Postreactor State of the Standard and Experimental BN-600 Fuel Kinds

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Abstract. Standard uranium dioxide and experimental kinds of MOX fuel operated in BN-600 reactor have been examined. In burn-up range 9-13 % FIMA uranium dioxide pellets are compatible with ChS-68 and EK-164 cladding steels: residual fuel-to-cladding gap is wide, high burn-up Rim-layer is formed, phases of cesium uranate and molybdate phases are generated, fuel swelling rate is 0.6-2.9% per 1% FIMA, maximum average porosity of pellets is 25%, depth of internal cladding corrosion does not exceed 65 μm. Globular inclusions of depleted uranium dioxide and formation of a fuel film on the cladding are typical for pellet MOX fuel. With burn-up to 11.6% FIMA strengthening of inside fuel element corrosion versus to standard fuel is not detected, moreover with a chromium getter applied at burn-up of 9% FIMA corrosion is not registered at all. Vibropac uranium gettered MOX fuel with burn-up to 10.1% FIMA does not chemically interact with ChS-68 steel cladding. However, an abnormal thermomechanical deformation of the cladding at the boundary between active core and lower blanket was observed due to getter nonuniform distribution and oxidation and high local concentration of splitted cesium as well. Minimum lattice parameter of UO_2 at the peak burn-up is 0.54587 nm and the same value for MOX fuel is 0.54521 nm.

Key Words: uranium dioxide, MOX fuel, structure, porosity.

1. Introduction

The last ten years out of 35 BN-600 reactor history show the most stable, accident-free and effective operation of the core of the third 01M2 modernization [1, 2]. The indicated period is characterized by regular control and experimental testing of standard and experimental nuclear fuel kinds in close cooperation with JSC "INM" carrying out a significant number of post irradiation examinations of fuel composition from spent fuel assemblies. The paper aimed to investigate operating capacity of standard and some experimental kinds of oxide fuel under life characteristics increase, burn-up depth and cladding materials combination in particular.

2. Research Data

The main operating characteristics of the fuel from the second 01M1 and third BN-600 core modernizations, examined at JSC "INM", are given in the Table. Active part (core column) of elements with enriched fuel is 1030 mm long. Enrichment of standard uranium dioxide fuel is 17, 21 and 26% for ^{235}U. Pu/(U+Pu) relation for MOX fuel based on depleted uranium is in the range between 22 and 25%. Lifetime of the examined fuel is 1.5-2 years and typical period of post irradiation cooling is 1-2 years.

TABLE. CHARACTERISTICS OF FUEL FOR MATERIAL-TESTING EXAMINATIONS.

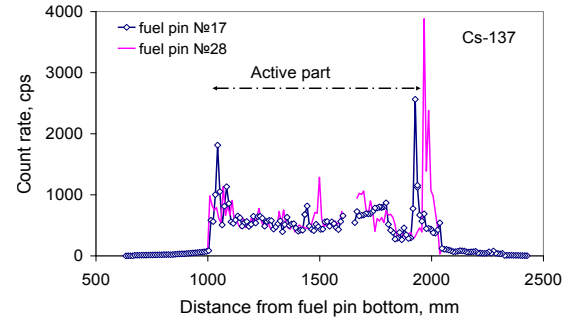
Fuel	Cladding	Maximum burn-up, % FIMA	Maximum linear load, kW/m	Maximum dose, dpa
UO_2 pellet (standard)	ChS-68-IAR* cw**	12	45	87
	EK-164-IAR cw	13	45	96
	EP-450	9	32	77
(U, Pu)O_2 pellet	ChS-68-IAR cw	12	46	85
(U, Pu)O_2+Cr pellet	ChS-68-IAR cw	9	44	58
(U, Pu)O_2+U vibropac	ChS-68-IAR cw	10	35	76
*IAR – induction arc remelting, **cw – cold-worked				

3. Research Techniques

Examinations include analysis of axial distribution of gamma-emitting fission products, swelling and fuel porosity measurements, metallography of fuel structural changes, fuel physicochemical and thermomechanical interaction with cladding material, X-ray diffraction analysis and oxygen ratio assessment after 1 year of cooling.

4. Research Results

After cooling for 1-2 years ^{137}Cs, ^{134}Cs, ^{106}Rh, ^{144}Ce, ^{144}Pr, ^{125}Sb, ^{154}Eu, ^{95}Nb, ^{95}Zr are registered in gamma-ray spectrum of fission products of standard and pellet MOX fuel in order of decreasing activity. Among mentioned nuclides highly volatile cesium tends to migrate to the junction boundaries of enriched fuel column and lower and upper blankets (*see Fig. 1a*). Other fission products follow power and burn-up shape with maximum values in the core centre (*see Fig. 1b*). Common feature for distribution behaviour is regular local concentration of fission products in gaps between fuel pellets (*see coordinates 1250-1800 mm in Fig. 1b*). There is even more intense cesium release from hot fuel to core edges and blanket region for vibropac fuel with uranium getter additive versus standard and MOX fuel without getter (*see Fig. 1c*). Similar 'self-cleaning' effect for fuel with uranium getter from splitted cesium is detected in BOR-60 reactor as well [3].



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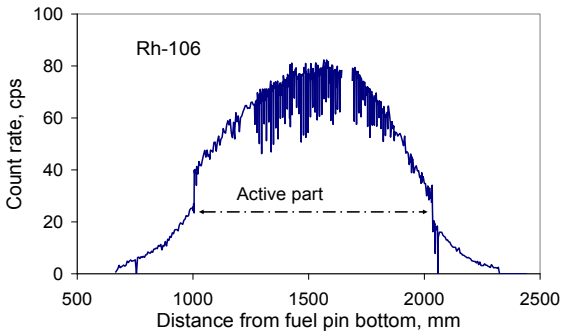

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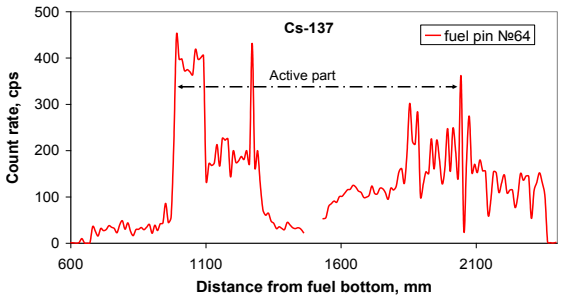


c

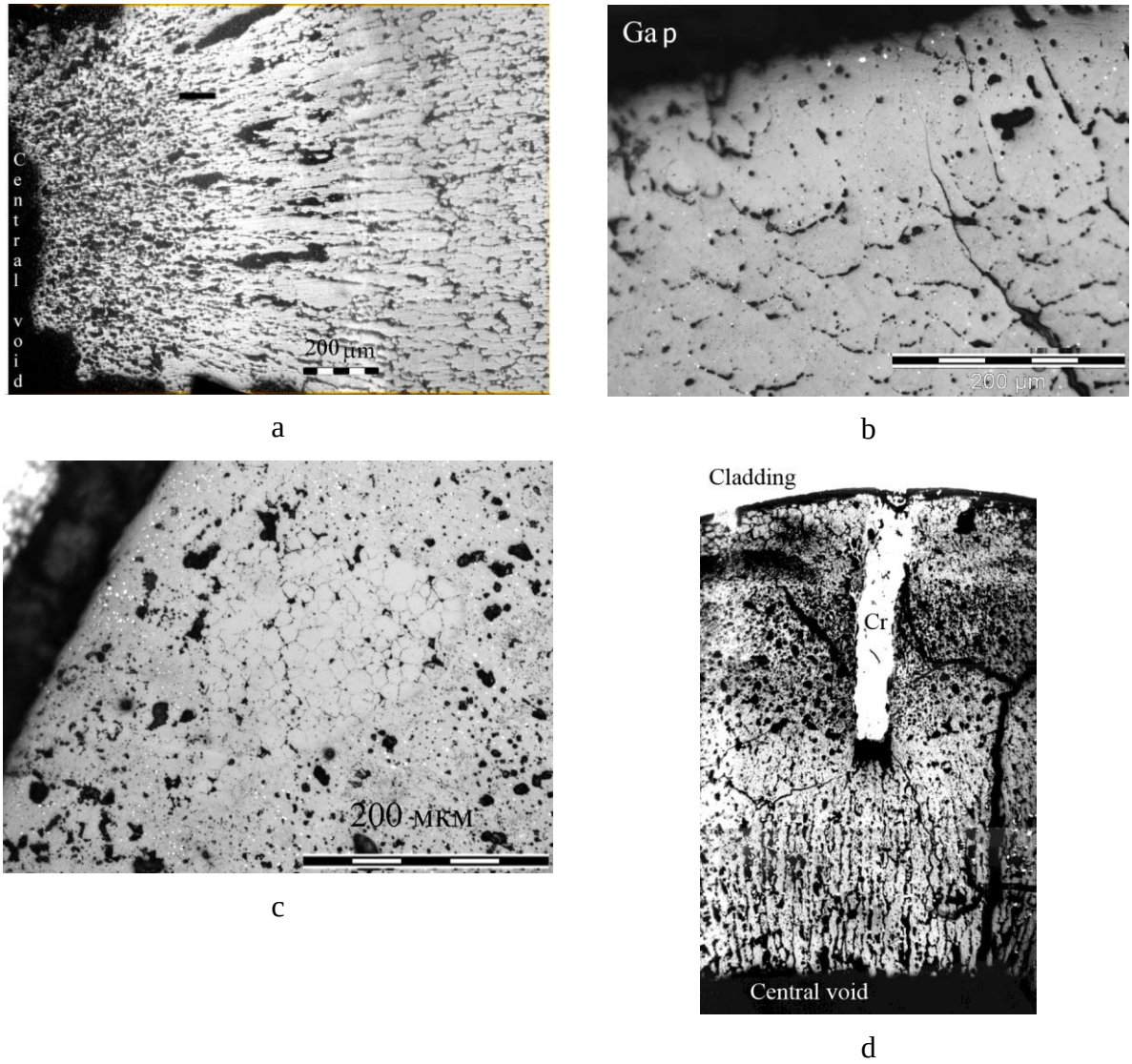
FIG. 1. Distribution of fission products along standard (a, b) and vibropac (c) fuel column

Structure of standard and experimental fuel kinds is determined by power density modes and heat removal and fuel-cladding interaction conditions. Due to low heat conductivity of all examined compositions is observed multizone structure in radial direction of fuel kernel after irradiation [4, 5]: central void, areas of columnar and equiaxed gross grains, initial grain periphery. As a rule at linear power rate beyond 25 kW/m there are no columnar grains. Otherwise they are formed sometimes in the middle of radius but around central void high-porosity recondensate migrating along the central void from hot centre to less hot areas is formed (*see Fig. 2a*).

In case of the third reactor core modernization under power rate below 45 kW/m (T<1700 ºC) the cold boundary of columnar grain area in radial direction of kernels does not increase for more than r/r_0=0.9 (r_0 – pellet radius, r – outer radius of columnar grains). An exception to this are singular cases of local heat removal faults in a wide gap or on pellet facet, which does not cause overheating or cladding depressurization (*see Fig. 2b*).

Specific aspect of MOX fuel structure in areas of low power density and pellet periphery are globular inclusions of depleted uranium dioxide without voids and traces of metal fission products. Globules appear during fuel etching aimed to identify grain boundaries (*see Fig. 2c*). Diameter of inclusions is 50-200 μm and their volume fraction is not more than 2% that makes them unregistered by X-ray analysis.

Physical and chemical interaction of standard and pellet MOX fuel with cold-worked ChS-68 steel appears as intergranular and general corrosion of cladding with depth beyond 65 μm at operating temperature above 580 ºC. Internal high-temperature pitting corrosion of EP-450 claddings at lower irradiation parameters reaches 86 μm [6]. For claddings made of EP-450 and cold-worked EK-164 steels the low-temperature corrosion from the fuel side as 56 and 55 μm deep, respectively, is additionally registered at 400−450 ºC.



a b c d

FIG. 2. Specific aspects of dioxide fuel restructuring: recondensate in central void (a), local overheating of pellet external surface (b), heterogeneous inclusion (c), getter removed from central void (d).

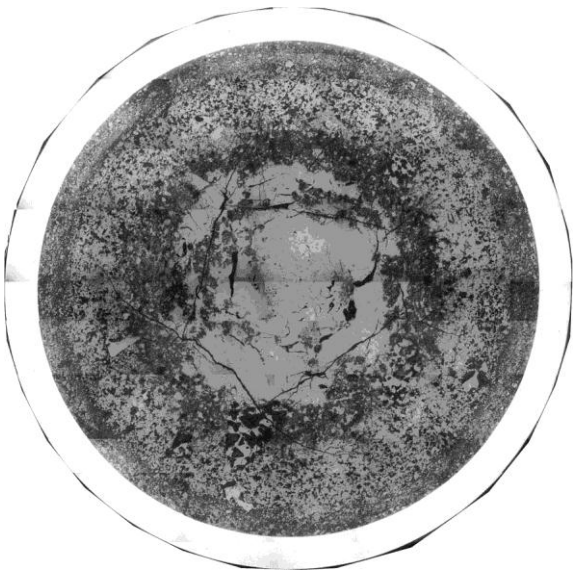
The unique case of MOX fuel irradiation with wire insert of chromium getter has showed complete absence of cladding corrosion from the fuel side all along the core column during the whole operation period. The fragments of chromium inclusion (T_{melt}=1857 ºC) after crystallization are localized under the cladding and also in cracks and fuel voids, indirectly showing the temperature of cold boundary of columnar structure development in radial direction (*see Fig. 2d*).

Structure of vibropac MOX fuel under high energy irradiation is rather conventional: central void and columnar grains are formed, initial granulate takes a narrow band on external perimeter. High-temperature corrosion of cladding from the fuel side does not occur. Uranium high density getter fraction results in its hardly predictable tendency to concentrate at the core-lower blanket boundary during fabrication. Subsequent oxidation of high uranium quantity during irradiation may be accompanied by volumetric changes of fuel composition, which cause abnormally high mechanical deformation of the cladding (*see Fig. 3a*).

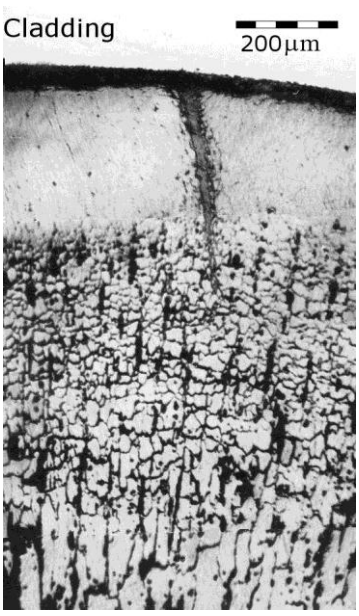
In case of pellet fuel there is a complicated dependence of kernel-cladding thermomechanical interaction. Closing of technological gap (75 μm as-fabricated wide) finishes with burn-up of

about 4% FIMA, but hot and yielding swelling fuel shows some creep features: central void diameter reduces and fuel has densification in periphery under the cladding (*see Fig. 3b*). Due to formation of porous periphery Rim-zone of high burn-up, with low mechanical strength, burn-up growth over 10% FIMA causes widening in residual gap (*see Fig. 3c*). Regions with cesium concentration on the core edges are marked with cesium uranates generating in pellet periphery (*see Fig. 3d*). A mixture of cesium molybdates with corrosion products from cladding material – so called *Joint oxide-gain* or *JOG-formation* – is sometimes registered inside technological gap in high-temperature core region (*see Fig. 3e*). Amorphous condition of cesium compounds rules out their identification by X-ray diffraction analysis. MOX fuel shows high adhesion to metal, therefore its films, up to 30 μm thick, appear at the undamaged internal surface of the cladding after the kernel is mechanically removed (*see Fig. 3f*).

Initial density of standard and pellet MOX fuel is 97% TD. Average porosity of sintered pellets is 2.5%, with only 0.4% for open voids [7]. An intense free fuel swelling up to contact with the cladding follows short stage of complete sintering during formation of initial radial structure of oxide fuel for the first hours of irradiation. After closing of technological gap at burn-up over 4% FIMA, the stage of constrained fuel swelling begins. Average constrained swelling rate for uranium dioxide during irradiation is 0.6−2.9% per 1% FIMA, with minimum rate value at 12-13% FIMA [7]. Typical behaviour for distribution of closed and general porosity of standard fuel along the core column depending on power density is given in *Fig. 4*. Evidently, at maximum linear power rate the fuel shows creep features and has densification in columnar grain areas and pellet periphery contacting with the cladding. In intermediate regions of fuel column with coordinates ~200 and 700−800 mm porosity is much higher than in the centre, therefore average swelling rate increases. At the same time in these regions central void narrowing up to 0.7 mm is registered, versus initial value of 1.75 mm [5, 7]. In case of pellet MOX fuel no significant contraction of central void along the core column is detected.



a



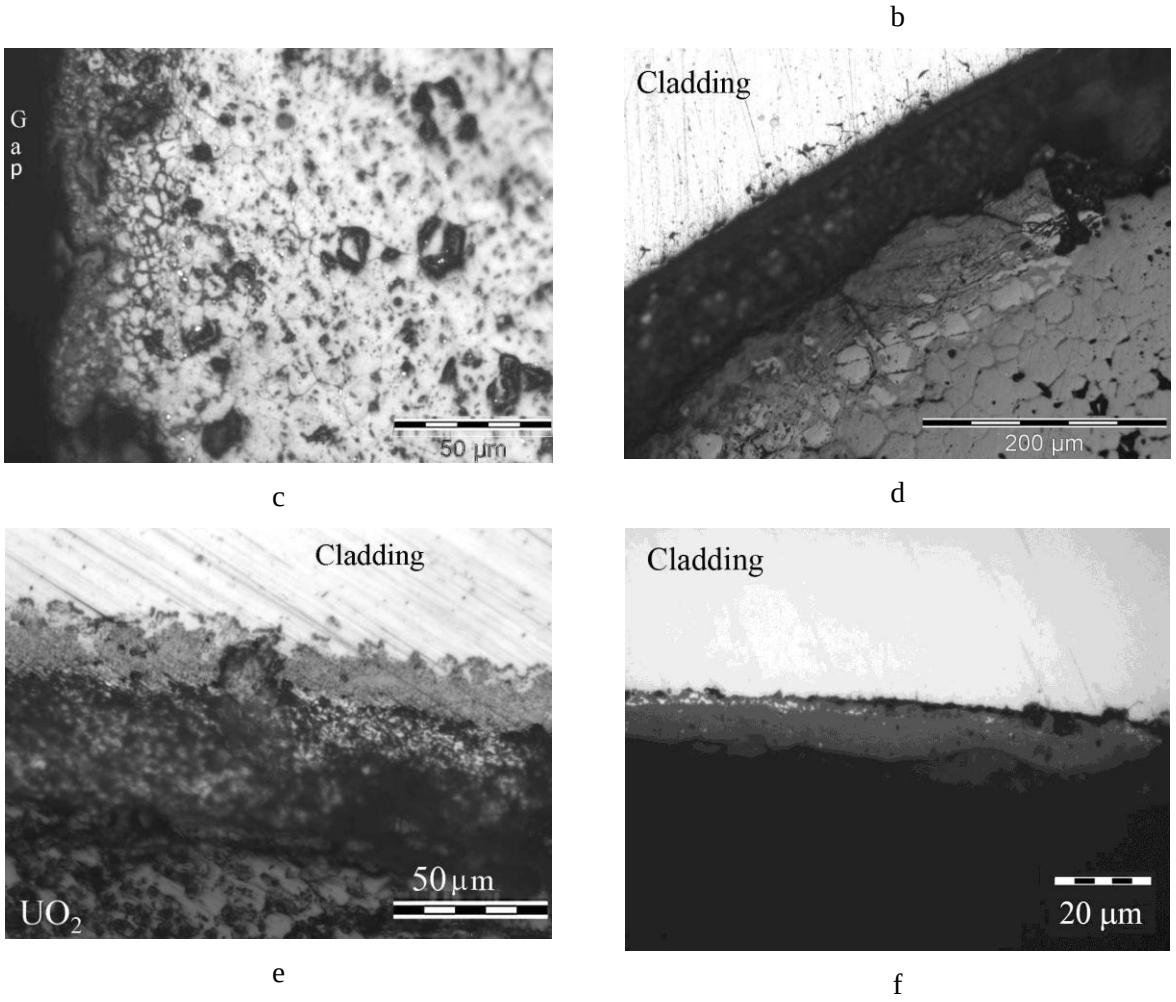


FIG. 3. Fuel-cladding thermomechanical interaction: low-porous centre core of uranium dioxide (a), densification of pellet periphery (b), Rim-layer (c), cesium uranate (d), JOG- fraction in the gap (e), MOX fuel film (f).

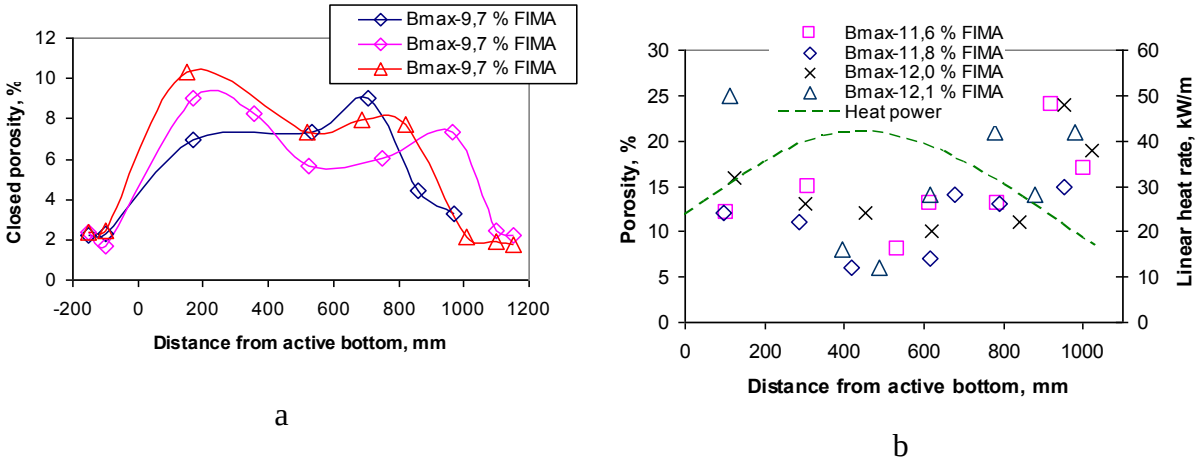
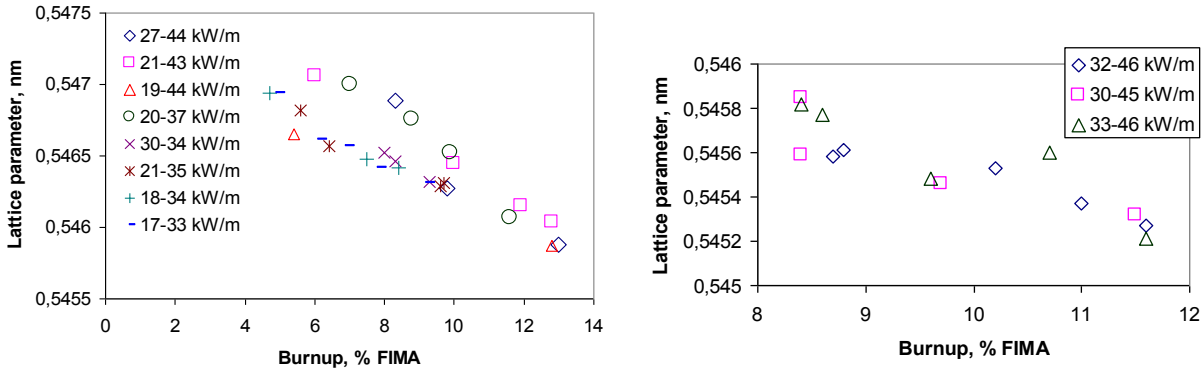


FIG. 4. Behaviour of closed (a) and general (b) porosity development along the core.

X-ray diffraction analysis of irradiated uranium dioxide and MOX fuel shows homogeneous solid solution with FCC fluorite lattice. Lattice parameter (LP) of high-temperature oxide fuel

contracts with burn-up increase due to accumulation of fission products, soluble in solid solution, and O/Me ratio increases (where Me=U+Pu).

LP of standard fuel beyond burn-up of 4-5% FIMA remains at the level of initial unirradiated state (0.54704 nm), and in the range from 5 to 13% FIMA decreases monotonically to 0.54587 nm (*see Fig. 5a*). MOX fuel with chromium getter in burn-up range of 4-9% FIMA keeps initial LP values (0.54595−0.54606 nm) [8]. Contribution of fission products is not registered or is compensated with multidirectional effect of rare earth elements. MOX fuel without getter is exposed to oxidation during irradiation, therefore at burn-up of 8.6% FIMA its LP decreases to 0.54548 nm [8], and at 11.6% FIMA of another assembly it decreases to 0.54521 nm (*see Fig, 5b*). Analysis of the latter value by LP calculation formula [9] a(nm)=0,54704–0,0077·(±0,0025)·(1–y)−0,007·(±0,001)·x for relation Pu/Me=0.22 in $(U_yPu_{1-y})O_{2+x}$ compound shows relation value for MOX fuel O/Me <2,05, regardless of fission product effect.



a b

FIG. 5. Burn-up dependence of lattice parameters for uranium dioxide (a) and MOX fuel (b)

5. Conclusion

State of standard uranium dioxide and experimental MOX fuel irradiated in BN-600 core for the last 10 years has been examined. The results of materials testing examination show quite reliable capacity and acceptable compatibility of pellet fuel with cold-worked ChS-68 and EK-164 cladding steels at attained burn-up levels of 12-13% FIMA.

Examination of standard fuel in EP-450 cladding and vibropac MOX fuel has been revealed a number of admonitions, which need additional measures to be eliminated.

6. References

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