RERTR 2022 – 42ND INTERNATIONAL MEETING ON
REDUCED ENRICHMENT FOR RESEARCH AND TEST REACTORS

OCTOBER 3-5, 2022
VIENNA INTERNATIONAL CENTRE
VIENNA, AUSTRIA

LOW ENRICHED NUCLEAR FUEL BASED ON URANIUM-ZIRCONIUM CARBONITRIDE: REACTOR TESTS AND POST-REACTOR STUDIES

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ABSTRACT

The creation of energy-intensive nuclear power plants requires the use of nuclear fuel capable of withstanding various effects of neutron fields, high temperatures and thermal stresses during operation. At present, such a fuel can be the uranium-zirconium carbonitride (UZrCN) with a low oxygen content (less than 0.1 wt. %), developed at the LUCH JSC, State Atomic Energy Corporation "Rosatom". Uranium-zirconium carbonitride combines the positive qualities of fuels based on uranium carbides and nitrides, namely: the heat conduction of UZrCN – based fuel is almost 10 times higher, the ultimate stress is almost 3 times higher, and the volumetric swelling is 3 times lower than that of UO_2; compared to UN, it has a reduced yield of gaseous fission products, less tendency to swell and a significantly higher operating temperature; compared to UC it has increased compatibility with structural materials. Due to its optimal characteristics, UZrCN is an attractive fuel material for use in various types of reactors. The limited use of this type of fuel is due to the small amount of information about the behavior and characteristics of UZrCN under irradiation, especially at deep burnup levels. To eliminate this problem, in the framework of RRRFR programme an international project is currently being implemented to study the properties and characteristics of UZrCN in conditions of irradiation to reach 5, 15, 40% fifa burnup with the energy release up to 600 W/cm^3. The project have been implemented step-by-step through the solution of a series of research and computational and experimental tasks. Starting from 2016, the tasks of developing the design of the experimental capsule and the irradiation device have been solved; for this, neutron-physical, thermophysical and strength computations of the experimental capsule and the irradiation device have been carried out, and a

program of out-of-pile experiments has been implemented. In 2019, a methodical experiment has been carried out in the SM-3 research reactor (JSC “SSC RIAR”, State Atomic Energy Corporation “Rosatom”) with a duration of 23.3 eff. days with the 0.63% fifa burnup reached and post-reactor studies of the irradiated fuel have been performed. Starting from July 2021, systematic preparations have been made for a reactor experiment to study the properties and characteristics of UZrCN in conditions of irradiation until the 5% fifa burnup has been reached. As a result of the work carried out, an experimental capsule, an irradiation device have been manufactured, and on December 19, 2021, a reactor experiment with a duration of 200 eff. days has started.

The critical facilities “Giacint” and “Kristal”, located in the scientific institution “JIPNR – Sosny”, National Academy of Science of Belarus are used for the preparation of critical facility experiments on the multiplication units, simulating physical characteristics of the cores using low-enriched UZrCN-based nuclear fuel (19,75% U-235) so as to use them in the reactors on fast and intermediate neutrons with gas or liquid-metal coolants. It is planned to investigate the fast-neutron critical assemblies with three types of fuel cassettes and different matrix materials (air, aluminum and lead). Also, it is planned to investigate the intermediate-neutron critical assemblies with a hydride-zirconium moderator. The side reflectors of these critical assemblies are beryllium (internal layer) and stainless steel (external layer).

The report describes in detail the results of the work carried out on irradiation of UZrCN in the SM-3 research reactor, as well as a description of the design and composition of critical assemblies with UZrCN- based fuel, and the results of calculations.

1 Introduction

The creation of energy-intensive nuclear power plants requires the use of nuclear fuel capable of withstanding various effects of neutron fields, high temperatures and thermal stresses during operation. At the moment, uranium nitride, uranium carbide, uranium sulfide and uranium silicide are considered as such fuel. In this case, the main attention is paid to uranium nitride. Fuel based on uranium nitride (UN) has high strength and thermal properties (in the range of operating temperatures, its thermal conductivity is ~ 7 times higher than that of UO_2), a high fissile element density (1.3 times higher than that of UO_2), therefore it is considered as a potential replacement for uranium dioxide [1]. However, this type of fuel has a significant drawback that affects its performance – low thermochemical stability. At elevated temperatures (above 1950 K in N_2-free atmospheres), the UN-based fuel dissociates, and the precipitation of the uranium metal phase is observed [1].

JSC "NII NPO "LUCH" has developed a fuel based on uranium-zirconium carbonitride (CNT). This fuel is a modification of UN, which retains its positive qualities while being thermochemically stable at temperatures up to 2500 K [1]. To stabilize the phase composition and expand the region of homogeneity, a dope additive in the form of zirconium was introduced into the UN fuel, carbon is necessary to block thermal dissociation. Uranium-zirconium carbonitride combines the positive qualities of fuels based on uranium carbides and nitrides. Comparison of characteristics of fuels based on UO_2, UN, UZrCN is shown in Table 1.

Due to its optimal characteristics, UZrCN is an attractive fuel material for use in various types of reactors. The limited use of this type of fuel is due to the small amount of information about the behavior and characteristics of CNT under irradiation, especially at deep burnup levels. To eliminate this problem, within the framework of the RRRFR program, a project is currently being implemented to study the properties and characteristics of CNTs under irradiation conditions to achieve burnups of 5, 15, and 40% fifa in the SM-3 high-flux research reactor (Dimitrovgrad, Russia). The parameters of the reactor experiments are given in Table 2.

Table 1 – Comparison of key characteristics of fuels [1,2]

Characteristic	UO_2	UN	U-Zr-C-N
Theoretical nucleus density of U, g/cm^3	10,97	13,5	12,8
Thermal conductivity, W/m·K	3,0	26,0	32,0
Melting point, K	3078	3123	3470
The content of oxygen impurity, wt.%	—	< 0,15	< 0, 1

Table 2 - Parameters of the reactor experiment

Fuel	U – Zr – C – N
T of fuel	not more than 1600 К
Energy release, W/cm^3	до 600
Burnup (fifa), %	5, 15, ≤ 40
Duration of the reactor experiment, year	≤ 5

The critical facilities "Giacint" and "Kristal", located in the scientific institution "JIPNR – Sosny", Minsk, Belarus [3, 4], are used for the preparation of critical facility experiments on the multiplication units, simulating physical characteristics of the cores using low-enriched UZrCN-based nuclear fuel (19.75% 235U) so as to use them in the reactors on fast and intermediate neutrons with gas or liquid-metal coolants.

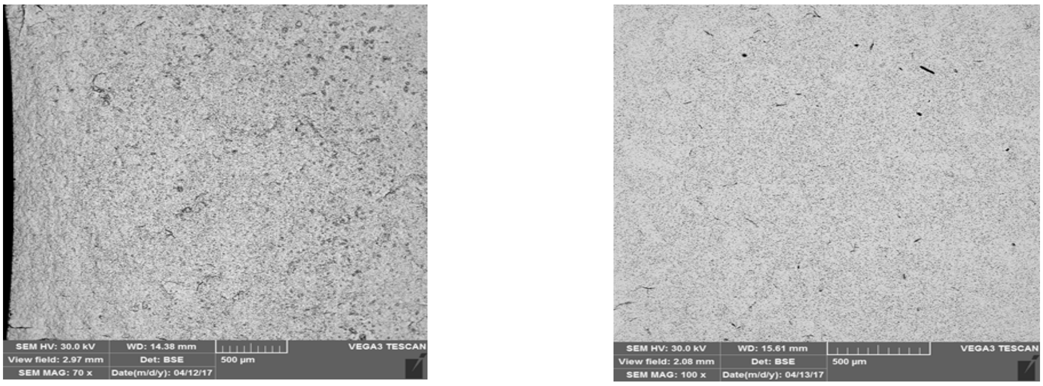
2 Reactor Tests of Low Enrichment Nuclear Fuel Based on Uranium-Zirconium Carbonitride

2.1. Description of the Investigated Fuel

To implement the program for conducting reactor experiments, a batch of fuel pellets was manufactured and produced. Fuel pellets with a height of 5 mm and a diameter of 8 mm have an enrichment of 19.75% in the U-235 isotope, the chemical composition of UZrCN (equimolar fuel composition for light components), a density of 12.3 g/cm^3. It should be noted that the resulting fuel has a high purity of oxygen impurities (less than 0.1 wt. %). The results of structural-phase studies of the manufactured fuel pellets in the initial state are shown in Figure 1. Cross sections of the fuel pellet in the initial state characterize the fuel as sufficiently dense and homogeneous without macro- and microdefects throughout the fracture and thin section (Fig. 1). Pores of a spherical shape with a size of 1-1.5 μm are homogeniously distributed over the cross section of the pellet and are localized mainly in the body of the grain (Fig. 1). The fuel microstructure appears to be homogeneous with equiaxed polyhedral grains 10–15 μm in size. The fracture surface characterizes it as mixed with the advantage of a transcrystalline fracture type (Fig. 1a). X-ray phase analysis has showed that the material of the fuel pellet consists of a single phase – a solid solution of U, Zr (C,N).

2.2. Design of the Experimental Capsule (EC) and the Irradiation Device

To implement reactor tests of UZrCN fuel pellets, an experimental capsule (EC) design was developed (Fig. 2). The experimental capsule contains 17 uranium-zirconium carbonitride pellets 8 mm in diameter and 5 mm thick (13 test pellets, two low-enriched auxiliary pellets (in the lower part of the column) to increase the uniformity of the temperature field, and two pellets with a central hole for inserting thermoelectric transducer (TET)). All pellets are installed by a tight fit in a non-sealed protective tungsten three-section shell with a thickness of 2.0 mm. A sealed molybdenum housing is outside the tungsten shell. A protective tungsten shell is necessary to eliminate the contact interaction of carbonitride fuel with a molybdenum housing, and it also helps to reduce the unhomogenious distribution of temperature along the height of the test fuel pellets. The EC cavity has been initially filled with helium at a pressure of 0.1-0.2 MPa. A compensation volume is provided in the lower part of the EC.



(a) (б)

Figure 1 – General view of the cross section of the fuel pellet in the initial state:
a) fracture section; b) thin section.

TETs are installed on the molybdenum housing and in the second (counting from the top) pellet for reliable control of the CNT temperature.

The irradiation device (ID) consists of a sealed EC located inside a protective stainless steel casing. During irradiation, the outer steel casing is in contact with water in the primary circuit of the SM-3 reactor. Reactor tests are carried out in channels 10 and 11 of the second group of reflectors of the SM-3 research reactor.

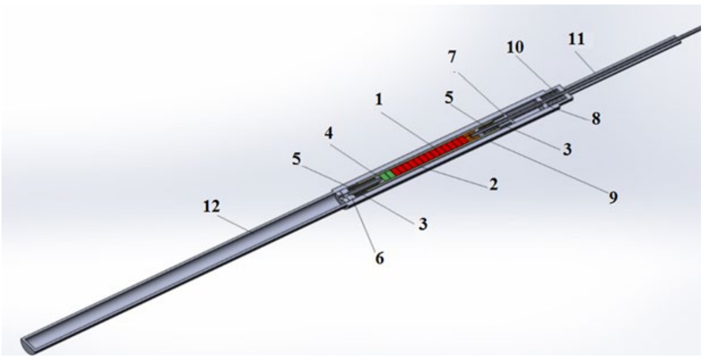


Figure 2 – Schematic representation of the experimental capsule:
1 - fuel column, 2 - tungsten shell, 3 - end springs (4 pcs.), 4 - tungsten table, 5 - Mo tube, 6 - lower molybdenum tube spacer, 7 - TET housing internal fixator (W), 8 - upper molybdenum tube spacer, 9 - sealed molybdenum housing, 10 - temperature compensation unit, 11 - Mo tube for TET output, 12 - volume for collecting gaseous fission products (GFP).

2.3. Confirmation Calculations

After developing the designs of the experimental capsule and the irradiation device using the MCU-RR computer program [5], which implements the algorithm for solving the neutron transfer equation by the Monte-Carlo method, neutron-physical calculations of the experimental parameters have been carried out using the geometric parameters of the specimens. The main task of neutron-physical calculations has been to determine the geometry of the absorbing hafnium screen, which is necessary to ensure the maximum energy release in the fuel up to 600 W/cm^3. For calculations, the core conditions corresponding to the middle and end points of the campaign have been used. The results of neutron-physical calculations have been used to determine the wall thickness and outer diameter of the hafnium screen and are equal to 2.5 and 59.5 mm, respectively. The dimensions of the screen have been chosen from the conditions for ensuring homogenous removal of heat released in the primary circuit by water. The results of neutron-physical calculations of energy releases in ID materials are given in Table 3.

Table 3 - Calculated value of energy releases in ID materials

Object	Calculated energy release, W/cm^3
UZrCN	509,9
W	64,1
Mo	27,83
Steel	17,71

On the basis of neutron-physical calculations, verification calculations of the temperature fields of the experimental capsule have been carried out. The calculations have been carried out using the ANSYS Mechanical finite element analysis software package [6]. When carrying out thermophysical and strength calculations, it is necessary to go by the conditions of the reactor experiment. Thermophysical calculations have been carried out in order to determine the temperature regime of the EC during irradiation and its compliance with the requirements of the specification. The calculation results are shown in Fig. 3.

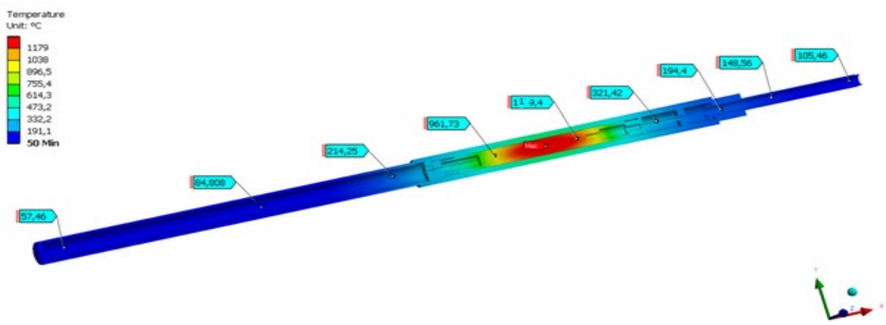


Figure 3 – Temperature field of the experimental capsule

It follows from Figure 3 that at a given level of energy release in the EC elements and the selected EC layout, the temperature in the center of the fuel pellet meets the requirements for the experiment.

The reactor experiment will be terminated when the GFP is fixed in the gas path of the SM-3 reactor; therefore, it is necessary to determine the level of stresses arising on the sealed Mo housing. Fig. 4 shows the results of calculating the equivalent thermal stress in the Mo housing (MPa). The level of the obtained stress values in the sealed housing outside the concentration zones does not exceed 94 MPa, which is significantly less than the yield strength of pure molybdenum. On this basis it can be concluded that under the indicated operational loads, the plastic flow of the capsule housing material will not occur, and the strength of the sealed housing is ensured.

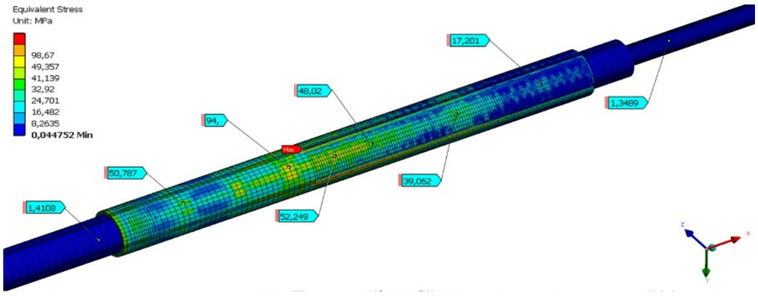


Figure 4 – Equivalent stresses in Mo housing

2.4. Methodological Experiment

A short-term methodical reactor experiment has been implemented as a preparation for long-term reactor experiments and confirmation of thermophysical calculations. Methodical reactor tests of the “ID CNT” irradiation device with an experimental capsule with uranium-zirconium carbonitride fuel have been carried out in cell No. 11 of the reflector of the SM-3 reactor from 06/01/2019 to 06/29/2019. The total exposure time was 23.28 effective days.

Changes in the temperature of the fuel, the molybdenum shell of the experimental capsule, and the pressure in the cavities of the ampoule and hanger during irradiation are shown in Fig. 5.

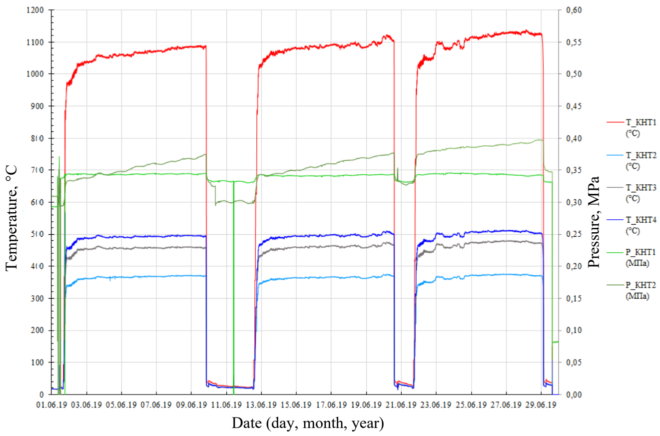


Figure 5 – Change in temperature and pressure in the irradiation device "ID CNT"

As can be seen from the graph shown in Figure 6, the fuel temperature has increased during the experiment. The increase in fuel pellet temperature is due to two factors. In each campaign of the SM-3 reactor facility, when the reactor is operated at a constant power level, there is a constant increase in the energy release and neutron flux density in the reflector channels due to the movement of the compensating elements [6]. Also, the increase in the temperature of the fuel pellet is due to a change in the thermal conductivity of the gaseous medium inside the experimental capsule due to the release of gaseous fission products from the fuel and, possibly, the release of nitrogen, which have a thermal conductivity several times lower than that of helium. In addition, it should be taken into account that the neutron flux density and energy release depend on the distribution of fuel burnup in the reactor core and on the loading of experimental channels, which change every campaign.

The average value of the volumetric energy release in the test pellets has been 516 W/cm^3 during the methodical reactor tests. The burnup achieved in the test tablets is 0.63% fifa.

2.5. Post-Reactor Studies

After conducting a methodical reactor experiment, a program of post-reactor studies of irradiated specimens has been implemented.

2.5.1. Results of Gamma Scanning of the Experimental Capsule

Fig. 6 shows the counting rate distributions for the fission product lines of Zr-95, Cs-137, and Nb-95 obtained by gamma-scanning the experimental capsule. The origin of coordinates on the diagram corresponds to the bottom edge of the capsule.

Based on the intensity distributions of the nuclides, the location of the fuel column and Cs-137 in the experimental capsule have been determined. As can be seen from the figure, all Cs-137 produced during irradiation completely remained in the fuel, that indicates the hermeticity of the EC during the experiment.

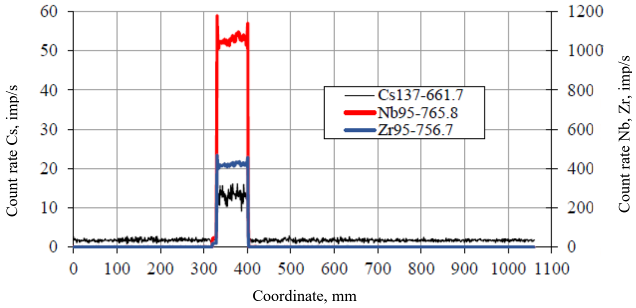


Figure 6 – Intensity distribution of the lines of some fission products along the length of the experimental capsule

2.5.2. Results of X-Ray Crystallographic Analysis of Fuel Pellets

X-ray diffraction analysis of the fuel has been carried out on specimens of two pellets located at the top of the fuel column. The X-ray patterns obtained as a result of the study make it possible to identify a cubic face-centered lattice (Fm-3m space group). No maxima belonging to other structures have been found in the X-ray diffraction patterns. The crystal lattice parameter (a) is 4.892 ± 0.0005 Å. Thus, according to the nameplate data, the phase composition of the fuel under irradiation have not changed.

2.5.3. Results of Studies *of Fuel Pellet Structure after Irradiation*

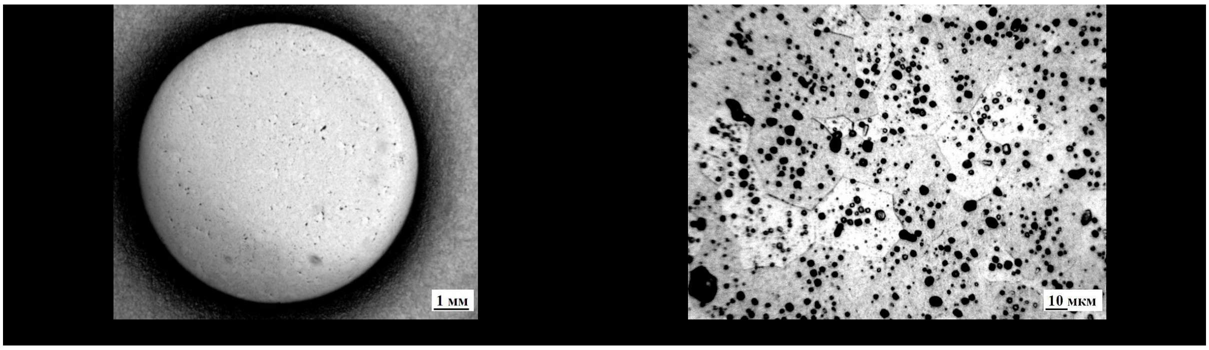
Metallographic analysis of the fuel structure has been performed on pellets located in the center of the fuel column.

The metallographic analysis of the fuel structure of this specimen has been performed at two levels along the pellet height: near the surface and closer to the pellet center. For these purposes, after studying the structure near the pellet surface, the specimen has been again subjected to grinding to a depth of about 1 mm, then polished and chemically etched.

The general view of the specimen after the first grinding, which has been carried out to level the surface of the pellet and to polish is shown in Figure 8. There are no cracks in the body of the pellet, the macrostructure of the fuel is almost homogenious in the cross section of the pellet and is shown in Figure 7.

2.5.4. Reactor experiment (5% fifa)

The results of the work on conducting a methodical reactor experiment have demonstrated the operability of the selected layout of the experimental capsule and irradiation device, the compliance of the experimental conditions with the specification requirements, and a high correlation of experimental and calculated data. Based on the foregoing, starting from July 2021, systematic preparations have been made for a reactor experiment to study the properties and characteristics of CNT under irradiation conditions until a burnup depth of 5% fifa. As a result of the work in 2021, according to the reissued design documentation (minor changes have been made to the design of the experimental capsule – the lower weld has been reinforced, and a spacer has been installed on the gas collector), the EC sealing unit, the experimental capsule and the irradiation device have been manufactured, and on December 19, 2021, the reactor experiment has been started with duration of 200 eff. days. Fig. 8 represents the first data on the temperature of fuel pellets and the pressure in the ampoule under ionizing radiation conditions.



(a) (b)

Figure 7 – Results of material science studies of irradiated pellets:
a) macro shot of a pellet after the first grinding and polishing; b) the microstructure of the fuel in the center of the pellet after the first grinding and etching

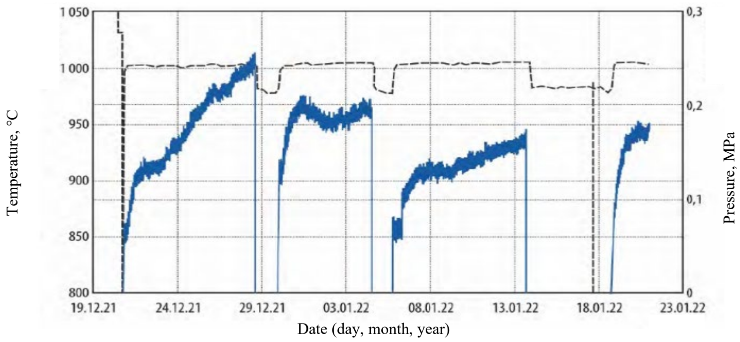


Figure 8 – Measurements of the temperature of fuel pellets (-) and pressure (---) in the ampoule during testing [7]

In March 2022, the reactor experiment has been stopped and the irradiation device has been moved to the spent fuel pool of the SM-3 reactor plant.

3 The Program of Experiments on the Fast and Intermediate-Neutron Critical Assemblies with 19.75% Enriched UZrCN Fuel at the Critical Facilities "Giacint" and "Kristal"

3.1. Fast-Neutron Critical Assemblies

At critical facilities "Giacint" and "Kristal" is planned to investigate fast-neutron critical assemblies with three types of fuel cassettes and different matrix materials (air, aluminum and lead). All of these critical assemblies will use Type 1 and Type 2 fuel rods.

Type 1 (Type 2) fuel rod consist of a fuel section, a cladding and end pieces (Fig. 9). The fuel section consist of cylindrical pellets, 10.75 mm in diameter, from uranium-zirconium carbonitride $U_{0,9}Zr_{0,1}C_{0,5}N_{0,5}$. The overall length of the fuel section is 500 mm. Ligature weight of the fuel section 540 g, overall uranium weight 489 g, the mass of U-235 is 96.7 g. Gaps between the fuel section pellets and the fuel rod cladding have a gas helium medium at 0.11–0.12 MPa. The overall length of the fuel rods is 620 mm. The lower end piece of Type 1 fuel rod consist of a lower plugs, a spring, gaskets and a pin. The upper end piece of the fuel rod consist of an upper plug (stainless steel). The fuel rod cladding is made from a tube (stainless steel) with the 12 mm outer diameter and the 0.6 mm thick wall. The material of the cladding, the lower plug and the upper end piece of Type 2 fuel rod is the niobium alloy.

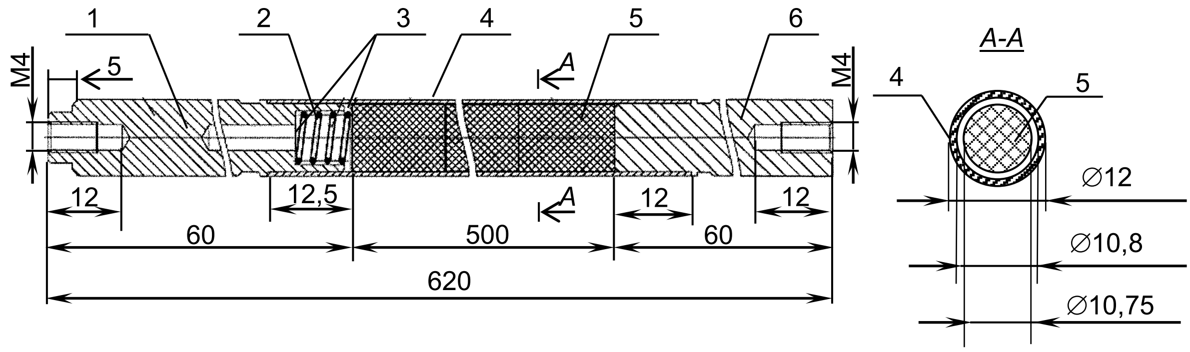


Figure. 9 – Type 1 (Type 2) fuel rod:
1 – lower plug; 2 – spring; 3 – gasket; 4 – cladding; 5 – fuel section; 6 – upper plug

Fast critical assemblies, simulate physical features of the cores of the future fast-neutron reactors with gaseous (F-20-A) and liquid-metal (F-20-L, F-20-AL) coolants. This fast critical assemblies represent a lattice of fuel cassettes with fuel rods with air or lead or aluminum as matrix materials, with a beryllium-steel reflector.

Description of the design, composition and calculation results of fast critical assemblies are presented in [8].The experiments are to be performed on three fast critical assemblies, the loading cartograms of which are shown in the Fig.10.

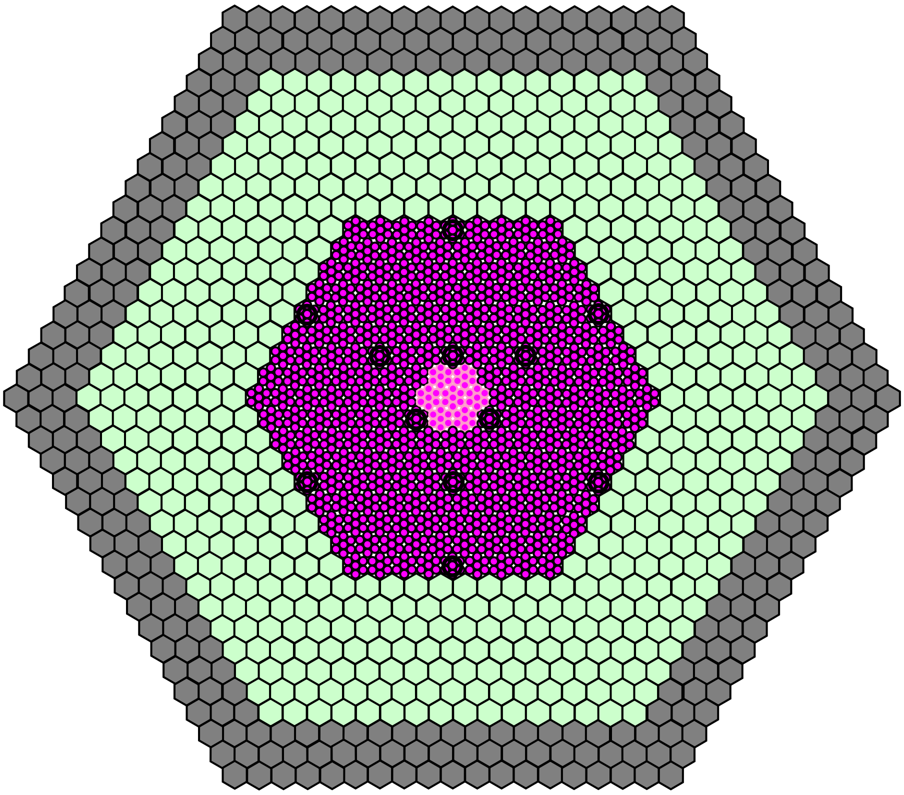
3.2. Intermediate-Neutron Critical Assemblies

Intermediate-neutron critical assemblies H-20-1 and H-20-2 with a hydride zirconium moderator contain fuel cassettes, cassettes for the control rods and cassettes and blocks of reflectors in the form of a hexagonal lattice.

Description of the design, composition and calculation results of these critical assemblies are presented in [8]. Loading cartograms of critical assemblies planned for the study are shown in the Fig.11.

4 CONCLUSIONS

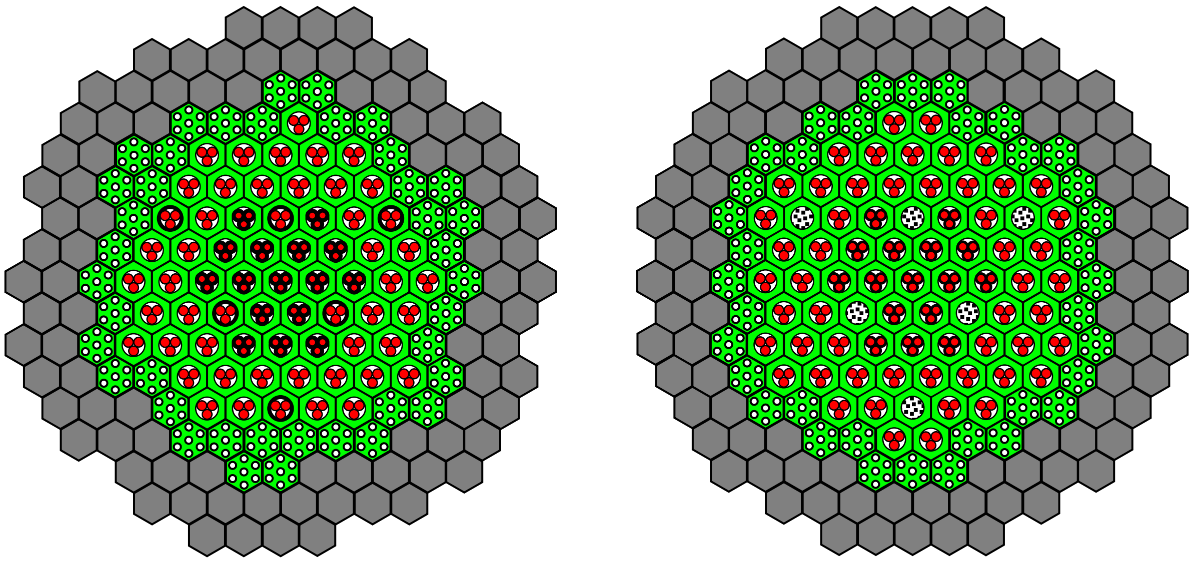
As a result of the study of the properties and characteristics of fuel based on uranium-zirconium carbonitride under irradiation conditions, a methodical reactor experiment has been carried out in the SM-3 reactor. Tests of the irradiation device "ID CNT" have been carried out in cell No. 11 of the reflector of the SM-3 reactor. The irradiation time of the device has been 23.3 effective days. The average value of the volumetric energy release in the test pellets during the methodical reactor tests has been 516 W/cm^3. The burnup achieved in the test tablets is 0.63 fifa.



— fuel cassette with 19 Type 1 fuel rods — fuel cassette with 19 Type 2 fuel rods

— control rod — beryllium reflector unit — steel reflector unit

Figure. 10 – The loading diagram of the F-20-A (F-20-L, F-20-AL) fast critical assembly



— cassette with control rod

(a)

— cassette with control rod

(b)

— fuel cassette with 3 Type 1 fuel rods

— fuel cassette with 3 Type 1 fuel rods

— zirconium hydride reflector cassette

— steel reflector unit

Figure 11 — Loading diagrams of H-20-1 (a) and H-20-2 (b) critical assemblies

As a result of the material science studies, it has been found that, as a result of the additional sintering of the fuel, the dimensions of the fuel pellets (height and diameter) have decreased during irradiation by approximately 1%, and the density has increased by approximately 3%.

As a result of non-destructive post-reactor studies (gamma-scanning of the experimental capsule), it has been found that the fuel column has no breaks, the length of the fuel column after irradiation have not changed within the measurement error. It has been also shown that the cesium-137 accumulated during the irradiation has completely remained in the fuel

The fuel microstructure is characterized by equiaxed grains. The fuel during irradiation retained a single-phase crystal structure with a face-centered cubic lattice (Fm-3m space group), but there is a slight decrease in the crystal lattice parameter.

Preparations have been made for a reactor experiment aimed at obtaining data on the behavior of uranium-zirconium carbonitride fuel at a burnup of 5% fifa.

Design documentation has been developed for critical assemblies containing fuel assemblies with nuclear fuel based on low-enriched UZrCN and air, aluminum or lead as a matrix material, simulating the physical features of the cores of promising fast reactors cooled with gas and liquid metal coolants. Also, design documentation has been developed for intermediate-neutron critical assemblies with a hydride-zirconium moderator. The elements of these critical assemblies have been made, and preparations are underway for experiments.

A program of experimental work at these critical assemblies was prepared and safety justification was carried out during their implementation.

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