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Irradiation and post-irradiation examination of uranium-free nitride fuel

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ABSTRACT

Two identical Phénix-type 15-15Ti steel pinlets each containing a 70 mm $Pu_{0.3}Zr_{0.7}N$ fuel stack in a 1-bar helium atmosphere have been irradiated in the HFR Petten at medium high linear power (46–47 kW/m at BOL) and an average cladding temperature of 505 °C. The pins were irradiated to a plutonium burn-up of 9.7% (88 MWd/kg_{HM}) in 170 full power days. Both pins remained fully intact. Post-irradiation examination performed at NRG and PSI showed that the overall swelling rate of the fuel was 0.92 vol-%/% FIHMA. Fission gas release was 5–6%, while helium release was larger than 50%. No fuel restructuring was observed, and only mild cracking. EPMA measurements show a burn-up increase toward the pellet edge of up to 4 times. All investigated fission products except to some extent the noble metals were found to be evenly distributed over the matrix, indicating good solubility. Local formation of a secondary phase with high Pu content and hardly any Zr was observed. A general conclusion of this investigation is that ZrN is a suitable inert matrix for burning plutonium at high destruction rates.

1. Introduction

Transmutation of degraded plutonium and americium in Fast Reactors or ADS systems may enable a reduction of radiotoxic inventories directed to geological repository by a factor of 100 [1], [2]. This could be achieved with conventional UO_2-based fuels, for which vast irradiation and back-end experience has been accumulated. However, the net transuranics (TRU) destruction rate may be significantly improved by using uranium-free fuels.

Candidate inert matrices for TRU destruction should ideally combine low neutron cross-section with high thermal conductivity and excellent irradiation behavior [3], but an additional key requirement is good aqueous solubility for the purpose of reprocessing and recycling. The low solubility rate of plutonium oxide in nitric acid [3] appears to require large-scale development of non-aqueous reprocessing methods. In contrast to uranium free oxide fuels, nitrides combine the advantages of maximum destruction rates with a good compatibility with the industrialized PUREX process [4].

The lack of basic and irradiation performance data on uranium free nitrides however necessitates a significant R&D program, before validation and qualification is possible. The participants of the FP5 European project CONFIRM [4] (Collaboration On Nitride Fuel IRradiation and Modeling) therefore agreed to perform theoretical and experimental studies on uranium free nitride fuel characteristics and performance under irradiation, with the financial support of the European Union. To investigate in particular their potential for fast burning at high linear power, within this program two identical segmented pinlets containing 10 ($Pu_{0.3}Zr_{0.7}$)N pellets each have been irradiated in the High Flux Reactor (HFR) at high linear power. The fuel pins were designed and fabricated by the Paul Scherrer Institute (PSI) and have been irradiated for 170 EFPD by the Nuclear Research Group (NRG) in the HFR in Petten.

The main results of the irradiation and the following post-irradiation campaign at NRG and PSI are reported here, followed by a comparison of the CONFIRM results with existing experience.

2. Fuel fabrication

PuZrN fuels in two stoichiometry's ($Pu_{0.2}Zr_{0.8}N$ as well as

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$Pu_{0.3}Zr_{0.7}N$) were prepared from the oxide by carbo-thermic nitriding of the mixed oxide powders. This was done by heating a homogenized mixture of zirconia and plutonium dioxide powders with 2.3 mol of carbon black per mole oxide for 24 h in a nitrogen atmosphere, after which the product was decarburized in a N_2/8% H_2 mixture. A second heat treatment was needed to lower the oxygen content to specifications (<1%). X-ray diffraction patterns from the nitride powder revealed lattice constants near the values expected from linear interpolation of PuN and ZrN (Vegard's law).

The resulting PuZrN powder was mixed with 0.5–1% zinc stearate. From the mixture, pellets were pressed using a hydraulic press (550 MPa) and sintered for 7 h under N_2 atmosphere at 1750 °C. Element analyses were performed on the pellets to determine the carbon, nitrogen and oxygen contents of the final material. After sintering, the pellet diameter was ground to the specified diameter of 5.53 mm on the centerless grinder. Relevant fuel data are summarized in Table 1. Post-irradiation electron microscopy on fuel samples revealed that a small amount of tungsten particles had been taken up in the pellets, most likely during the sintering in the tungsten furnace.

Alpha-autoradiography was performed for a sample pellet in order to characterize the homogeneity of the Pu distribution in the pellets. Axial and radial cut ceramographies were exposed for five seconds to a Kodak CN85 nitrate cellulose foil. The etched foils were scanned with a negative scanner. The resulting scans were processed with a program, which assigned an alpha-emittance to each pixel. The 3D histogram data are averaged over 100 μm^2, where the reference bar in the back represents twice the total mean (Fig. 1).

The 15-15Ti fuel pins provided by CEA (5.65 mm inner, 6.55 mm outer) were filled with helium (purity 99.9999%) at pressures of 1.03 (upper pin, CONF-30-U, upper pin, CONF-20-U, lower pin, CONF-20-L) and 1.04 bar (lower pin, CONF-30-L) before welding. The pins CONF-30-U and CONF-30-L were chosen for irradiation in the HFR in Petten.

3. Irradiation conditions

The two CONFIRM pins have been irradiated in a sodium-filled sample holder in position G7 of the HFR in Petten, for an effective duration of 170.9 full power days in 6 irradiation cycles. Both pins were oriented with the fuel stacks towards the core center line and maximum (vertical) flux, and plenums towards top and bottom of the reactor core. A 0.5 mm hafnium tube was used as a neutron shield to harden the neutron spectrum and reduce fuel power.

Temperatures in the CONFIRM sample holder as well as reactor power, vertical position and control rod level were recorded by means of a computerized data acquisition and storage system

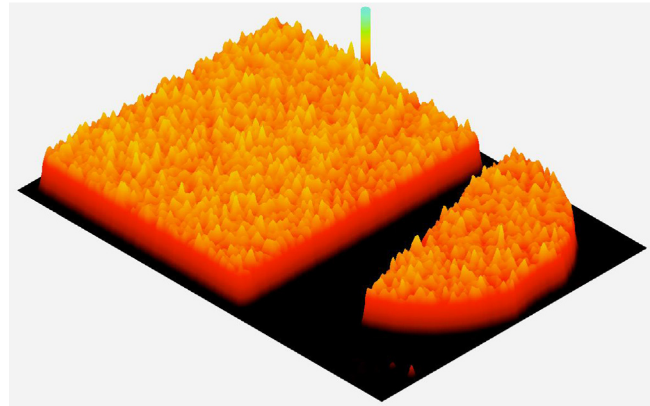

Fig. 1. Alpha-autoradiogram of $Pu_{0.2}Zr_{0.8}N$ pellet indicating a nearly homogeneous distribution of Pu.

(DACOS). The scan interval was 10-s and these 10-s data blocks were reduced to 10 min data blocks and subsequently stored on tape for off-line evaluation.

Thermocouples were located in recesses on the outside of a TZM shroud surrounding the two coupled pins. The observed thermocouple temperatures (Fig. 2) are close indicators of cladding temperatures of the two pins. A finite element model of the as-built experiment was used to derive that outer clad temperatures were 25 °C above the thermocouple temperatures, practically independent of linear power. The (axially) central cladding temperatures of the two fuel stacks were roughly constant over the course of the irradiation, respectively at 505 ± 22 °C and 501 ± 20 °C (1σ, outer clad temperature). Higher temperatures were registered near the end points of the fuel stacks. This was interpreted as resulting from power peaking.

Following the irradiation, fuel burn-up calculations were performed following neutron flux analysis based on activation monitor sets. These calculations are considered accurate to within ±5%. The calculated in-core fuel powers, 442 W/cm on average, are given in Fig. 3. Adopting a thermal conductivity of 12 W/mK, based on measurements carried out by CEA on $Pu_{0.25}Zr_{0.75}N$ [6], pellet central and surface temperatures of 1600 and 1200 K at beginning of irradiation were inferred. At the end of irradiation gap closure has to be taken into account (see section non-destructive examinations). Considering gap closure and the calculated linear power drop from burn-up, fuel central temperature at the end of irradiation was ~1075 K and edge temperature ~800 K.

The burn-up of both fuel pins has been calculated using the

Table 1
Summary of data on the (Pu,Zr)N fuel pellets loaded in the CONFIRM capsules.

	Unit	Upper pin 30% (CONF-30-U)	Lower pin 30% (CONF-30-L)	Upper pin 20% (CONF-20-U)	Lower pin 20% (CONF-20-L)
Material		$Pu_{0.3}Zr_{0.7}N$	$Pu_{0.3}Zr_{0.7}N$	$Pu_{0.2}Zr_{0.8}N$	$Pu_{0.2}Zr_{0.8}N$
Pellet batch		N-1173	N-1171	N-1176	N-1177
Pellet density	g/cm^3	7.49	7.42	6.69	6.97
	%T.D.[a]	80.7	79.9	78.2	81.5
Pellet mean diameter	mm	5.533	5.529	5.527	5.539
Stack length	mm	72.0	70.1	67.5	68.5
Total weight	g	12.932	12.453	11.344	11.005
Total Pu content	g/cm^3	3.49	3.57	2.78	2.65
Fissile Pu content	g/cm^3[b]	3.05	3.12	2.43	2.31
C content	wt-%	0.83	0.84	1.11	0.58
O content	wt-%	0.15	0.36	0.21	0.17
N/M		0.934	0.914	0.951	0.947

[a] TD($Pu_{0.3}Zr_{0.7}N$) = 9.28 g/cm^3; TD($Pu_{0.2}Zr_{0.8}N$) = 8.55 g/cm.^{3}.
[b] Pu-vector (2003): 0.1% Pu-238, 87.2% Pu-239, 11.4% Pu-240, 0.4% Pu-241, 0.2% Pu-242, 0.8% Am-241.