Preparation for reactor tests of uranium-zirconium carbonitride fuel

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Abstract. Uranium-zirconium carbonitride based fuel (CNF) is a consequential modification of UN retaining almost all the advantages of UN. This fuel boasts rather high uranium content and high heat conductivity. Uranium-zirconium carbonitride fuel is safer due to its higher tolerance (inertia) to accident processes development. Thanks to its properties, the CNF is an attractive candidate material for using in reactors of various types. The main drawback of UZr(CN) is insufficient amount of data regarding its performance and behavior under irradiation, especially at high burn-ups. To address this problem, preparations are currently underway to perform a reactor experiment with the goal to study the properties of the CNF after reaching ≈ 7% burn-up. The parameters of the reactor experiment are as follows: cladding temperature not exceeding 800 K, power density not exceeding 750 W/cm³. For the purposes of reactor testing of the CNF pellets at high burn-up, an experimental capsule installed into the irradiating device has been developed. To verify the choice of designs of both the experimental capsule and the irradiating device thermophysical calculations were made and a programme of pre-irradiation experiments was completed. This paper elaborates on the results of the calculations demonstrating that the reactor tests fit the set goals and objectives.

1. Introduction

Uranium carbide, nitride, silicide and sulfides are currently being evaluated as promising nuclear fuels. However, most attention is now paid to uranium nitride which has shown good thermophysical and mechanical properties and is considered a promising uranium ceramics fuel capable to replace uranium dioxide. Within the operational temperature range, nitride fuel boasts a heat conductivity ~ 7 times higher and a density 1.3 higher as compared with the oxide fuel; as well as good compatibility with stainless steel claddings of fuel elements. However, besides its unquestionable advantages, the mononitride fuel is plagued by a significant drawback affecting the choice of its operational temperature conditions inside reactors, namely its low thermochemical stability. The problem is that at a temperature exceeding 1950 K in vacuum or nitrogen-free atmosphere dissociation of the mononitride occurs resulting in a rapid shift of the original composition beyond the field of homogeneity attended by formation of metallic uranium phase. A team of employees of the LUCH FSUE has developed a uranium-zirconium carbonitride based fuel (CNF) with low oxygen content (below 0.1 % mass), which is a consequential UN modification retaining all the advantages of the
mononitride and boasting a better high-temperature stability. It should be noted that any and all properties of the CNF are of stochastic nature, and disperse considerably, that is the stoichiometry, content and nature of impurities, structure of the fuel are all defined by the specific fabrication process. Therefore, the utmost attention should be paid to the fabrication process and the impurities, oxygen in particular, since the oxygen content in the carbonitride fuel compound is the key process factor defining its performance, first of all – swelling, fission gas release and apparently – its compatibility with fuel element claddings. Uranium-zirconium carbonitride fuel with low oxygen content has a number of advantages as compared with the counterparts, namely: its fissile element density is 1.3 higher than that of uranium dioxide, its heat conductivity is 10 times better and operational temperature is 700 K higher; unlike uranium mononitride it has a better thermochemical stability (operational temperature is 1300 K higher) [1,2]. Table 1 presents the comparison of the key characteristics of the fuels.

Table 1. Comparison of the key characteristics of the fuels

| Characteristic                        | UO₂   | UN    | U-Zr-C-N |
|---------------------------------------|-------|-------|----------|
| Theoretical U nuclei density, g/cm³³  | 10.97 | 13.5  | 12.8     |
| Melting point (inert medium), K       | 3100  | 3120  | 3120     |
| Permissible operational temperature, K| 2200  | 1700  | 2900     |
| Heat conductivity, W/mK               | 3.0   | 28    | 32       |
| Oxygen impurity content, % mass       | -     | < 0.15| < 0.1    |

The main obstacle to wide application of this fuel is insufficient knowledge of its behavior and performance under irradiation, especially, at high burn-ups. To solve this problem an experiment is scheduled to be carried out in the SM-3 reactor of the “SSC RIAR” JSC (Russia) to reach a burn-up of ≈ 7 %. The parameters of the reactor experiment are as follows: temperature at the claddings – not exceeding 800 K, duration of the reactor experiment – 3 years; power density – not exceeding 750 W/cm³.

2. The design of the experimental capsule and irradiating device

To perform reactor testing of the UZrCN pellets under burn-up, an experimental capsule (Fig. 1) to be installed into an irradiating device has been developed. In the course of the experiment it is planned to test a statistically representative and relevant batch of regular pellets under controlled irradiation conditions (temperature and neutron flux). The parameters of irradiation are to be monitored throughout all the campaigns (duration of one campaign – 10 days). The experimental capsule (EC), as shown on Fig. 1, is a sealed cylindrical container with a wall thickness of 1.8 mm, with plugs welded from the top and bottom. There is a hole for a thermoelectric transducer installation in the top plug. The pellets are encased in a sealed heavy-duty molybdenum envelope (in designing the envelope, the principle of minimization of welds and their location in the low-temperature zone beyond the fuel stack was followed). The envelope’s thickness was chosen basing on the five-fold margin of strength with maximum gas pressure inside as high as 1.0 MPa. The high margin of strength was defined on the grounds of deterioration of the envelopes mechanical properties in the course of testing. To eliminate the possibility of undesirable contact between the CNF and the molybdenum envelope in the course of testing, a non-sealed 1.0 mm thick tungsten jacket separated into several segments is inserted into the helium gap. Besides its main function, this jacket preserves the geometry of the test object, takes part in alignment of the fuel stack relative to the axis of the experimental capsule and enables bypassing the axial temperature gradients. Using the tungsten jacket also makes it possible to achieve the azimuthal uniformity of the gas-filled gap virtually without impacting the pellets mechanically. A 250 μm gap is ensured between the CNF pellets and the capsule wall, with the
internal space of the capsule filled with pure helium (under a positive pressure ~0.01 MPa). Grooves for thermocouples are made in the molybdenum envelope to ensure continuous monitoring of the temperature. Reactor testing of the experimental capsule is planned to be carried out using a specially designed irradiating device inside the cells of the second reflector row of the SM-3 reactor. The irradiating device (ID) consists of the experimental capsule and a high-temperature strength steel shell (EP 912-VD or stainless steel). To ensure the specified power density it is proposed to use a composite screen consisting of an absorbent (hafnium) and a displacer (steel).

![Diagram of the experimental capsule layout](image)

**Figure 1.** Experimental capsule layout

### 3. Neutronic calculations

To perform neutronic calculations of the fuel pellet testing conditions, simulation of the irradiating device was conducted using IMCOR_SM simulator created on the basis of MCU-RR precision code which implements the algorithm of solution of the neutron transfer equation via Mote-Carlo method. Two states of the core corresponding to the middle and the end of the campaign were used for the calculations. The simulation used the depth of immersion of the corner compensating units equal to 127.5 mm for calculation of the conditions at the middle of the campaign. For calculation of the conditions at the end of the campaign the corner compensating units were fully extracted. At these positions of the compensating units the calculated reactor reactivity lay within 0.1±0.7 %Δk/k). As a result of the neutronic calculations performed, the configuration of the hafnium screen was defined ensuring average power density at about 750 W/cm³ in the fuel pellets in the experimental capsule. The screen is supposed to be made of 38 GFI-1 wires. The diameter of the screen is chosen so that the primary circuit water could uniformly flow over the screen providing its reliable cooling. Table 2 presents the values of neutron fluxes averaged by volume in the radial zones of the primary fuel pellets.

| Table 2. Neutron flux densities in the radial zones of the fuel pellets |
4. Thermophysical calculations
Thermophysical calculations of the EC and ID were conducted in order to select the width of the gap between the experimental capsule and the irradiating device so that the temperature at the molybdenum envelope wouldn’t exceed 800 K throughout the entire duration of the tests. For this case, power densities in the fuel and structural materials were set up basing on the results of the neutronic calculations.

The calculations were carried out using ANSYS Mechanical software complex for finite element analysis in an axial-symmetrical set-up, taking into account axial heat fluxes and heat exchange by radiation between the ID structural elements. As the boundary conditions, convective heat exchange between the coolant and the Mo envelope was set up, with adiabatic conditions at the butt-ends of the analyzed area. The calculations took into account thermal expansion of materials, radiation and contract thermal resistance. Power density in the fuel pellets was taken into account according to Table 2. The calculations were carried out for the ordered mesh with the total quantity of elements at about 35,000.

Widths of the gaps between the ID elements were varying and assumed to be as follows:
- the calculations assumed the gap between the fuel stack and the protective tungsten jacket as equal to 10 μm;
- the gap between the protective tungsten jacket and the molybdenum envelope - 250 μm;
- between the molybdenum envelope and the steel ampoule - 200 μm.

Results of the calculations of temperature of the fuel and the structural elements of the ID are presented on Fig. 2 to 4.

| Neutron flux density \( \cdot 10^{13} \text{ n/(s\cdot m}^2) \) | over 0.1 MeV | from 0.1 MeV to 1 keV | from 1 keV to 0.5 eV | below 0.5 eV |
|---|---|---|---|---|
| 7.08 | 3.17 | 2.44 | 8.11 |

Figure 2. Radial temperature profile in the central plane of the core
As follows from the calculations, at the preset parameters of the reactor experiment taking into account thermophysical properties of the materials of the EC and ID, the condition of the temperature of the molybdenum envelope staying below 800 K is satisfied. The calculations were carried out at axial-symmetrical setup conditions, i.e. ensuring the ideal conditions of the EC assembly. In real-life conditions failure to assure symmetry is possible; therefore the following EC layout where the tungsten jacket comes into contact with the molybdenum envelope has been studied. For the purposes
of analyzing the effect of eccentricity, the gap between the steel protective jacket and the steel shell was not taken into account.

The results of the calculation are presented on Fig. 5. As follows from the calculations, at the preset parameters of the reactor experiment taking into account thermophysical properties of the EC materials, the condition of the temperature of the molybdenum envelope staying below 800 K is satisfied.

Figure 5. EC temperature pattern in case of eccentricity of the fuel stack

Basing on the afore-stated temperature calculations a conclusion can be made that eccentricity of the fuel stack results in the fuel temperature reduction of at least 100 K. Maximum overheating of the molybdenum envelope will not exceed 90 K.

5. Conclusions

At present, the layout of the experimental capsule has been chosen to perform the reactor experiment to acquire the data on behavior and performance of the uranium-zirconium carbonitride-based fuel under irradiation. The analytical studies of neutronic, thermophysical and thermomechanical parameters of the experimental capsule and the irradiating device have been carried out in order to substantiate the possibility of reactor testing of the CNF fuel to achieve 7% burn-up using the chosen layout of the experimental capsule.

The thermophysical and mechanical calculations performed show that the temperatures keep below the maximum allowable values. The results of analysis of the model with the off-center fuel stack demonstrate preservation of the air-tightness of the Mo envelope and reduction of the fuel temperature. The proposed layout is capable to ensure implementation of the reactor experiment.

References

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[2] Bakhin A.N., Vishnevsky V.Yu., Tukhvatulin Sh.T., Galev I.E., Kotov A. Yu. 2019 Studies of Properties of Uranium-Zirconium Carbonitride: Preparation for Reactor Experiment J. Natural and Technical Sciences. (Moscow: Sputnik plus) vol 3 pp 92-96.