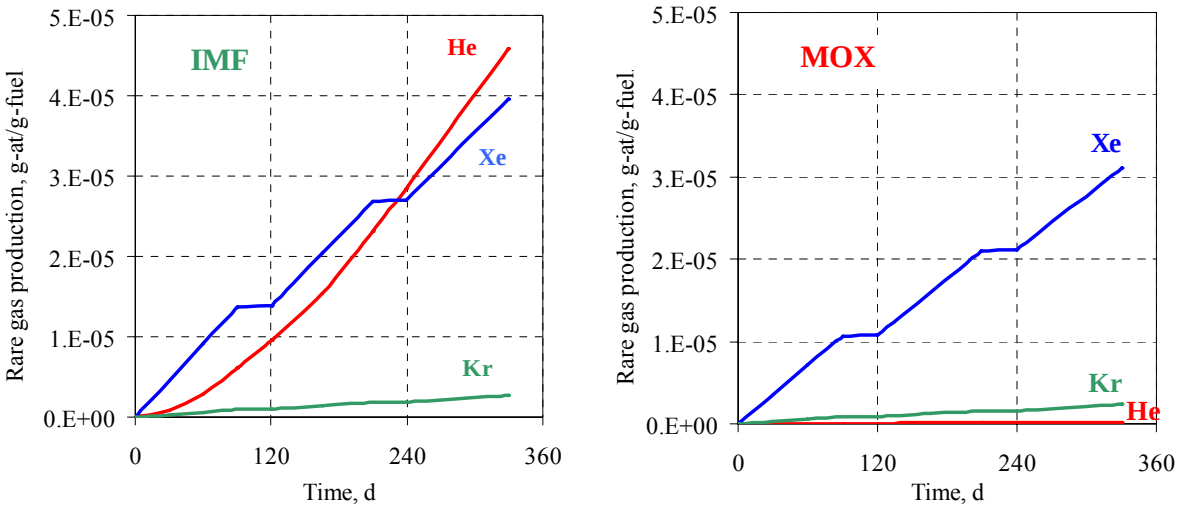
Figure 5. **Linear power (a) and central temperature (b) evolution**



Under considered conditions MA incineration is very similar for $(Am,Pu,Th)O_2$ and for IMF: 12.6 at. % of Am disappears after three cycles of irradiation, but at the same time curium is produced in amount equivalent to 5.1 at. % of the initial americium. It is interesting to note that the curium content decreases and the plutonium content increases during the reactor stop-periods. This is mainly explained by alpha-decay of ^{242}Cm and ^{244}Cm, which generate ^{238}Pu and ^{240}Pu respectively.

The amount of generated helium in the samples containing americium increases very rapidly with time (Figure 6a). After three cycles of operation, its concentration is about the same as that of Xe and Kr together. Due to alpha-decay of americium and curium, the amount of helium within these fuels continues to increase during the reactor maintenance too. In the MOX fuel, He contribution into the produced rare gases is negligible (Figure 6b).

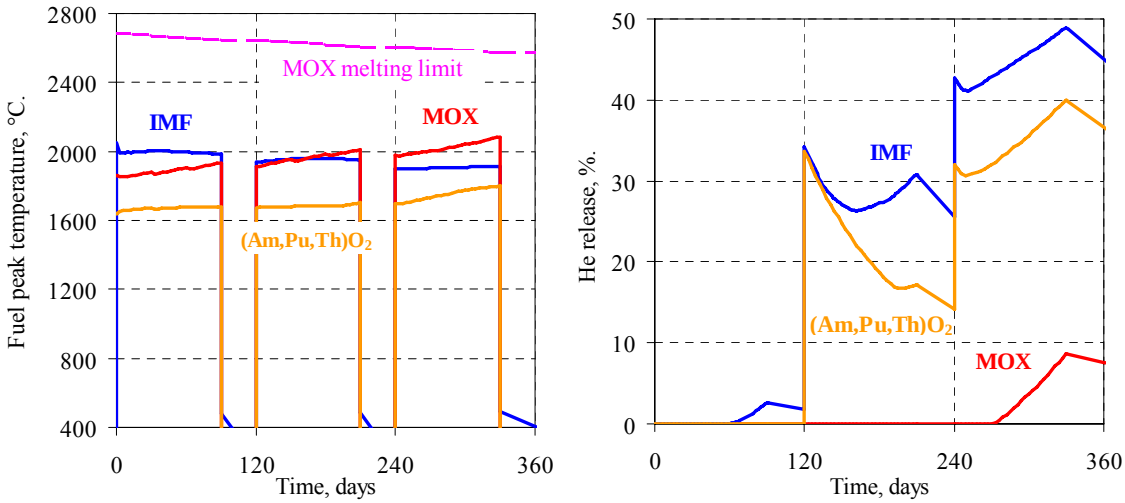
Figure 6. **Production of rare gases: Xe, Kr and He, - in fuel: (a) – IMF, (b) – MOX.**



(a) (b)

The central temperatures of the peak fuel pellets in function of operation time are presented in Fig. 7a. The temperature levels are rather high: at the beginning of operation, the maximum temperature of MOX is about 1860 °C °C, for $(Am,Pu,Th)O_2$ – 1650 °C, and ~ 2000 °C in the IMF rod. A higher temperature in the IMF fuel at start is mainly explained by a lower thermal conductivity. For MA fuel with thoria matrix two factors play the role: a higher thermal conductivity of the ThO_2 matrix and a lower LHGR.

Figure 7. **Evolution of the fuel maximum temperature (a) and gas release from the fuel (b)**



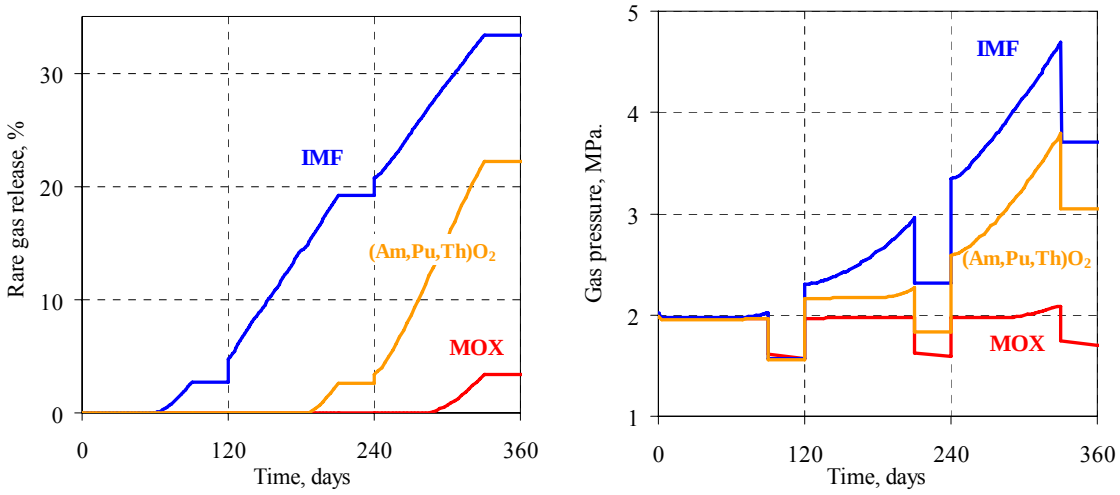
(a) (b)

In the driver MOX and in (Am,Pu,Th)O_2, the peak temperature increases with irradiation, demonstrating a domination of the irradiation induced degradation of thermal conductivity. By the end of the operation period, the peak temperatures reach 2085 °C in MOX and and 1760 °C in (Am,Pu,Th)O_2. However, these values are still significantly below the melting limit of 30% MOX [11]. In the IMF fuel this effect is not observed because of a high defect structure of the initial non-irradiated material. The peak temperature in this rod slightly decreases with time, mainly due to a decrease of LHGR. It also remains rather below its estimated melting temperature.

The right-side picture in Figure 7 shows the release of helium from the fuel pellets in the three rods. This release starts in mid first cycle in IMF rod, where temperature is the highest at this moment and the amount of the accumulated helium is sufficiently high (see Figure 6a). A rapid temperature increase at the start-up of the second cycle causes a "burst" release of helium that was accumulated during the cold shutdown period due to alpha-decay of curium and americium. After the restarting, a fraction of the released helium decreases with time because the He generation rate is higher at the beginning of the cycle, than its release rate. Then, the release rate increases with the raise of the He concentration in the fuel. These effects are also observed in the third cycle. Helium release from MA fuel with thoria matrix follows similar behaviour but starts a bit later because of a lower temperature. In the MOX, helium concentration is very low (see Figure 6b), and it might be supposed that it remains captured by micro pores and other structure defects until mid third cycle (Figure 7b).

The obtained results on the rare gas release and pressure evolution in the studied rods are presented in Figure 8. They demonstrate clearly the importance of helium in the fuel with americium and curium.

Figure 8. **Release of rare gases (c) and pressure evolution in the rods (d).**



At the end of the third cycle, more than 33 % of the produced rare gases are released from IMF, about 22 % from (Am,Pu,Th)O_2 and only about 3.3 % from MOX. Pressure in the rods follows the gas release evolution and, just before the reactor shut down in the third cycle, attains 4.6 MPa in IMF rod, 3.8 MPa in the rod with the thoria matrix fuel and only 2.1 MPa in the driver MOX rod. These values are significantly below the design limit of 12 MPa.

Conclusions

The preliminary modelling of the thermomechanical behaviour of the driver fuel rod with 30 % RG Pu MOX and two experimental fuel rods with americium-plutonium dioxide in the thoria and in the YSZ matrices has been performed with the MACROS-II code under typical irradiation conditions of the research ADS MYRRHA. The obtained results show that all three fuel rods can survive without fuel melting or cladding damage during at least three cycles of operation (3 x 90 days) in the hottest channel of the subcritical core even if a conservative scenario of operation foreseen at the constant maximum neutron flux. These results suggest that the same operation regime can be extended for a few cycles more. The modelling is under way to determine a limit of the fuel life.

The helium, generated in fuels containing americium and curium, can contribute significantly in their irradiation behaviour and in the internal pressure build-up in the fuel rods. The respective effects, especially possible "burst" release after a cold period, must be taken into account in MA fuel design and in modelling of future irradiation experiments with this kind of fuels and targets.

Acknowledgments

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REFERENCES

[1] H. Aït Abderrahim, et al., *Nuclear Instruments & Methods in Physics Research*, **A 463** (2001) 487-494.

[2] V. Sobolev, et al, "Fuel Activities Related to ADS development at the SCK·CEN Application to MYRRHA". *Proc. of the European Working Group "Hot Laboratories and Remote Handling". Mol, Belgium, September 25-27, 2002.*

[3] MCNPX User's Manual, Version 2.3.0. Ed. S. Waters, *Los Alamos National Laboratory Report* LA-UR-02-2607, April 2002.

[4] "ORIGEN 2.1: Isotope Generation and Depletion Code, Matrix Exponential Method", *Oak Ridge National Laboratory, Oak Ridge, Tennessee. CC-371.* August 1996.

[5] S. Lemehov, P. Van Uffelen, V. Sobolev, "MODELLING THE BURNUP EFFECTS IN ACTINIDE FUELS WITH THE MACROS CODE". *Proc. of the Enlarged Halden Programme Group Meeting", Storefjell, Norwary, September 8-13, 2002. Report HRP-359.*

[6] E. Malambu et al., "MA and LLFP Transmutation Performance Assessment in the MYRRHA Small-scale ADS". *This Conference, Session V: Transmutation: ADS.*

[7] W. S. Yang, "Blanket design studies for maximizing the discharge burnup of liquid metal cooled ATW systems". *Annals of Nuclear Energy, 29 (2002) 509*.

[8] W. Seelmann-Eggebert, "KARLSRUHE NUKLIDEKARTE". KfK, Karlsruhe, 5. Auflage 1981.

[9] Z. Zhu, P. Yung, H. Klein, *Journal of Nuclear Materials, 202 (1993) 222-227*.

[10] F. Barbier, G. Benamati, C. Fazio, A. Rusanov, *J. of Nuclear Materials, 295 (2001) 149-156*.

[11] SCDAP/RELAP/MOD3.1 Code Manual; Vol. IV: MATPRO – *A library of Materials Properties for Light-Water-Reactor Accident Analysis. Ed. D.T. Hagrman. INEL Report NUREG/CR-6150*, Idaho, USA, 1993.