ENGINEERING DESIGN FOR IN-PILE TESTS IN RESEARCH REACTOR AND INVESTIGATIONS IN FAST CRITICAL ASSEMBLY OF U-Zr-C-N LEU FUEL

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ABSTRACT

U-Zr-C-N fuel is a high density, high temperature fuel that has potential for application in different types of reactors, comprising research reactors, including of conversion HEU on LEU fuel. In the past, reactor tests of U-Zr-C-N HEU (96% U-235) fuel have been performed to ~ 0.6% burnup. However, reactor-testing data is still needed at high burnup to confirm the optimal performance of this fuel. As a part of U-Zr-C-N LEU (19.75% U-235) fuel testing activities under RRRFR Program, a research experiment will be performed to ~ 40% burnup in the high-flux SM-3 reactor in Dimitrovgrad, Russia, and "Giacint" and "Kristal" critical facilities in Minsk, Belarus will be used to determine neutronics characteristics of critical and subcritical assemblies, modeling physical features of cores of reactors and ADS systems, cooled gas and liquid-metal coolants. The fuel material is uranium zirconium carbonitride ($U_{0,9}Zr_{0,1}C_{0,5}N_{0,5}$) with ~ 12,5 g/cm^3 density and ~ 11,3 g/cm^3 uranium density. Material being investigated for possible use as a cladding – Nb, Mo, W and stainless steel. This paper describes the design of the experiment that will be performed in the SM-3 reactor and discusses the results of different calculations and basic pre-pile experiments that have been performed to show that the experiment design will meet all objectives. The description of the design and the composition of the critical and subcritical assemblies with U-Zr-C-N fuel, the results of calculation are presented also.

1. Introduction

Among the types of nuclear fuel considered for potential application in different types of reactors a high density, high temperature uranium-zirconium carbonitride U-Zr-C-N compositions are of interest [1]. They are compatible with the structural materials, and their radiation resistance is acceptable, which is stimulating develop and test on such fuel.

As part of this effort, reactor tests using U-Zr-C-N HEU (96% U-235) have been performed to low burnup of 0.6% fifa that show the fuel has optimal irradiation performance characteristics. In general, U-Zr-C-N fuel has better thermo-physical properties than UO_2 fuel. U-Zr-C-N is a high uranium density fuel (that requires relatively low U enrichment) that can be employed at relatively high operating temperatures (≥2500K). The zirconium in the fuel stabilizes the phase composition, and the carbon blocks relatively low temperature dissociation typical for UN. The fuel has a thermal conductivity almost 10 times higher, a strength limit almost 3 times higher, and volumetric swelling almost 3 times lower than UO_2; has high resistance to overheating during accidents (4 times higher than UO_2); has lower fission gas emissions and swelling compared to UN fuel; and, has a relatively smooth transition to a corium dioxide phase during extreme accidents. The main disadvantage for U-Zr-C-N is the limited amount of data on the irradiation performance of the fuel, particularly at high burnups.

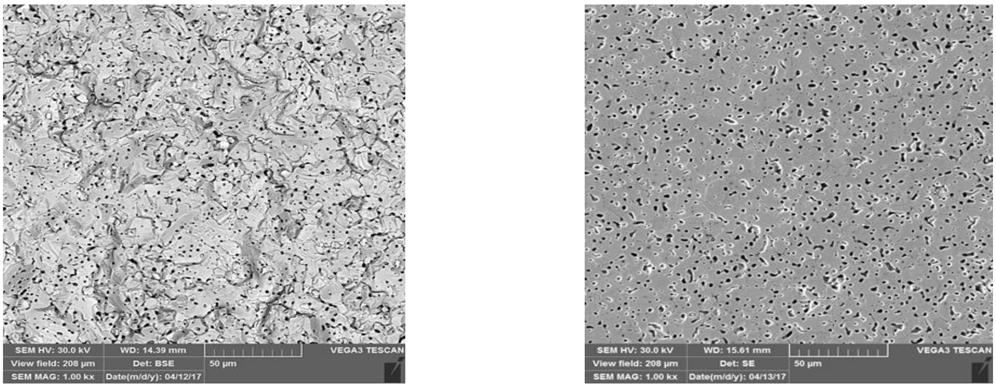
In order to produce the high-burnup data that is needed for demonstrating the optimal characteristics of U-Zr-C-N, a reactor test will be performed up to ~40% burnup (with some U-Zr-C-N pellets being removed after 5 and 15% burnup) in the high-flux SM-3 research reactor located in The State Scientific Center-Research Institute of Atomic Reactors, Dimitrovgrad, Russia. The fuel that will be tested has a composition of $U_{0.9}Zr_{0.1}C_{0.5}N_{0.5}$, a density of 12.5 g/cm^3, an enrichment of 19.73% (uranium-235), and an uranium density of 11.3 g/cm^3. Over 1700 effective days of irradiation will be required to achieve the targeted burnup.

It is planned to use the “Giacint” and “Kristal” critical facilities located in The Joint Institute for Power and Nuclear Research - Sosny of the National Academy of Sciences of Belarus, Minsk, Belarus, to carry experiments at critical assemblies on fast neutrons, simulating physical features of the cores of fast reactors, cooled by gas and liquid-metal coolants.

This paper describes the design of the experiment that will be performed in the SM-3 reactor and discusses the results of different calculations and basic pre-pile experiments that have been performed to show that the experiment design will meet all objectives. The description of the design and the composition of the critical and subcritical assemblies with U-Zr-C-N fuel, the results of calculation are presented also.

2. Engineering design for in-pile tests in research reactor SM-3

The FSUE «SRI SIA «LUCH» has developed the technology for production of carbonitride fuel by the carbothermic recovery method with a reduced content of oxygen impurity (less than 0.1% by weight). The results of structural phase research of the fuel pellets are presented in Fig. 1. The longitudinal sections of the fuel pellet in the original, state characterize the fuel as sufficiently dense and homogeneous without macro- and micro-defects over the entire fracture section and polished section. The spherical pores with a size of 1-1.5 μm are evenly distributed across the pellet section and located in the grain body, predominantly. The pore sizes and their shape in the polished section differ, to some extent, from similar pore sizes and shape in the fracture section due to etching and crystallographic orientation of some grains. The fuel microstructure is homogeneous with polyhedral equiaxial grains of 10-15 μm. The fracture surface characterizes it as mixed of a predominantly transcrystalline fracture. X-ray analysis showed that the fuel pellet material comprises one phase, i.e., solid solution of U,Zr(C,N) with the lattice spacing a=4.895Å. Local X-ray microanalysis demonstrated the absence of impurity elements in the fuel material. (The quantitative data of impurity element on metalloids (C,N) are exaggerated). The fuel pellet micro hardness was HV=880 kg/mm^2 (8.6 GPa). It was established that the (U,Zr)C,N fuel composition was an optimal version of high-temperature fuel by the aggregated thermophysical and operational properties.



(a) (b)

Fig. 1 Microphoto of U-Zr-C-N: a – fracture section; b – polished section

The main problem preventing broad use of this highly promising fuel is related to the fact that its behavior under irradiation has been studied very little. A series of experiments and research has been conducted in order to justify the use of U-Zr-C-N as a new fuel; however, attestation of highly pure U-Zr-C-N in the reactor with a deep burnup level has not been made. Therefore, the basic problem of this work is to study the behavior of carbonitride fuel and its properties after long-term irradiation in the reactor and to identify the radiation component in the changing characteristics of this fuel. The obtained data will make it possible to offer U-Zr-C-N fuel as substitution of the oxide fuel. For justified and predicted use of U-Zr-C-N fuel in research and gas-cooled reactors, it is required to conduct systemic reactor tests, out-of-pile certification of the U-Zr-C-N properties, as well as parallel simulation of reactor thermal processes in electrothermal facilities. Based on the conditions of the planned use of U-Zr-C-N fuel, the parameters of the reactor experiment in the SM-3 reactor have been generated in order to identify the properties and characteristics of the uranium-zirconium carbonitride, such as: temperature in the center of the fuel pellet up to 1600 K limited by the compatibility with the turbo-electric generators; the burnup depth up to 40% fifa ensuring the zone operating capacity without reloading for megawatt reactors up to 10 years, and energy release at 520 W/cm^3 defined by the experiment period not more than 5 years. The tests will be performed in the irradiation device (ID). This article presents the priority version of the ID. The ID scheme (Fig. 2) was chosen given the previous rector tests of U-Zr-C-N fuel. During the experiment it is planned to test a statistically representative batch of standard pellets in the controlled radiation conditions (temperature and neutron flux). The radiation parameters should be controlled throughout all campaigns (one campaign lasting 10 days).

Neutron physics, thermo-physical and strength calculations, as well as series of out-of-pile experiments were made. The neutron physics (NP) testing conditions for experimental capsules in the irradiation device were calculated for the simulator IMCOR_SM [2], built on the precision program MCU-RR [3], using the algorithm for the solution of then neutron transfer equation by the Monte Carlo method. The calculations were made with due account for the state A3, correspondent to the middle and end of the campaign. The calculated energy release values in the fuel pellets and in the structural elements of the irradiation device show that the highest energy release values (519 W/cm^3) at the end of the campaign were recorded in the pellet with enriched fuel, installed in the center of the fuel pile. The NP calculations showed that the energy release in the U-Zr-C-N pellets exceeds the nominal specified energy release values by 17%. In order to maintain the nominal energy release values in the fuel for 5 years, it was suggested to install a hafnium screen around the ID. The neutron physics calculations were used to determine the heat conditions of the irradiation devices during the reactor tests. The calculations were made using the software ANSYS Mechanical [4] in a 2D axially symmetric version, as shown in Fig. 2. The calculations took into account the heat expansion of materials, the irradiation and the contact heat resistance. The energy release in the structural materials is taken according to the neutron physics calculation data. The calculations were made on the orderly grid with a total number of elements of about 35,000. The results of calculations of the temperature field in the ID per ANSYS code are presented in Fig. 3.

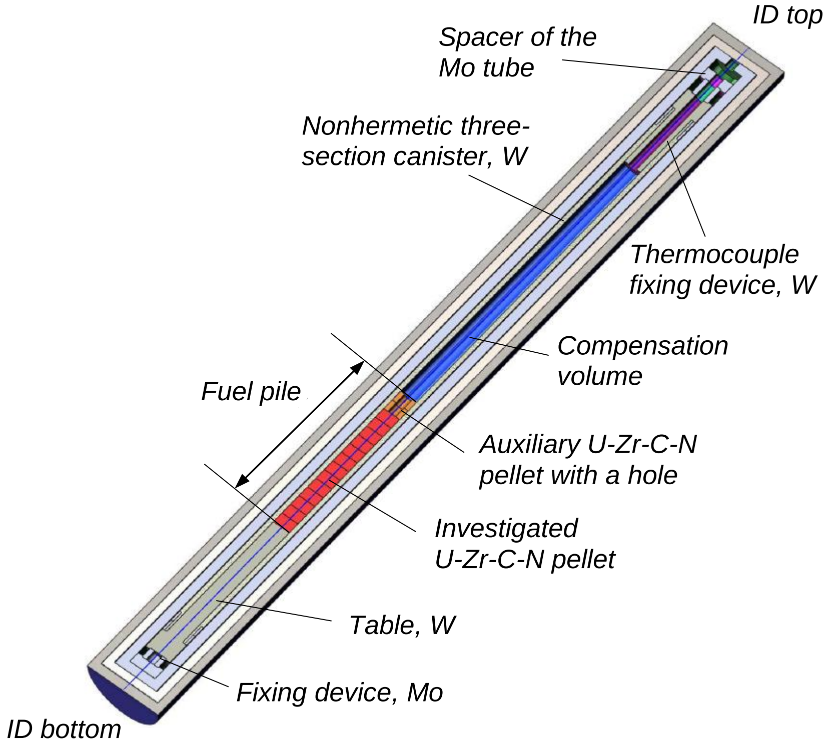


Fig 2. The irradiation device

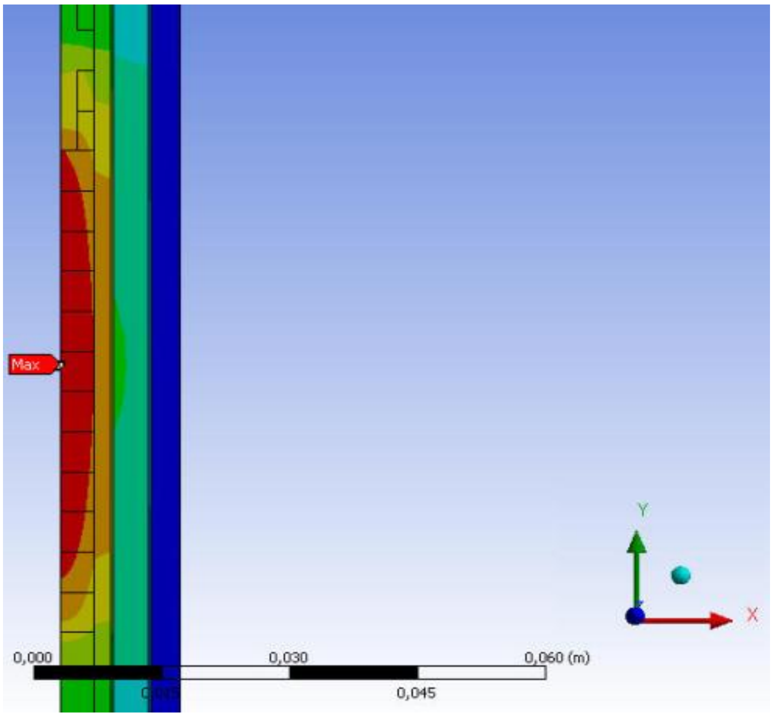
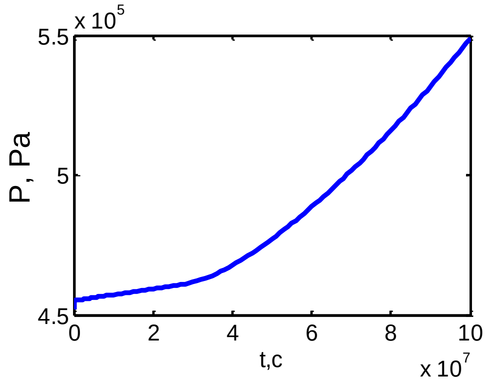


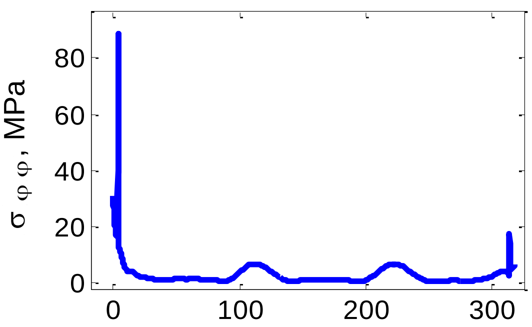
Fig 3. The temperature field of ID with heat release 519 W/cm^3

It follows from the calculations that at these parameters of the reactor experiment, given the thermophysical properties of ID materials, the condition of maintaining the U-Zr-C-N temperature below 1600 K is fulfilled.

The condition for the end of the reactor experiment was the recording of gaseous fission products (GFPs) in the gas stand of the SM-3 reactor; therefore, the strength calculations were made for the Mo shell as the element that preserved tightness of the experimental capsule and prevented the release of the GFPs to the gas stand. It was found that the subshell maximum gas pressure was 5.5 atm (Fig. 4a), which was insignificant for possible breaking of the Mo shell strength. The stress in the Mo shell will be determined only by the inhomogeneous temperature along the length and radius. The equivalent stress values are shown in Fig. 4b.



(a)



(b)

Fig 4. Gas pressure under the cladding versus time (a), equivalent stresses (b) in the molybdenum shell

The calculations results for the Mo shell indicate that the maximum equivalent stresses do not exceed 80 MPa, testifying to tightness of the ID throughout the campaigns. The performed calculation modeling of U-Zr-C-N pellet testing characteristics as part of the ID demonstrated possible burnup of 40% fifa during the projected period of 5 years (1700 effective days of the reactor operation).

Following the mathematical calculations, a number of experiments were performed in order to confirm the correct selection of materials for the shell and evaluate the performance of the ID: 1) the contact compatibility of U-Zr-C-N fuel with the shell refractory material; 2) the thermal strength of the Mo shell; and 3) the contact resistance of the IR simulator set.

The methods used to define the contact resistance of the ID simulator set were based on the known method of direct determination of heat conduction of the coaxial set of bushings for the one-dimensional steady-state radial heat flux. This work used pre-reactor thermal tests for full-scale fuel rods of different nuclear power facilities. These methods were developed by the enterprise FSUE «SRI SIA «LUCH» at the PARAMETER facility [5] using methods of direct or indirect heating by electrical current. The clad fuel rods with a composite pellet-type fuel core efficiently use the gradient heating by means of central ohmic heating. Figure 5 shows a schematic view of the working section. Processing of the experimental results by the radial one-dimensional heat flow demonstrated the following results for the contact resistance: the gap between the molybdenum hood and the protective steel cladding, the original gap of 20 μm, and the thermal resistance in vacuum $(3.25\pm1.0)\cdot10^{-3} m^2K/W$. It should be noted that in the version a) the share of the radiant component of the effective heat conduction at the used temperatures may be up to 40%, according to the calculations. The remaining part refers to the conduction part of the effective heat conduction.

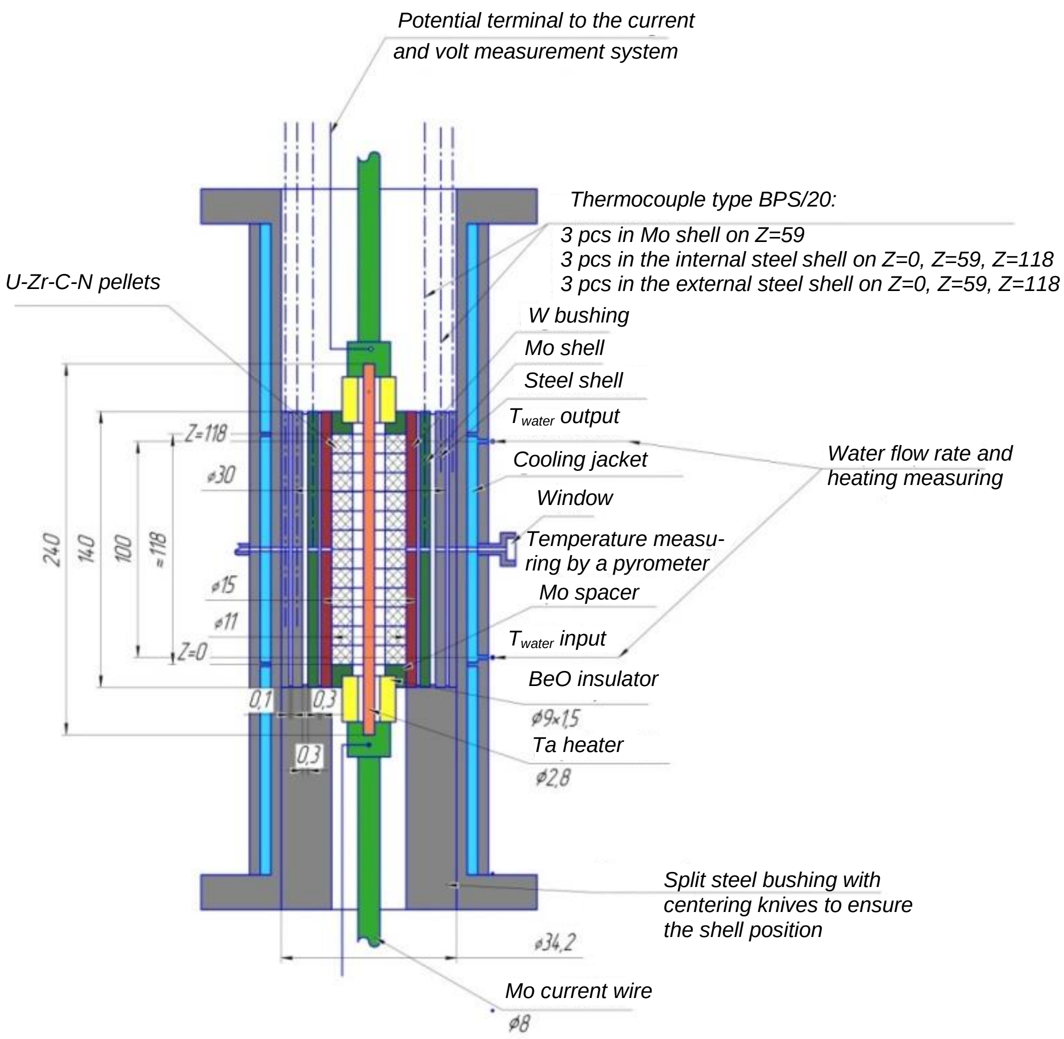


Fig 5. The schematic view of working area with the model capsule and ID elements

It was required to justify the use of the selected refractory materials in the experimental capsule elements and to estimate its performance capacities in conditions of the reactor experiment during 5 years. It is impossible to justify the compatibility of the fuel with the cladding material by using direct experiments at the specified temperature. It is necessary to develop accelerated testing methods making it possible to predict the results for the specific processes of interest for the given resource. Given a rather high level of operation temperatures, it is suggested that tungsten or molybdenum should be selected as the experimental capsule cladding material, at least at the initial work stage, since these materials possess the maximum required range of the needed properties. The criterion for accelerated testing for diffusion compatibility in the first approximation can be the root mean square diffusion mixing of the diffusion atom as the product of the diffusion factor by the time:

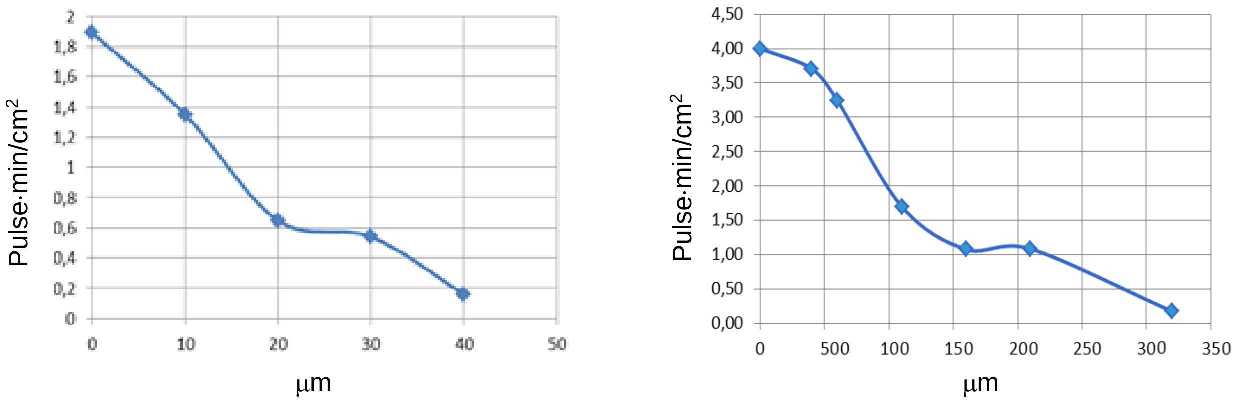
$$x^2 = D \cdot t \tag{1}$$

The temperature dependences of the diffusion factors U in W and Mo from the constant source are as follows:

$$D_W = 0.45 \cdot \exp\,(-399200/RT),\ \text{cm}^2/\text{s} \tag{2}$$

$$D_{Mo} = 0.32 \cdot \exp\,(-341100/RT),\ \text{cm}^2/\text{s} \tag{3}$$

For the total assumed the experimental capsule operation time equaling 30000 hours and the contact area temperature equaling ~1600 K the value (D·t), for example, for Mo, makes $2.57 \cdot 10^{-4} cm^2$. The same root mean square diffusion mixing (D·t) can be implemented over a shorter period of time at higher temperatures by increasing the diffusion factor by the exponential law [6]. The acceleration scale is equal numerically to the diffusion factor ration at the nominal and increased temperatures. For instance, the ratio of the diffusion factor U in Mo at the increased temperature 2073 K to the diffusion factor at 1600 K makes 346. When tungsten is used, the relevant ratios make 936. The tests were made according to methods of high-temperature annealing in a vertical vacuum furnace with mandatory certification of the samples, when the time of testing was 150 hours (calculated from the conditions of the diffusion factor ratio at the nominal and increased temperatures) at 2073 K. The depth of U-Zr-C-N fuel interaction with the spacer element material was determined using radiometric devices. The testing objects were polished W and Mo spacer elements and standard U-Zr-C-N pellets. Figure 6 presents the radiometric results.



(a) (b)

Fig 6. Radiometric results: a – W spacer element; b – Mo spacer element

It follows from the charts that the depth of uranium diffusion to the tungsten spacer element is an order of magnitude lower compared to the molybdenum spacer element. The uranium diffusion to the tungsten is insignificant making 40 µm maximum, and this testing mode corresponds to the reactor experiment at T =1600 K with the time period 90 000 hours (the calculated theoretical depth of diffusion for this time period was 54 µm); hence, W is the priority material contacting U-Zr-C-N.

The experiments for determination and confirmation of the Mo shell strength were made in an induction furnace with the heating/cooling rate 22 K/sec, the testing temperature 1600 K, the number of cycles corresponding to the number of reactor campaigns over 5 years, namely 350, and the inner pressure by nitrogen inside the capsule 10 atm, the inner pressure simulating output of GFPs. The testing object was a full-scale model of a tight Mo shell of the ID. Following the tests, the ampoule was tested to tightness by He pressure 1.0 MPs, with no pressure drop recorded over 10 minutes, signifying that cycling did not damage tightness of the welded joints and the shell. No change of the ampoule geometry was recorded. Neither there were recorded any changes in the welded structure of the joints, cracks, burns-thru, unfilled areas or craters, foreign inclusions, incompletely fused edges, overlapping areas over 0.2 mm, expansions (or narrowing) and uneven height over 0.2 mm.

3. Engineering design for investigations in fast critical assemblies of the critical facilities “Giacint” and “Kristal”

The critical facilities “Giacint” and “Kristal” are used to prepare the experiments on the criticality of multiplying systems simulating physical features of the core of the future fast-neutron reactors with gaseous and liquid-metal coolants. Lattices of fuel cassettes in a matrix from air, aluminium and lead were investigated.

These fast critical assemblies represent a lattice (39 mm pitch) of fuel assemblies with fuel rods based on U-Zr-C-N (19.75% U-235) and with air or lead or aluminum as matrix materials, with a beryllium-steel reflector. The critical assemblies included a core, a side reflector, top and bottom axial reflectors and a control and protection system’s (CPS) rods.

The cores of critical assembly, comprising fuel assemblies, is surrounded by several rows of Be and steel reflector units. These elements of the critical assemblies are placed on the stainless-steel support grid. The neutron detectors are attached on special arms around the critical assemblies.

There are six types the fuel cassettes. The fuel cassette type 1 (type 2) with air as matrix materials is unclad and comprises 7 fuel rods of type 1 (7 fuel rods of type 2) and end pieces (Fig. 7). The fuel rods are placed in a cassette with a 14 mm pitch over the hexagonal grid and fixed by the end pieces. The turn-key size of the design model of the cassette is 38.5 mm; the total length of the design model of the cassette is 947 mm. All upper and lower end pieces of the cassettes are made from stainless steel.

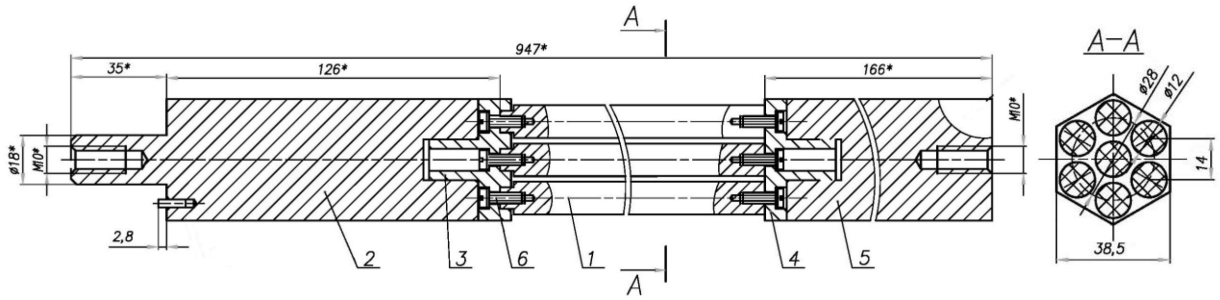


Fig 7. The design model of fuel cassette type 1 (type 2): 1 – fuel rod of type 1 (type 2); 2 – shank; 3 – lower tube plate; 4 – upper tube plate; 5 – head; 6 – screw

The fuel cassette type 3 (type 4) is unclad and comprises 7 fuel rods of type 1 (7 fuel rods of type 2), a lead matrix, and end pieces (Fig. 8). The fuel rods are placed in casing pipes 13x0.3 mm (stainless steel) with a 14 mm pitch over the hexagonal grid. The fuel rods are fixed in the cassette by means of the upper and lower tube plates. The lower end piece comprises a shank (stainless steel), a hexagonal lead prism, and a lower tube plate (stainless steel). The upper end piece of the cassette comprises an upper tube plate (stainless steel), a hexagonal lead prism, and a head (stainless steel). The turn-key size of the design model of the cassette is 38.5 mm; the total length of the design model of the cassette is 947 mm.

The fuel cassette type 5 (type 6) is unclad and comprises 7 fuel rods of type 1 (7 fuel rods of type 2), a aluminum matrix, and end pieces (Fig. 9). The fuel rods are placed in casing pipes 13x0.3 mm (stainless steel) with a 14 mm pitch over the hexagonal grid. The fuel rods are fixed in the cassette by means of the upper and lower tube plates. The lower end piece of the cassette comprises a shank (stainless steel) and a lower tube plate (stainless steel). The upper end piece of the cassette comprises an upper tube plate (stainless steel) and a head (stainless steel). The turn-key size of the design model of the cassette is 38.5 mm; the total length of the design model of the cassette is 947 mm.

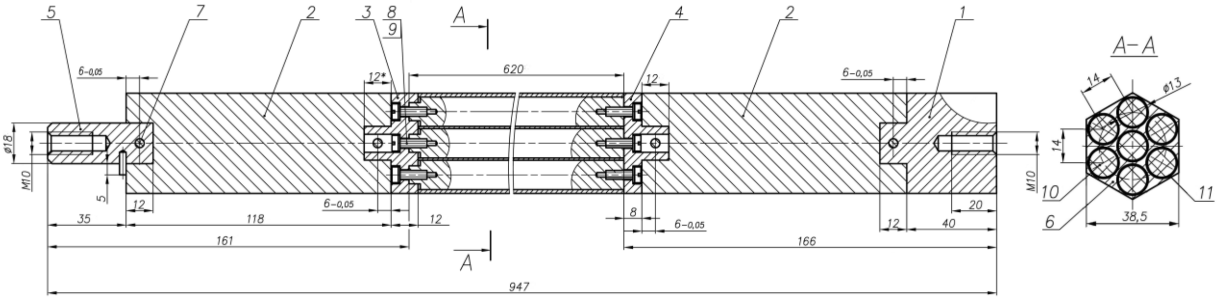


Fig 8. The design model of fuel cassette type 3 (type 4): 1 – head; 2 – lead prism; 3 – lower tube plate; 4 – upper tube plate; 5 – shank; 6 – lead matrix; 7 – pin; 8 – washer; 9 – screw; 10 – fuel rod type 1 (type 2); 11 – casing pipe

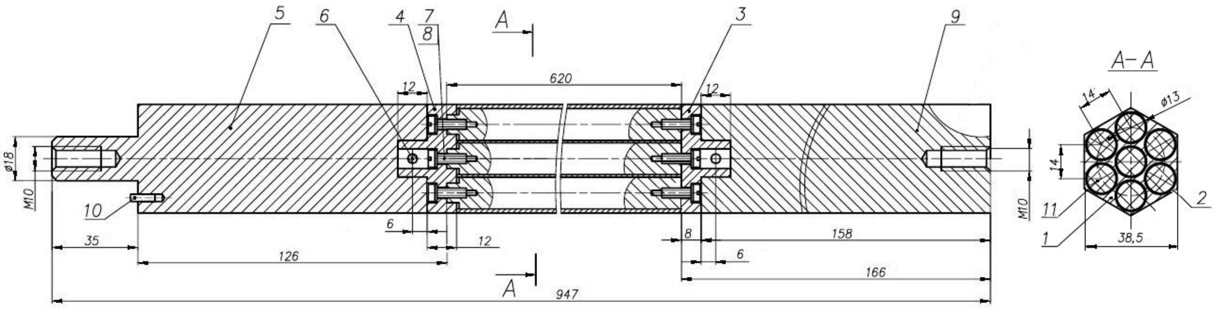


Fig 9. The design model of fuel cassette type 5 (type 6): 1 – aluminum matrix; 2 – casing pipe; 3 – upper tube plate; 4 – lower tube plate; 5 – shank; 6 – pin; 7 – washer; 8 – screw; 9 – head; 10 – pin; 11 – fuel rod type 1 (type 2)

The fuel rod type 1 (type 2) comprises a fuel core, cladding, and end pieces (Fig 10). The fuel rod cladding has the outer diameter 12 mm and the wall thickness 0.6 mm. The fuel rod comprises pellets, 10.75 mm in diameter and 14.7 mm in height, from uranium-zirconium carbonitride $U_{0,9}Zr_{0,1}C_{0,5}N_{0,5}$. The enrichment by U-235 was 19.75%. The gaps between the fuel rod cladding pellets and the fuel rod cladding contain He under ~0,11 MPa. The total core height is 500 mm. The total length of the fuel rod is 620 mm. The material of the fuel rod clad and end pieces (plugs) is stainless steel (fuel rod type 1) or alloy NbZr-1 (fuel rod type 2).

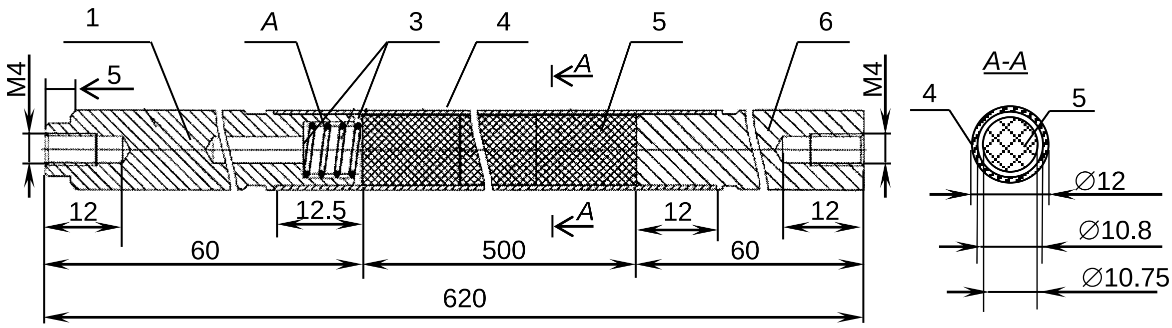


Fig 10. Fuel rod type 1 (type 2): 1 – lower plug; 2 – spring; 3 – gaskets; 4 – cladding; 5 – fuel core; 6 – upper plug

The side reflector of the critical assemblies is several rows of the beryllium and stainless steel reflector units. The beryllium reflector unit represents a hexagonal beryllium prism with turn-key size 38.5 mm and the length 872 mm. To the bottom of the unit is attached to the shank. The head (stainless steel) with the turn-key size 38.5 mm and the length 40 mm is fixed to the upper part of the unit. The total length of the beryllium reflector unit unit is 947 mm, and the shank length is 35 mm. The steel reflector unit represents a hexagonal prism from stainless steel with the turn-key size 38.5 mm. To the bottom of the unit is attached to the shank. The total length of the steel reflector unit is 947 mm, 35 mm shank length.

The experiments are to be performed on three fast critical assemblies. The core and reflector compositions of these critical assemblies are presented in Tab. 1. The calculation results of K_{eff} made by the Monte Carlo method using the MCNP-4C [7] and MCU-PD [8] computation codes are presented in Tab. 2. Figure 11 represent the loading chart of the fast critical assemblies.

The critical assembly	The fuel cassette, pcs						Beryllium reflector unit, pcs	Steel reflector unit, pcs
	type 1	type 2	type 3	type 4	type 5	type 6		
Type 1	210	7	—	—	—	—	504	306
Type 2	—	—	210	7	—	—	504	306
Type 3	—	—	—	—	210	7	504	306

Tab 1: The core and reflector compositions of the fast critical assemblies

The critical assembly	K_{eff} calculation result		β_{eff} calculation result
	MCNP-4C	MCU-PD	MCU-PD
Type 1	1,00494 ± 0,00021	1,00993 ± 0,00030	0,007408
Type 2	1,01672 ± 0,00021	1,01244 ± 0,00027	0,007362
Type 3	1,01412 ± 0,00020	1,01619 ± 0,00037	0,007367

Tab 2: The calculation results of the fast critical assemblies

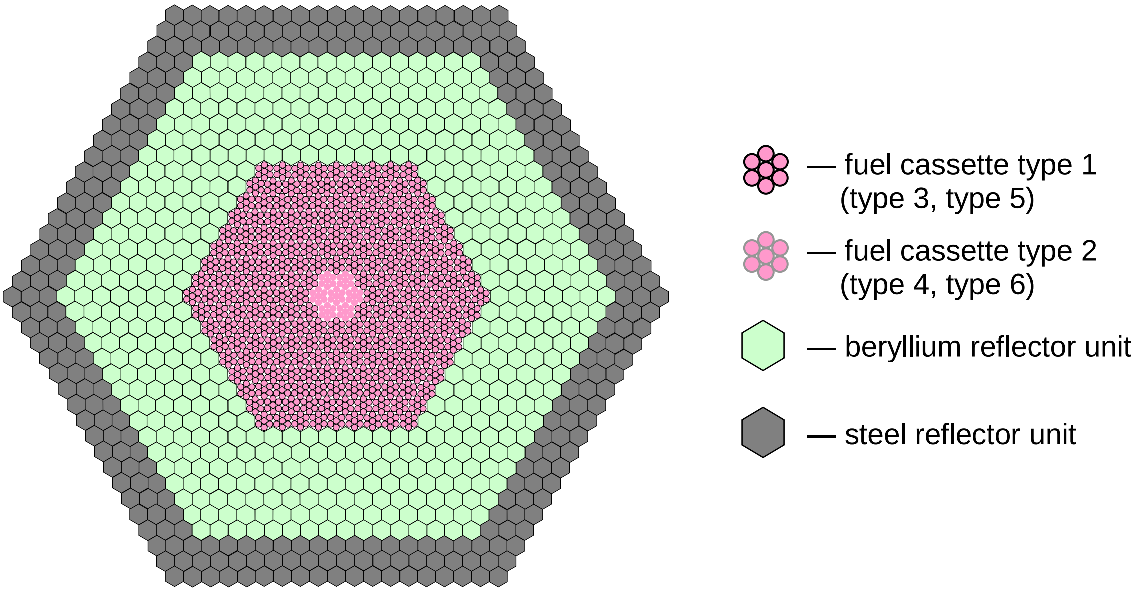


Fig 11. Loading chart of the critical assembly type 1 (type 2, type 3)

4. Conclusions

Nominal calculation studies of neutron physics, thermophysical and thermomechanical parameters of the experimental capsule and the irradiation device were made in order to justify possible reactor testing of U-Zr-C-N fuel reach 40% fifa burnup during an estimated period of up to 1700 effective days. The results of neutron physics, thermophysical and strength calculations show that it is possible to reach the required burnup values in the investigated U-Zr-C-N pellets at the specified reactor experimental conditions. The program of out-of-pile experiments was implemented that allowed confirmation of the structure and materials selected for the irradiation device.

It is planned to use the “Giacint” and “Kristal” critical facilities to carry experiments at critical assemblies on fast neutrons, simulating physical features of the cores of fast reactors, cooled by gas and liquid-metal coolants.

5. References

[1] Alekseev S.V., Zaitsev V.A. Nitride fuel for nuclear energy. Moscow, Technosfera, 2013.
[2] Marikhin, N. Yu., Modernized Software: Simulator of the SM Reactor Core (IMCOR_SM). Description and User Manual. Report of the JSC «SSC RIAR». 2014, O-6264.
[3] Gomin, E. A., MCU-4 Status. 1 Problems of Atomic Science and Engineering. Series: Physics of Nuclear Reactors, 2006, Issue, pp. 6–32.
[4] ANSYS Mechanical User Guide, ANSYS Inc. Southpointe.
[5] Tikhonov, L. V., Kononeko, V. A. et al., Structure and Properties of Metals and Alloys. Reference Book. Mechanical Properties of Metals and Alloys. Kiev, Naukova Dumka Press, 1986.
[6] Ulybyshev, E., Zborovskii, V. G., Kozhakin, A. N., Likhanskii, V. V., Sorokin, A. A., Soldatkin, D., M., and Khoroshilov, A. V., Determination of Crack Resistance of the Nuclear Fuel Tablet in Out-of-Pile Experiments Using Uneven Radial Heating. Enterprise Laboratory. Diagnostics of Materials, No. 9 (80), 2014.
[7] MCNP – A General Monte Carlo N-Particle Transport Code, Version 4C / Ed. J. F. Briesmeister, Report LNL, LA-13709-M, 2000.
[8] Application Description and User’s Manual for MCU-PD program, Report NRC “KURCHATOV INSTITUTE”, №36-10/30-11, Moscow, 2011.