Development of innovative fast reactor nitride fuel in Russian Federation: state-of-art

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Abstract

The nitride fuel is selected as an advanced fuel for fast reactors in Russia. Within the framework of "PRORYV" project a comprehensive program of calculation-experimental study of mixed uranium-plutonium nitride performance for BN-1200 and BREST-OD-300 reactors has been developed. The program provides for works to improve the fabrication technique, composition and structure of nitride fuel, to measure out-of-pile properties, to carry out reactor tests in the MIR, BOR-60 research reactors and in the BN-600 commercial reactor, as well as post-irradiation examination (PIE) of all experimental fuel assemblies (FA).

For the nitride fuel fabrication the carbothermal synthesis technology of nitride oxide powders, which are the product of the current radiochemical industry, is used. The laboratory technique of carbothermal synthesis of starting powders developed at JSC "VNIINM" is implemented on a larger scale at JSC "SCC" in Seversk, where the possibility of full-scale production of experimental FAs of BN-600 reactor is created. Nitride fuel pellets have been fabricated for more than 500 fuel pins for all BN-600 experimental FAs. Today 8 FAs are under irradiation in the BN-600 reactor. PIEs of one FA have been completed.7 dismountable FAs with 7 nitride pins in each are under irradiation in the BOR-60 reactor. Fuel and fuel pins have been fabricated at JSC "VNIINM".

All BOR-60 and BN-600 experimental nitride fuel pins are intact.

Key words: mixed nitride uranium-plutonium fuel, carbothermic synthesis, pre-reactor properties, experimental fuel elements, BOR-60, BN-600, BN-1200, BREST-OD-300

1 Introduction

Within the «PRORYV» project it has been developed a comprehensive program of computational and experimental validation of the mixed nitride fuel performance for BN-1200 reactors and BREST-OD-300. At this stage of the program the task is to justify the initial stages of operation of the reactor facilities. For BREST-OD-300, this corresponds to a maximum fuel burn-up of ~ 6at% with a maximum damage dose of up to 85 dpa, for BN-1200 a maximum fuel burn-up of ~ 7,5at% with a maximum damage dose of up to 95 dpa. The program includes improvement of the manufacturing technology, composition and structure of nitride fuel, pre-reactor researches of fuel, made on the technology of carbothermic synthesis, reactor testing of fuel elements at the research reactor facilities MIR, BOR-60 and the commercial reactor BN-600, post-irradiation examination of all experimental assemblies for individual programs.

2 Technological and pre-reactor researches

By 2004, JSC VNIINM has developed a laboratory technology for the production of mixed nitrides obtained by the method of hydrogenation-nitriding of metals using the vortex layer device (ABC-150), that allows to produce a high-quality powder mixture in a short time. According to this technology fuel was produced for 15 experimental fuel elements with a lead sublayer for dismountable FAs of the BOR-60 reactor. In BOR-60 two FAs with seven fuel elements each were irradiated up to maximum burn-up 4,3at% and 5,5at%, respectively. In 2008-2011 JSC VNIINM has developed and implemented a laboratory technology for the carbothermic synthesis of mixed uranium-plutonium nitride from the initial oxides and the production of fuel pellets from it also using the ABC-150 unit. More than 60 experimental fuel elements have been made in order to study the behavior of the fuel produced by carbothermic synthesis, which are currently being tested in the BOR-60. The parameters of fuel pellets, depending on the type of fuel element, varied within the following limits: density (12-13) g/cm^3, plutonium content (12-20) weight%, O_2 <0.15, C <0.15, pellet diameter (5.8 - 9.3) mm.

The laboratory technology of mixed nitride fuel with carbothermic synthesis of initial powders was implemented at JSC SCC, where it is possible to produce a full-scale experimental fuel assemblies for testing in the BN-600 reactor.

The proven technology based on experimental products was implemented on a pilot scale in a pilot production (SCC), which is planned to be used in the production of a large batch of experimental fuel assemblies in the coming years to justify the reliability of nitride mixed fuel. During testing the technology, the experience gained was used in the designing of the plant for the industrial production of nitride fuel.

According to this technology fuel pellets for more than 500 fuel elements were produced for all experimental fuel assemblies (EFA) with mixed nitride fuel, which are irradiated in the BN-600 reactor core. As a result of the improvement of the technology, modes for obtaining the initial oxides of uranium and plutonium, the processes of carbothermic synthesis and sintering were optimized, and the technological process at the manufacturing plant was formalized.

The pre-reactor researches program includes:

- researches aimed at determining the physical-mechanical properties; thermophysical, structural, diffusion properties, thermochemical stability and high-temperature creep of mixed nitride fuel;

- researches aimed at determining the missing properties of cladding material: physical-mechanical, thermophysical, high-temperature creep, long-term strength and stress rupture ductility;

- bench tests of mock-ups (fragments of mock-ups) of fuel elements, including spacer elements (small liquid metal stands).

Research methods should ensure that the data are obtained due to temperature ranges that cover the fuel operational parameters, approval of methods for confirming the measuring tolerances of the fuel and structural materials characteristics should be carried out. The analysis revealed the necessity to obtain the additional data on the properties of mixed nitride fuel (pre-reactor researches), including using the nitride which imitates a irradiated fuel: thermal creep, thermal conductivity, Young's and Poisson's modulus, thermal expansion coefficient, and studying the behavior of nitride at high temperature.

Development and improvement of computational methods of research, including codes of a new generation, require improving the accuracy of measurements of the characteristics of mixed nitride fuel using modern facilities and advanced techniques. For this purpose in 2013-2015 facilities were established and metrological approval of the main measurement methods was carried out.

Researches of pre-reactor properties were carried out in JSC VNIINM. The fabrication of the samples with different plutonium contents was carried out using mononitrides obtained from the initial oxides by carbothermic synthesis. The samples fabrication were carried out in a dry nitrogen atmosphere (H_2O, O_2 - not more than 100 ppm of each). For their production, the following were used: the vortex layer device ABC-150, the hydraulic press machine, the synthesis and sintering shaft-type furnaces. Research of the structure and composition of the experimental samples was carried out on their longitudinal sections using scanning electron microscopes-microanalysers. Six samples with plutonium contents of 5,10,12,15,25,40 mass% were unspecified chosen for the researches. Photo of the sample surface in a secondary electrons was taken at several areas of the section at various magnifications. Analysis of the obtained data on the homogeneity of the plutonium distribution indicates that there are no areas of inhomogeneity for all samples.

The thermal conductivity of samples of mixed nitride uranium-plutonium fuel was measured by the laser burst method in vacuum due to the temperature range of 673 – 1873K, plutonium content of 5-40% by mass and pellet density of 83.2 - 93.7%. The obtained curves of thermal conductivity are consistent with the foreign experimental and calculated data up to 1600K (Fig. 1). There are no foreign data for temperatures above 1600K. The thermal conductivity of nitride fuel increases with density increasing. With an increase in density from 11 to 13.5 g/cm^3, the thermal conductivity coefficient increases approximately by 70%.

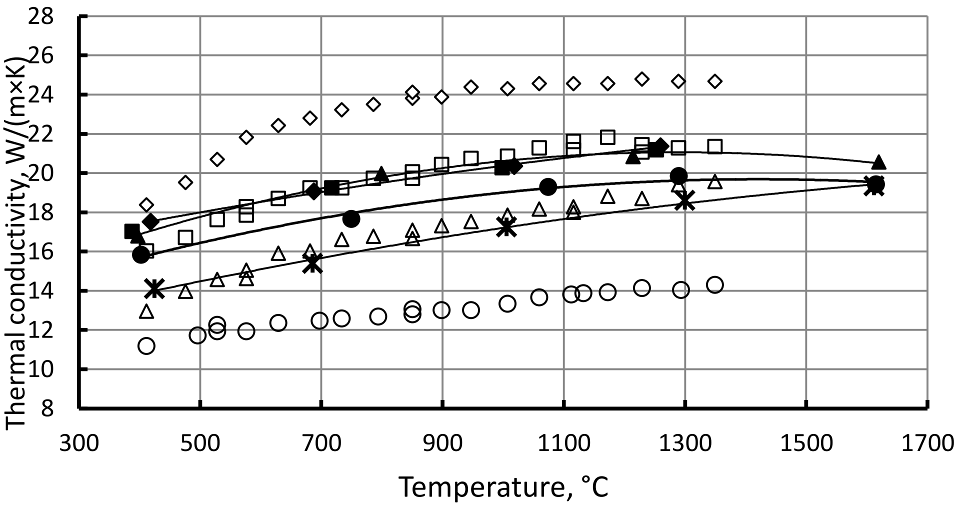

Figure 1 - Temperature dependences of thermal conductivity $(U_{1-x}Pu_x)N$, reduced to theoretical density, obtained at VNIINM (confidence bounds of the total error $\delta = \pm 5\%$) and in [1]: ◇ - UN [1], Δ - $U_{0,65}Pu_{0,35}N$ [1], ▲ -$U_{0,95}Pu_{0,05}N$ [VNIINM], ■ - $U_{0,85}Pu_{0,15}N$ [VNIINM], Ж - $U_{0,60}Pu_{0,40}N$ [4], □ - $U_{0,80}Pu_{0,20}N$ [1], ○ - PuN [1], ◆ - $U_{0,90}Pu_{0,10}N$ [VNIINM], ● - $U_{0,75}Pu_{0,25}N$ [VNIINM]

The high-temperature creep of a mixed nitride fuel with a plutonium content of 10% by mass and a density of 12.95 g/cm^3 was measured by uniaxial compression at a constant stress of 20 MPa and temperature of 1323-1723 K.

The coefficient of linear thermal expansion was measured on a high-temperature dilatometer adapted for mixed fuel, Fig.2.

The thermochemical stability of uranium nitride and mixed uranium-plutonium nitride was studied by the method [3]. Thermogravimetric researches were carried out in a dynamic helium atmosphere (purification class is 7.0 - 99.99999%) using a thermal analyzer in a glove box with an inert atmosphere and a gas cleaning system combined with a quadrupole mass spectrometer.

Mass loss of mixed nitride and uranium nitride at the isothermal time for 30 min in the range of 1800-2100 °C passes at a constant rate in one stage (Fig. 3). Mixed nitride begins to lose mass at a lower temperature than uranium nitride. Herewith, the rate of mass loss of mixed nitride at 2000 °C is greater than that of uranium nitride at 2100 °C.

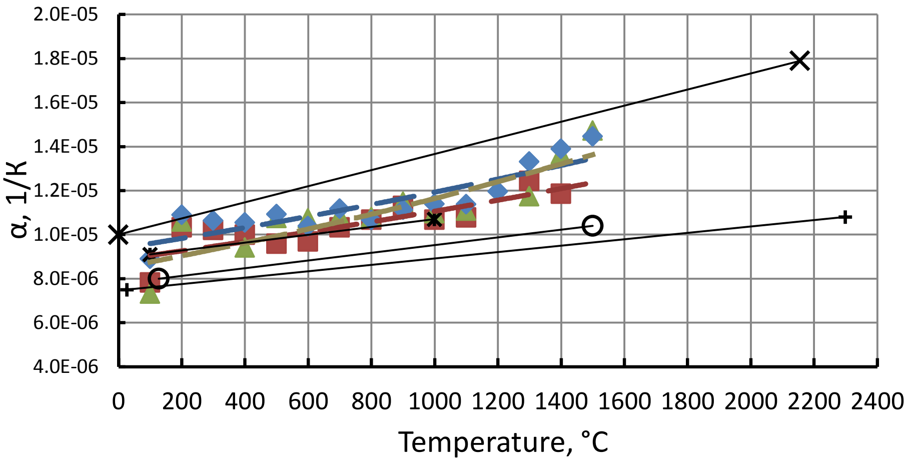

Figure 2 - Dependence of the linear thermal expansion coefficient of the samples: ◆ ($U_{0,95}$ $Pu_{0,05}$)N , ■ ($U_{0,90}$ $Pu_{0,10}$)N, ▲ ($U_{0,85}$ $Pu_{0,15}$)N from the temperature (the limit of the permissible relative tolerance of measurements of linear increments is 3%) in comparison with the literature data [2].

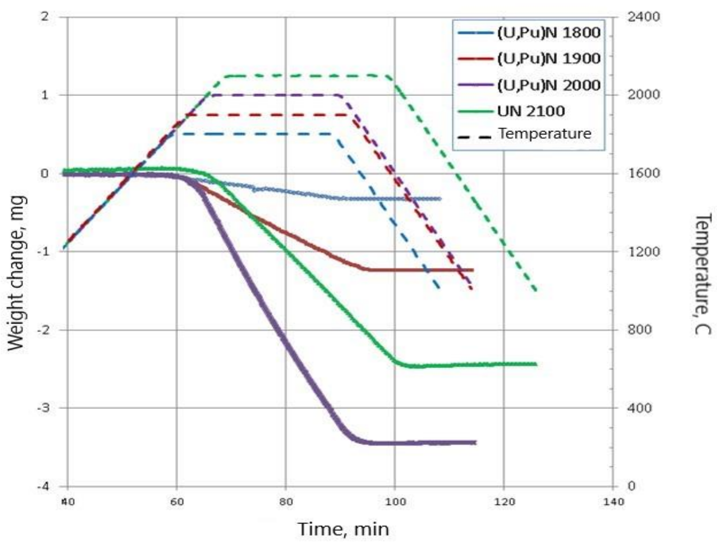

Figure 3 - Results of thermogravimetric researches of uranium nitride at the temperature of 2100°C and mixed uranium-plutonium nitride ($U_{0,5}Pu_{0,5}$)N at temperatures 1800°C, 1900°C and 2000°C.

2. Reactor testing

The purpose of the tests is to obtain the experimental data on the changes in the physical-mechanical, thermophysical, and irradiation properties of mixed nitride fuel and fuel cladding at different temperatures, damage dose and burn-up to form the correlations which are necessary to substantiate the fuel elements performance.

The main objective of the experiment in the research reactor MIR is to measure the temperature directly in the process of irradiating at the center of the fuel pellets, which will allow us to obtain the experimental data necessary for verification of the temperature model of the nitride pin with gas sublayer, as well as gas release and fuel column elongation. Currently, the researchers are carried out in the MIR reactor using an irradiation device, which includes 7 fuel elements, 6 of which are equipped with sensors for measuring the pressure of gaseous fission products under the cladding, fuel column elongation and fuel temperature. The maximum fuel temperature during testing was 1050 °C.

Tests of nitrides in the BOR-60 reactor are carried out in dismountable fuel assemblies, which are provided with the possibility of intermediate unloading of individual fuel elements for post-irradiation examinations. In the BOR-60 reactor, it is planned to test experimental fuel elements of different sizes with different clad materials, with different size of fuel-cladding gap with mixed nitride fuel of various densities made by carbothermic synthesis developed at VNIINM. Moreover, tests are planned for mixed nitride with Am and Np. At present time, 7 dismountable experimental fuel assemblies (EFA) are under irradiation in the BOR-60 reactor with 7 fuel elements in each, in EFA-4 are lead-bonded fuel elements. As of 01.05.2017 in EFA-4 fuel elements with cladding made of ferrite-martensite EP823 steel, the maximum burn-up is 4at% and maximum damage dose is 58% dpa.

In addition to obtaining the experimental data on the nitride properties, a statistically representative reactor substantiation of fuel elements with mixed nitride for BREST and BN-1200 reactors in the EFA of the BN-600 reactor is required. A total of 15 EFA were loaded into the BN-600 reactor. The irradiation of CEFA-1, CEFA-2, CEFA-3, CEFA-6, CEFA-7, EFA-4 and EFA-5 were successfully completed by the autumn of 2016. The main characteristics of these EFA are presented in Table 1. The irradiation of eight EFA continues. All fuel elements are sealed.

CEFA are, so called, combined EFA, each of which contains 4 fuel elements with nitride mixed fuel, and the rest - with oxide fuel. The design of the first fuel elements for testing in the BN-600 reactor was determined at the insistence of Rosenergoatom in such a way that the fuel element geometry and the standard cladding material ChS68-ID were used in order to increase the safety of the tests. Such tests are not fully representative for the fuel elements of BREST and BN-1200 types according to the geometry of the fuel element and the irradiation parameters, first of all, on the burn-up rate of nitride fuel. However, they make it possible to obtain data on nitride swelling and gas release in shorter time, depending on the fuel temperature at various burn-ups, and also on the corrosion compatibility of the fuel with the cladding. Due to the limited experience of irradiation of nitride fuel elements in fast reactors and the absence of such experience in the BN-600 reactor, the first irradiation of fuel elements in the BN-600 was decided to be carried out as part of a combined EFA for safety reasons.

Table 1. Parameters of EFA with mixed nitride, irradiation of which is completed in the reactor BN-600

№ EFA	Composition of EFA	Contents of the experiment	Irradiation time
CEFA-1	4 fuel elements with mixed nitride fuel and 123 standard fuel elements BN-600 with UO_2 fuel	Cladding size 6,9x0,4mm Pellet density 85% Cladding made of ChS68-ID steel Max burn-up 5,5at% Max linear power 390 W/cm	April 2013 - October 2014
CEFA-2	4 fuel elements with mixed nitride fuel and 57 fuel elements of BN-1200 type with MOX fuel	Cladding size 9,3x0,6mm Pellet density 85% Cladding made of EK164-ID steel Max burn-up 5,0at% Max linear power 440 W/cm	April 2014 April 2016
CEFA-3	4 fuel elements with mixed nitride fuel and 57 fuel elements of BREST type with UO_2 fuel	Cladding size 9,7x0,5mm Pellet density 84% Cladding made of EP823-Sh steel Max burn-up 4,5at% Max linear power 370 W/cm	April 2014 - October 2016
EFA-4	61 fuel elements of BN-1200 type with mixed nitride fuel	Cladding size 9,3x0,6mm Pellet density 85% Cladding made of EK164-ID steel Max burn-up 5,0at% Max linear power 470 W/cm	October 2014 - October 2016
EFA-5	61 fuel elements of BREST type with mixed nitride fuel	Cladding size 9,7x0,5mm Pellet density 84% Cladding made of EP823-Sh steel Max. burn-up 3at% Max. linear power 390 W/cm	October 2014 - October 2016
CEFA-6 CEFA-7	4 fuel elements with mixed nitride fuel and 123 standard fuel elements BN-600 with UO_2 fuel	Cladding size 6,9x0,4mm Pellet density 85% Cladding made of ChS68-ID steel Max. burn-up 3,8at% (CEFA-6) 7,4at% (CEFA-7) Max. linear power 390 W/cm	October 2014 - October 2015- CEFA6, October 2014 - October 2016- CEFA-7

3. Fuel post-irradiation examinations

Post-irradiation examinations of fuel element are aimed to obtain the additional data on the properties of mixed nitride: fuel swelling rate, gas release, corrosion interaction with the cladding of the ferritic-martensitic and austenite classes, ductility of the cladding under reactor irradiation and thermomechanical interaction with mixed nitride fuel.

According to comprehensive program of computational and experimental study, one fuel pin from EFA-1 of BOR-60 was unloaded with a maximum burn-up of 1.3at%, later the second fuel pin was unloaded with a maximum burn-up of 3.2at%. Currently, post-irradiation examinations of these two fuel elements with cladding made of steel EP823-Sh have been completed at JSC SRC NIIAR. Comparative post-irradiation examinations of four fuel elements with mixed nitride and neighboring driver fuel elements of CEFA-1 irradiated in BN-600 have been completed, the maximum burn-up of nitride was equal to 5.5at%, of neighboring oxide fuel pins - 7.2at%. Non-destructive examinations of CEFA-6 fuel elements have also been completed, the maximum burn -up of nitride was 3.8at%. Post-irradiation examinations confirmed that all fuel pins remained their performance.

The following main results are obtained.

1. The irradiation led to length increase of the fuel pins and the ovality of the cladding, compared with oxide fuel pins (Figures 4,5). As shown by a comparative analysis of the results obtained with the PIE data of the fuel pins of the CEFA-1, CEFA-6, and the fuel pins of the Russian-French BORA-BORA experiment [4], the fuel pins size changes were apparently caused by the thermo-mechanical interaction of fragments of the cracked fuel pellets with the cladding and do not depend on the dose and nitride burn-up. Arises in this case the inelastic deformation of the cladding does not limit the fuel elements performance, which is confirmed by the results of the BORA-BORA experiment with the maximum burn-up of mixed nitride of 12.1at%. The specific feature of pins design of BOR-60 EFA-1 and of BN-600 CEFA-1 is the increased diametrical gap between nitride pellets and the cladding (up to 0.4 mm).

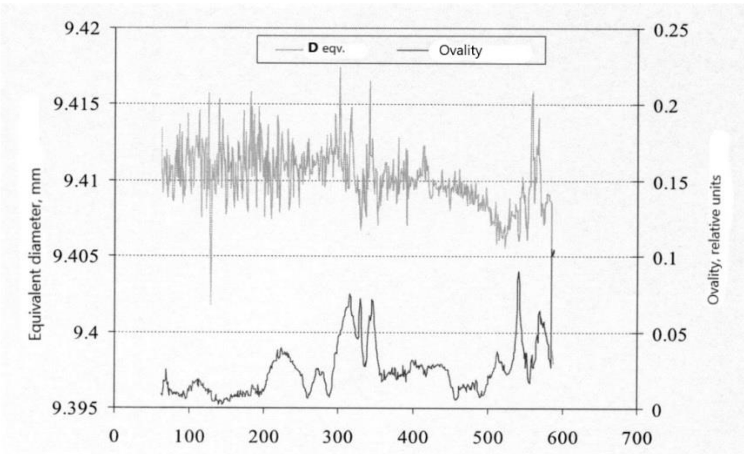

Figure 4 - Diameter and ovality changes of the cladding along the length of the fuel element of BOR-60 EFA-1 with a maximum burn-up of 1.3at%

Figure 5 - Top end of CEFA-1 after irradiation in BN-600. (Values in Figure 5 note the numbers of fuel elements with nitride fuel.)

2. Krypton and xenon release from the fuel of two fuel pins of BOR-60 EFA-1 is consistent with the results of gas release under the cladding of fuel pins of BN-600 CEFA-1 and CEFA-6, Fig. 6. The obtained results are also agreed with the data of the BR-10 reactor and with the foreign data, where the gas release increases with burn-up increase and a noticeable increase in gas release takes place after an incubation period, the duration of which depends on fuel temperature. As for all fuel elements with nitride fuel, there is increased helium forming and its release under the cladding, which should be taken into account at fuel performance proving

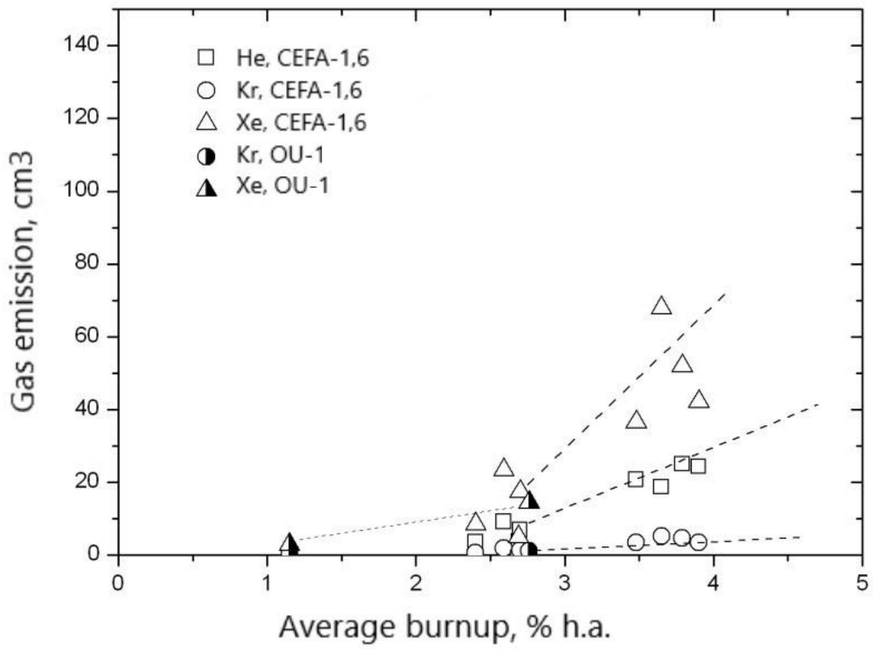

Figure 6 – Gas release from the nitride of BOR-60 EFA-1 (OU-1), CEFA-1 and CEFA-6 of BN-600

3 Metallographic researches have shown that the claddings of both types of fuel elements (nitride and oxide) of CEFA-1 had the corrosion damage from the internal side. The type of the corrosion of the cladding of neighboring fuel elements with oxide and nitride fuel of CEFA-1 irradiated the BN-600 reactor up to burn-up of 7.3at%(oxide) and 5,5at % (nitride) differs. The maximum depth of corrosion damage to the nitride cladding on the fuel side was 150 μm, with a maximum corrosion depth of 50 μm for oxide fuel elements. The type and depth of corrosion of the oxide pins correspond to the data obtained from the research of a large number of standard fuel elements of the BN-600 reactor with cladding made of ChS68-ID steel. Features of corrosion of fuel elements with nitride fuel are its greater depth, ulcerous character, and also the presence of corrosion damage in the lower parts of the fuel elements. In addition to the corrosion, the effect of nitride fuel on the cladding of fuel element led to the nitriding of its internal surface. The depth of the nitrided layer can be as high as 30-50 μm according to X-ray microanalysis and microhardness measurements.

Corrosion of the cladding on the nitride side did not exceed 50 μm for fuel elements of BOR-60 EFA-1 with a ferritic-martensitic cladding EP823-Sh. Correlation of corrosion depth with burn-up was not found. Nitriding of the inner surface of the cladding was observed only for the fuel element with maximum burn-up and did not exceed 10 μm. The detected nitriding in the sample M3 of the fuel element № 7 did not affect on the mechanical properties of the cladding in the upper part of the fuel element.

3 Tests of annular cladding samples of the fuel elements with nitride and oxide of CEFA-1 showed a noticeable difference in their mechanical properties. When tested under the identical conditions, the fuel element oxide cladding showed higher strength and plastic characteristics, compared to the nitride cladding.

The obtained data on the mechanical properties of the cladding made of EP823 steel of two fuel elements EFA-1 correspond to the previously obtained data for EP-823 and EP-450 steels irradiated as cladding material with oxide fuel. Steels ductility decreasing in the lower parts of fuel elements is caused by low-temperature irradiation embrittlement, which appears in the irradiation temperature range of 280–460°C. Further increase in temperature does not lead to ductility decrease, even at doses of 80-90 dpa. It should be noted that the minimum cladding temperature for the fuel element of the BREST reactor is 460 °C, and thus the low temperature irradiation embrittlement factor will be minimized in these fuel elements.

4 Based on the results of the computational researches of the CEFA-1 fuel pins with real irradiation history, conducted by the codes DRAKON and BERKUT, it can be concluded that, in general, the expected results were obtained for the mixed nitride swelling, gas release, cladding deformation, which is important from the point of view of adequate prediction of fuel life time. The results of the pretest calculations of the expected post irradiation examinations results of CEFA-6 by the DRAKON and BERKUT codes correspond to the non-destructive examinations data on gas release, cladding deformation, fuel-cladding gap presence and pin integrity.

Conclusion

The development of uranium-plutonium nitride fuel is carried out in accordance with the comprehensive program of computational and experimental validation of the performance of mixed nitride fuel for BN-1200 and BREST-OD-300.

It has been developed the technology of manufacturing pallets of mixed uranium-plutonium nitride fuel by carbothermic synthesis method. The laboratory technology of mixed nitride fuel with carbothermic synthesis of initial powders was implemented at JSC SCC, where it is possible to produce a full-scale experimental fuel assembly for testing in the BN-600 reactor. More than 500 fuel elements were produced using this technology for testing in MIR, BOR-60 and BN-600 reactors. Researches are continuing to improve the composition and structure of MNUP fuel in order to increase ductility, reduce crack resistance and fuel swelling speed.

According to the results of proven technology at SCC it has been created a pilot experimental technological line and construction of industrial production is underway, the launch is scheduled for 2020.

It has been carried out a complex of pre-reactor researches of the physicomechanical properties of mixed nitride fuel, the results of which make it possible to perform the minimum necessary justification for the reactor tests of experimental fuel elements. Researches will continue to obtain data in the required temperature range depending on the density, composition, and structure of the fuel composition.

In accordance with the comprehensive program of computational and experimental validation of dense fuel:

- tests of experimental fuel elements are carried out in the reactor MIR with mixed nitride fuel, equipped with sensors for in-reactor control of fuel temperature, extension of the fuel core and gas emission from fuel;

- more than 60 fuel elements of different modifications with mixed nitride fuel were loaded into the BOR-60 reactor with the dismountable irradiation devices;

- 15 experimental fuel assemblies with mixed nitride fuel were delivered to the BN-600, 7 of which were irradiated and completed in a planned manner without signs of unsealing, the irradiation of the remaining 8 EFA with mixed nitride fuel is going on.

Currently, SRC NIIAR has completed post-reactor researches of two fuel elements with cladding from EP823-SH steel with a maximum burnup of 3.2% h.a., and a maximum dose of 48 dpa. Comparative post-reactor researches of four fuel elements with MNUP fuel and neighboring standard fuel elements CEFA-1 and BN-600 have been completed, the maximum burnup of nitride fuel was 5.5% h.a, fuel oxide of neighboring fuel elements was 7.2% h.a., a maximum dose of 55 dpa. Primary researches of CEFA-6 fuel elements have also been completed, the maximum burnup of MNUP fuel was 3.8% h.a. Post-reactor researches confirmed that all fuel elements remained their work capacity.

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