A High Density Uranium Zirconium Carbonitride LEU Fuel for Application in Fast Reactors

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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, including fast reactors. In the past, reactor tests using U-Zr-C-N fuel have been performed to low burnup. However, reactor-testing data is still needed at high burnup to confirm the optimal performance of this fuel type. The SM-3 reactor, which is a high-flux reactor located in Dmitrovgrad, Russia, will be used to test a U-Zr-C-N ($U_{0.9}Zr_{0.1}C_{0.5}N_{0.5}$) fuel to ~40% burnup. The fuel will then be examined to determine its performance during irradiation. The fuel that will be tested has a density of 12.5 g/cm³ and an enrichment of 19.73% (uranium-235), and the uranium density of this fuel material is 11.3 g/cm³. Over 1200 effective days of irradiation will be performed in the SM-3 reactor and discusses the results of different calculations that have been performed to show that the experiment design will meet all objectives.

Key Words: nuclear fuel, carbo-nitride fuel, fast reactors, reactor testing.

1. Introduction

For many years, Russian researchers have developed and tested a high density, high temperature U-Zr-C-N fuel (CNF) for potential application in different types of reactors, including fast reactors. As part of this effort, reactor tests have been performed to low burnup that show the fuel has optimal irradiation performance characteristics. In general, U-Zr-C-N fuel has better thermo-physical properties than uranium-dioxide fuel. [1] [2] CNF 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 uranium nitride. 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 UO₂; has high resistance to overheating during accidents (4 times higher than UO₂); 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 CNF is the limited amount of data on the irradiation performance of the fuel, particularly at high burnups. The optimal characteristics of CNF make it an attractive candidate for use in different types of reactors, including fast and high temperature gas-cooled reactors.

In order to produce the high-burnup data that is needed for demonstrating the optimal characteristics of CNF, a reactor test will be performed up to ~40% burnup (with some CNF pellets being removed after 5 and 15% burnup) in the high-flux SM-3 reactor located in Dmitrovgrad, 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/cm³, an enrichment of 19.73% (uranium-235), and an uranium density of 11.3 g/cm³. Over 1200 effective days of irradiation will be required to achieve the targeted burnup. This paper will describe the design of the experiment and will discuss the results of performed calculations that show a planned reactor test in the SM-3 reactor will meet all the testing objectives.

2. Experiment Design

Experimental capsules have been designed to allow for reactor testing of CNF pellets to 5%, 15%, and 40% burnup contained in an irradiation device. An experimental capsule is a leak-tight cylindrical canister with 2-mm-thick walls that is welded at the top and bottom with plugs. The capsule will be made of W or possibly a W alloy (e.g., W-4Ta). There will be a 300 μ m gap between the CNF pellet and the capsule wall, and the capsule will be filled with pure He (with an excess pressure of ~0.01 MPa). Each CNF pellet will be ~100 mm long and four pellets will be contained in a capsule.

The irradiation device, as shown in FIG. 1, consists of three experimental capsules Mo shells with 4-mm-thick walls contained in protective heat resistant steel (EP 912-VD or stainless steel X12H10T). This experimental assembly is held in X12H10T steel. The Mo shell has grooves for six thermocouples. The steel casing will be exposed to water at a temperature between 50 and 70°C during the irradiation. The irradiation device also has an external neutron absorbing Hf screen to equalize the energy released during the irradiation test. It is planned to perform the reactor tests of the experimental capsules in the designed irradiation devices in positions 10 and 11 of the second reflector row of the reactor SM-3 (FIG. 2).



FIG. 1. Schematic diagram of the irradiation device.



FIG. 2. Diagram showing a cross-section of the SM-3 reactor core. (1 - central beryllium unit; 2 - beryllium inserts; 3 - beryllium units of the reflector; 4 - central compensating unit; 5 channel and its size; 61 - Cells of the core with a fuel assembly; 41 - compensating unit, AP-1 actuator; - actuator A3 in the beryllium insert)

3. Supporting Calculations

The neutron physics conditions for the experiments were calculated using the MCU-RR computer program, and the geometric parameters that were employed are listed in TABLE 1. The program implemented an algorithm for solution of the neutron transfer equation by the Monte Carlo method. The goal of the neutron physics calculations was to determine geometric characteristics of the absorbing hafnium screen, ensuring maximum energy release in CNF fuel (up to 500 W/cm³) in the irradiation device with a heat-resistant steel capsule. The calculations considered the conditions of the core, correspondent to the midpoint and the end of the campaign. The neutron physics calculation results were used to determine the outer diameter of the hafnium screen equaling 59.5 mm, and the wall thickness equaling 2.5 mm. The screen dimensions were selected to ensure heat release with the first circuit washed with water in a uniform manner.

Designation	Beginning Campaign		End of Campaign	
	Scheme 1	Scheme 2	Scheme 1	Scheme 2
Gap between CNF pellet and cladding (microns)	300	300	150	150
Dimensions of W cladding (mm)	12.6 x 2.0	12.6 x 2.0	12.6 x 2.0	12.6 x 2.0
Gap between cladding and cover (microns)	100	100	100	100
Dimensions of Mo cover (mm)	22.0 x 4.6	20.6 x 4.0	22.0 x 4.6	20.6 x 4.0
Gap between Mo cover and protective case (microns)	500 / 300	100	500 / 300	100
Dimensions of protective case (mm)	34.0 x 5.5 /33.6 x 5.5	28.0 x 3.5 /28.1 x 3.5	34.0 x 5.5 /33.6 x 5.5	28.0 x 3.5 /28.1 x 3.5
Gap between protective case and steel case (microns)	500 / 300	300 / 350	500 / 300	300 / 350
Dimensions of Irradiator Unit steel case (mm)	42.0 x 3.5	35.6 x 3.5	42.0 x 3.5	35.6 x 3.5
Thickness of experimental capsule (mm)	5	5	5	5
Height of experimental capsule (mm)	100	100	100	100
Height of Irradiator Unit (mm)	318	318	318	318

TABLE I: GEOMETRIC PARAMETERS OF THE EXPERIMENTAL CAPSULE AND IRRADIATION DEVICE.

Based on the calculated values of the neutron flux density in the experimental capsules for the SM-3 reactor operating at 90 MW, the duration of the tests was estimated. To reach 40% U-235 burnup, around 25 or 26 SM-3 campaigns are needed (more than 1200 days). Based on the calculated value of energy release for the CNF pellets, it was shown that the target energy release of 500 W/cm³ could be met by slightly increasing the thickness of the Hf screen.

During irradiation of the CNF, the thermal physical properties of the gaseous medium in the gap between the fuel and the ampoule cladding will change. The egress of volatile fission products from the fuel (mainly, Xe^{133} and Kr^{89}) results in the formation of a gaseous mixture *He*-*Xe-Kr* in the gap between the fuel and the cladding, whose thermal physical properties deteriorate compared to the original properties of helium specified during the design of thermal physics calculations. A mixture of He-Ne, simulating the mixture of gaseous fission products is fed to the original gas gaps in order to maintain the CNF and ampoule temperature at a quasi-stationary level during the irradiation. Nitrogen released due to dissociation of CNF at elevated temperatures is also considered.

To aid in the selection of the optimum conditions for testing of the CNF in the reactor, it was necessary to predict thermal physical properties of gaseous media in the irradiation device

depending on the content of individual components and the temperature. It was determined that during the irradiation the heat conductivity coefficient of the mixture He-Xe-Kr varies in a range from 0.10 W/(m K) to 0.46 W/(m K) with the content up to 0.6 relative fractions of Xe-Kr and the temperature 1500 K. The broad range relationship of the fission products changes the heat conductivity coefficient of the mixture He-Xe-Kr insignificantly.

The stable heat conditions of the experiment capsules and irradiation devices are ensured by varying the compositions of the gas media used to fill the ampoule and capsule gaps. The predicted temperatures for the CNF pellets and claddings were used to determine the gas compositions in the gaps. These temperature calculations were performed using the codes ANSYS, COMSOL, and PARAM-TG. The calculations were made with iterations, taking into account changes in the model geometry due to heat expansion, given the dependence of thermal physical properties of solids, liquids and gases on temperature. The thermal condition of the fuel section was simulated, taking into consideration variations of the CNF fuel geometry and fission product gas egress. The swelling of CNF pellets and the composition of gaseous media are taken as constant for the calculations. The calculations were made for two core conditions, correspondent to the beginning and end of the campaign, reaching 40% burnup of fissionable atoms. The calculations were made for three arrangements of the CNF pellets and spacers.

At the beginning of life, a 60%He-40% Ne mixture is used for the capsule that is removed after 5% burnup. For the capsule removed at 15% burnup, a 90%He-10%Ne mixture is used. For the capsule irradiated to 40% burnup, a 100%He gas composition is used. For the 40% burnup case, estimations show the final composition of gaseous mixture under the experimental capsule cladding is 60% (Xe+Kr) - 40% He (mole). It was assumed that the fission product gases were fully egressed from the CNF pellets. Given the mechanisms of gaseous fission product migration and diffusion from the CNF, it is predicted that the share of stable Xe and Kr isotopes in the gaseous mixture will decrease to ~ 20% (mole).

4. Conclusions

This paper has described the design of an experiment using CNF pellets that will be performed in the SM-3 reactor up to a burnup of 40%. Calculations have been performed that indicate the irradiation experiment will be accomplished successfully, and all testing goals will be met. Initial gas compositions have been identified for use in technological gaps that will maintain target temperatures, and the final gas compositions after different levels of irradiation have been determined.

5. References

- [1] Alekseyev S.V., Zaitsev V.A. Nitride fuel for nuclear energy. Moscow: Technosfera, 2013. P. 218-223, 239.
- [2] Andreev D.V., Gayduchenko A.B., Domantovskiy A.G., Nevzorov V.N., Remiz E.M. "The Effect of Irradiation on the Structure of Carbonitride Fuel" in: Problems of Atomic Science and Engineering. Series: Physics of Nuclear Reactors. 2013, Issue 1. P.36-43.