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ON THE DEVELOPMENT OF TOLERANT FUEL — A NEW GENERATION FUEL

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Abstract

The concept of tolerant fuel is considered as applied to water-cooled power reactors. The concept is based on eliminating the steam-zirconium reaction. For this, two work areas, i.e., using the physical and the thermodynamic barriers, were considered. Physical barrier presupposes exclusion of contact between water and zirconium, and the thermodynamic barrier (the most radical method) envisages replacement of the alloy containing zirconium with other materials inert to water when exposed to high temperature in the reactor core (~ 1200 °C). Consequences of the most devastating accidents at the nuclear power plants in the world were discussed: Three Mile Island, Chernobyl and Fukushima. The latest accident in Japan brought to the fore the concept of tolerant nuclear fuel, i.e., being resistant to accidents. Work orientation in creating the tolerant fuel is indicated. Main attention is paid to materials and technologies applied to tolerant fuel. General requirements to safety analysis of the reactor facility fuel system currently developed in the Russian Federation and abroad, as well as current safety criteria for fuel elements, under design-based accidents are presented. Procedure for calculating justification of the safety criteria fulfillment for fuel elements under design-basis accidents is briefly considered. Main characteristics of the new generation materials under development for reactor cores as applied to tolerant fuel are presented. Based on comparing the proposed materials as the tolerant fuel for the fuel element claddings, composite materials based on the heat-resistant SiC/SiC ceramic system could be recommended, and as far as fuel materials are concerned — materials with increased density, uranium capacity and thermal conductivity values, i.e., nitride fuel and fuel made of uranium silicide

Keywords

Reactor core safety, fuel element cladding operation, zirconium alloys, silicon carbide, fragility, composite, tolerant fuel

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Introduction. Reactor core operational safety in water-cooled thermal reactors is an important goal of their development and is associated with safety operation of the fuel elements cladding made of zirconium alloys.

Three major accidents at nuclear power plants were registered in the history of nuclear power engineering: Three Mile Island (1979), Chernobyl (1986) and Fukushima (2011). Each accident occurred for its own reasons, and the reactors at these nuclear power plants were of different design.

These disasters triggered severe industry crises and brought about serious changes. The accident at the Three Mile Island nuclear power plant stimulated introduction of new safety standards, creation of an organization for their development and monitoring of their observance.

Accidents at the Chernobyl and Fukushima NPPs showed particular hazard of the vapor-zirconium reaction appearing, when temperature of the fuel elements cladding rises after losses of coolant and bursts of reactivity. Accident at the Fukushima NPP brought to the fore the concept of tolerant nuclear fuel, i.e., the accident tolerant fuel.

These accidents started in different ways, but in all cases the steam-zirconium reaction became the main destructive factor.

The Accident Tolerant Fuel (ATF) term originated precisely after the accident at the Fukushima NPP. As defined by the IAEA, it is the fuel that withstands accidents accompanied by loss of coolant.

Tolerant fuel concept. The tolerant fuel concept is based on elimination of the steam-zirconium reaction. For this, physical and thermodynamic barriers could be applied.

Physical barrier excludes any contact between water and zirconium. One of the options for such a barrier is deposition of protective coatings on the fuel element cladding; various chromium-containing coatings are considered for this purpose.

Another radical way is replacing the alloy that contains zirconium with other materials being inert to water at high core temperatures.

Thermodynamic barrier excludes possibility of a steam-zirconium reaction due to the fact that temperature of 900 °C, at which the reaction starts, will not be reached even in the emergency situations. This could be achieved by raising the fuel pellet thermal conductivity. In case of the uranium dioxide, temperature on the fuel element cladding surface is 300–400 °C, and inside the pellet it is ~ 1500 °C. The gradient is very high, and, as a result, stored heat appears that could initiate reaction of steam with zirconium.

Dense fuel with a higher thermal conductivity does not provide such a gradient; the temperature difference is not very significant. Dense fuel also has another advantage, i.e., the increased uranium content. Due to this, the fuel campaign could hypothetically be increased, and performance of the core could be improved. And with zirconium coated claddings or claddings made of other materials, the increased uranium content could compensate for the loss of reactivity.

Of course, the simplest and fastest of all options for the fuel element cladding to implement is application of the protective coatings on the fuel element cladding, since changing the fuel assembly (FA) design is not required, and interference in the production technology is minimal. Although in the latter case, many technological problems appear [1].

At present, serious attention is paid to materials and technologies as applied to tolerant fuel, namely, to coating on claddings, thin-walled high-strength steel claddings, fuel with increased thermal conductivity (UN and U_3Si_2 — uranium nitride and silicide), ceramic cladding and fuel element cladding made of composite material based on carbide silicon (SiC) [2–7].

Thermal neutron capture cross section in silicon carbide claddings is 25 % lower than in claddings made of zirconium alloys. Silicon carbide is corrosion resistant in water at elevated temperatures [8–10].

Experimental samples of silicon carbide pipes, i.e., fuel element cladding based on a SiC matrix, obtained by various following methods [1, 11] are used as objects of the study:

– isostatic pressing and subsequent high-temperature annealing;

– fiber frame deposition or impregnation with silicon carbide;

– silicon carbide deposition from the gas phase on a monolithic frame.

Disadvantage of the ceramic material (matrix), including that based on silicon carbide, is its fragility. Matrix fragility could be eliminated by increasing its impact strength, tensile strength, bending strength and cyclic loading strength and thermal shock resistance, i.e., by creating ceramic composites.

This concept of tolerant fuel determines development of new fuel and structural materials for operation, including those used in new generation reactors with increased parameters in temperature and radiation dose.

Requirements to fuel safety analysis. LOCA (Loss of Coolant Accident) is one of the most dangerous accidents in reactors Such an accident occurs, when the coolant leaves the core; as a result, the fuel element cladding is first heated to high temperatures (1100–1200 °C) and oxidizes in steam, and then is drastically cooled by water due to the emergency cooling system operation. After such an impact, the cladding shells embrittle (lose their plasticity), which complicates the core unloading after an accident, transportation and storage of fuel assemblies.

The main requirements for fuel elements in the LOCA design-basis accidents include ability to cool fuel elements, including those with modified cladding geometry, without destroying them (ability to withstand thermal shocks) and possibility to ensure the reactor core disassembly after an accident (absence of cladding fragmentation).

Guaranteed exclusion of undesirable fuel damage due to possible erroneous actions of the operator and violations of the technological regulations requirements is ensured by introduction of the reactor core automatic protection against the exceeding linear power. The WWER-1000 design provides for the reactor protection according to the local parameters: linear power and crisis heat transfer margin. In addition, local core protection system is envisaged based on the readings of neutron detectors installed behind the reactor vessel.

General safety requirements for the reactor plant fuel system are set in Russian and foreign regulatory documents, for example, OPB-88/97. NP-001-97 (PNAE G-01-011-97); PBYA RU AS–89 (PNAE-G-1-024-90); NUCLEAR ENERGY AGENCY of the OECD; Fuel Safety Criteria Technical Review; CSNI/R (99) 25; OECD, Paris (2001); IAEA-TECDOC-1381 and others. Reactor operation safety criteria are as follows:

– fuel system should not be damaged in all design modes, except for emergency;

– in emergency modes, the fuel system should not be damaged to the extent preventing insertion of the control rods;

– heat abstraction from the fuel system to the final receiver should always be ensured;

– for the radiation situation analysis, conservative estimate of the depressurized fuel elements number should be determined.

In case of a design-basis accident with the initiating event of rupture of the Du 850 (WWER-1000) main circulation pipeline at the reactor inlet, safety criteria provided in the Table should be fulfilled, as they determine the maximum damage design limit for fuel elements developed by the authors of [12] and included in the current regulatory document PBYA RU AS-89.

Recommendations on the procedure of calculating substantiation for safety criteria fulfillment in fuel elements exposed to design-basis accidents. Linear loads on fuel elements, which are determined in the process of safety analysis, are used in calculating fuel amount in case of an accident. These calculations are also required to assess the fuel element state during the accident ramification, in determining fuel and cladding parameters, and make it possible to establish thermomechanical and corrosive fuel element behavior exposed to deteriorating heat removal in the core and rapid alteration in power [12].

Safety criteria in force for fuel elements in design-basis accidents

PBYA RU AS-80 statement	Criteria introduction objective
Fuel element maximum damage design limit corresponds to not exceeding the following limiting parameters: – fuel element cladding temperature — not more than 1200 °C; – cladding oxidation local depth — not more than 18 % of the initial wall thickness; – reacted zirconium proportion — not more than 1 % of its mass in the fuel elements cladding	Absence of the self-sustaining steam-zirconium reaction is necessary to ensure the core cooling. Limiting cladding embrittlement is necessary to prevent fuel elements fragmentation exposed to thermal shock, cooling water supplied to the core (T = 60 °C), which would ensure possibility of unloading the core fuel assemblies and subsequent transportation thereof. Limiting the amount of hydrogen generated in the vapor-zirconium reaction is necessary to prevent formation of the explosive mixture
Technical design should demonstrate that in design-basis accidents with rapid increase in reactivity, specific threshold energy of the fuel elements destruction at each campaign moment is not increasing and fuel melting is excluded. Enthalpy averaged over the fuel cross-section in a fuel element — no more than 230 cal/g (963 kJ/kg)	Absence of interaction between molten fuel and cladding is necessary to maintain the core cooled geometry and the possibility of fuel assemblies unloading. Absence of fuel elements fragmentation under rapid energy release in an accident with increasing reactivity is necessary to maintain the fuel assemblies' geometry and the possibility of their unloading

Fuel element state evaluation under the accident development, determination of the fuel and cladding parameters are carried out using the calculated fuel codes, which make it possible to establish thermomechanical and corrosive behavior of the fuel element exposed to deteriorating heat removal in the core and rapid alteration in power.

Procedure of computational analyzes to substantiate fulfillment of the fuel elements safety criteria under the design-basis accidents includes the following [12]:

– analysis of the neutron-physical characteristics (NPC) of the fuel loading operation developed using the neutron-physical codes, core division into groups of fuel elements by power and burnup;

– thermophysical calculations of normal operation according to the START-3A code based on the NPC loads, data generation on the thermophysical characteristics of fuel elements of various groups for thermohydraulic calculations of accidents;

– thermohydraulic calculations of accidents accompanied by LOCA and rapid increase in the reactivity-initiated accident (RIA) using the integral codes, generation of initial data on the fuel elements temperature scenarios with different power and burnup;

– calculations of the fuel elements thermomechanical behavior in an accident according to the RAPTA-5A fuel code, determination of criterion thermomechanical and corrosion characteristics, depressurization forecast for fuel elements of each group in terms of power and burnup;

– based on numerical distribution of fuel elements into groups with different power and burnup determination of the oxidized zirconium fraction in the core and of the number of depressurized fuel elements with different burnup.

Fuel development and licensing for improved and economical fuel cycles is carried out on the basis of experimental data, operating experience and detailed design studies [13]. Verification codes certified by Gosatomnadzor of the Russian Federation (START-3A, RAPTA-5A) are only used. Acceptance criteria are formulated with sufficient level of conservatism, and the safety factors values correspond to the established normative values. New fuel creation is impossible without improving technology of its automated production, which contributes to ensuring high operational reliability of domestic nuclear fuel corresponding to the world level.

Key characteristics of materials for reactor cores. Main characteristics of such materials include [14, 15]:

– thermal, mechanical and chemical properties;

– lack of restrictions on burnup and fuel cycle length;

– provision of reserves according to operating parameters (power distribution, peak factors, safety margins, etc.);

– preservation of reserves in terms of reactivity coefficients and controlled physical parameters;

– reloading, transportation and storage (isotopic composition, doses, handling);

– compatibility with the core existing infrastructure.

Requirements to the cladding materials. Ensuring reliability of fuel elements exposed to design-basis and beyond design-basis accidents requires introduction of claddings with the following properties:

– high melting point;

– resistance to high-temperature corrosion of up to 1500 °C;

– slow reaction kinetics with steam, high activation energy of the oxidation process;

– sufficient strength and creep resistance at 1200–1500 °C;

– reduced cladding deformation to ensure the core cooling; sufficient ductility to prevent the fuel elements fragmentation;

– compatibility with the coolant;

– reliability under normal operating conditions;

– acceptable neutron absorption.

JSC VNIINM materials and technologies in relation to the tolerant fuel. With regard to tolerant fuel, the following materials [1, 14, 15] are being developed: cladding coatings; thin-walled high-strength steel claddings; fuel with increased thermal conductivity retaining fission products; ceramic claddings.

Chrome coatings (high-speed ion-plasma magnetron sputtering — HIPMS)

Advantages:

– the rate of chromium-plated fuel elements oxidation at temperatures of 1100–1200 °C is an order of magnitude lower than the rate of the zirconium pipes oxidation. After autoclave experiments on oxidation in water vapor at the pressure of 18 MPa and temperature of 1100–1200 °C, mechanical properties of the claddings are retained (no hydrogen embrittlement) in contrast to the zirconium claddings;

– introducing into production requires minimal changes in the existing design and production of the fuel elements.

Disadvantages:

– experimental data on the coating state before and after irradiation is missing;

– thermal neutron capture cross section by the chromium isotopes (3.05 barn) is 15 times greater than that of the zirconium isotopes (0.185 barn);

– production of the active short-lived isotope ^{51}Cr ($T_{1/2}$ = 28 days) and alteration in chemical composition as a result of the vanadium accumulation.

Coatings using the corrosion-resistant alloys (Al–Ti–Cr, Al–Ti–Cr–N) (HIPMS)

Advantages:

– high corrosion resistance.

Disadvantages:

– lack of data on strength, thermophysical parameters and corrosion resistance, as well as on these alloys' behavior under irradiation;

– in alloys containing nitrogen, ^{14}C would be accumulated under the action of neutron irradiation, which could entail problems associated with storage and processing of spent fuel elements;

– cross sections of thermal neutrons capture by coatings (Al is 0.231 barn; Cr is 3.05 barn; Ti is 6.09 barn) based on chromium are 16 times higher than the cross sections of the Zr (0.185 barn) capture. This would require introduction of fuel with enrichment of more than 5 % in ^{235}U or the use of a thinner cladding;

– alteration in the alloy chemical composition, and, consequently, in its properties under prolonged irradiation.

Coating with the Zr–Cr–N corrosion-resistant alloy (HIPMS)

Advantages:

– high corrosion resistance (*corrosion tests were carried out only in the open air*).

Disadvantages:

– data on the coating behavior before and after irradiation are missing;

– ^{14}C accumulation would entail problems connected to storage and processing of the spent fuel elements;

– cross sections of the thermal neutrons capture by coatings (Cr is 3.05 barn; N is 1.91 barn) based on chromium are 16 times higher than cross section of the Zr capture (0.185 barn). This would require introduction of fuel with enrichment of more than 5 % in ^{235}U, or the use of a thinner cladding;

– alteration in the alloy chemical composition of the alloy, and, consequently, in its properties during prolonged irradiation.

Development of zirconium pipes coated with austenitic steel by the pipe-in-pipe method

Advantages:

– low oxidation rate in water vapor; high melting point;

– introduction into production requires minimal changes in the existing design and production technology of the fuel elements.

Disadvantages:

– experimental data on the coating state before and after irradiation are missing;

– increased capture of thermal neutrons by the coating compared to the zirconium shell (Cr is 3.05 barn; Ni is 4.49 barn; Mo is 2.48 barn);

– production of short-lived radioactive isotopes of chromium, radioactive cobalt and molybdenum;

– decrease in plasticity at high temperatures, alteration in the chemical composition.

Thin-walled high-strength steel claddings. Claddings made of austenitic corrosion-resistant steel.

Advantages:

– low oxidation rate in water vapor; high melting point.

Disadvantages:

– alterations in chemical composition and, consequently, in properties under irradiation;

– production of the short-lived radioactive isotopes of chromium, radioactive cobalt and molybdenum;

– decrease in plasticity at high temperatures;

– increased thermal neutron capture in comparison with a zirconium cladding (Cr is 3.05 barn; Ni is 4.49 barn; Mo is 2.48 barn). This would require the use of enriched fuel of more than 5 % in ^{235}U, or introduction of a thinner cladding.

Fe–Cr–Al steel claddings

Advantages:

– alloy oxidation rate in water vapor at 1200 °C is 100 times lower than that of the zirconium alloy.

Disadvantages:

– experimental data on the cladding non-reactor properties and its behavior under irradiation are missing;

– the melting point temperature (T_m (Fe–Cr–Al) = 1500 °C) is by 355 °C lower than that of the zirconium alloy (T_m = 1855 °C);

– cross section of the thermal neutrons captures by the Fe–Cr–Al alloy (Fe is 2.56 barn; Cr is 3.05 barn; Al is 0.331 barn) is 13–14 times higher than that of zirconium (Zr is 0.85 barn). This would require the use of enriched fuel of more than 5 % in ^{235}U, or introduction of a thinner cladding;

– alteration in the alloy chemical composition and, consequently, in its properties exposed to prolonged irradiation.

Fuel with increased thermal conductivity. Uranium nitride (UN)

Advantages:

– nitride fuel density is 30 % higher than that of the UO_2, which makes it possible to design more compact and uranium-intensive cores with lower enrichment compared to the UO_2;

– UN thermal conductivity (λ_{UN} (1100 °C) = 22.8 W/(m · K)) is an order of magnitude higher than the UO_2 thermal conductivity coefficient (λ_{UO_2} (1100 °C) = 2.8 W/(m · K));

– temperature difference between the fuel center and the cladding in high-density fuel is 10 times lower in comparison with the same value in UO_2;

– heat removal from the fuel having higher thermal conductivity is significantly greater in comparison with UO_2.

Disadvantages:

– capture of thermal neutrons by ^{14}N (99.63 % content in nature, capture cross section 1.1 barn) with subsequent nuclear transformation into the ^{14}C long-lived radioactive isotope;

– spent fuel containing this isotope in amount exceeding the natural concentration is a source of biological threat. To dispose of spent fuel, it is necessary to elaborate a new process for its reprocessing guaranteeing the absence of this isotope leak from the radioactive waste storage facilities.

Uranium silicide (U_3Si_2)

Advantages:

– silicide fuel density is 11 % higher as compared to the UO_2, which makes it possible to design more compact and uranium-intensive cores with lower enrichment as compared to UO_2;

– ability to retain gaseous fission products; thermal conductivity of U_3Si_2 ($\lambda_{U_3Si_2}$ (1100 °C) = 17.3 W/(m · K)) is 6 times higher than the UO_2 thermal conductivity coefficient (λ_{UO_2} (1100 °C) = 2.8 W/(m · K));

– temperature difference between the fuel center and the cladding in high-density fuel is 6 times lower compared to the same value in the UO_2. Heat removal from fuel with higher thermal conductivity is significantly larger in comparison with UO_2.

Disadvantages:

– lower melting point temperature of the silicide uranium fuel T_m (U_3Si_2) = = 1665 °C compared to the UO_2 T_m (UO_2) = 2840 °C.

Composite ceramic fuel (UO_2 + Cr_2O_3 + SiC; UO_2 + BeO)

Advantages:

– thermal conductivity of the composite ceramic fuel is higher than the UO_2 thermal conductivity.

Disadvantages:

– lack of systematic data on mechanical, strength and thermophysical properties of the fuel, as well as on its behavior exposed to irradiation.

Cermet fuel

Advantages:

– proven technology (used in the nuclear submarine fleet).

Disadvantages:

– required increased enrichment.

Ceramic claddings

Advantages:

– SiC dissociation temperature (~ 2545 ± 40 °C) is 2 times higher than the cladding temperature in case of a design-basis accident accompanied by the loss of coolant;

– is not reacting with water vapor of up to 1300 °C; extremely low casing swelling value;

– cross sections for the thermal neutrons capture by silicon carbide (Si is 0.71 barn; C is 0.035 barn) are lower than those of zirconium.

Disadvantages:

– problems with cladding handling and use connected to fragility thereof under normal conditions;

– gas permeability of the SiC claddings;

– problems with the fuel element sealings (plug and cladding welding);

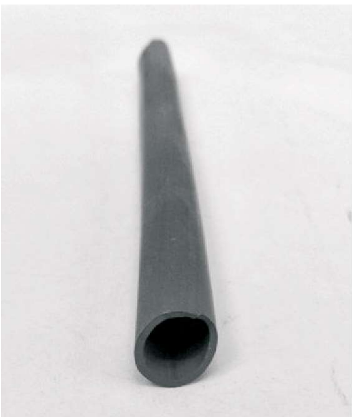
– expensive production of SiC fibers.

SiC/SiC composite materials are the ceramic matrices reinforced with high-strength ceramic fibers and appear to be the most complex materials, since they are significantly different from metals due to specific production process, low ductility, anisotropy, etc. Potential advantages include high (up to 1000 °C) heat resistance, chemical and dimensional stability.

NIIgrafit JSC together with JSC VNIINM developed a technological scheme of producing thin-walled long pipes based on the SiC/SiC heat-resistant ceramic system using a reinforcing frame made of the silicon carbide fibers [1, 15].

The main operations include: charge preparation by fine grinding of the original fillers together with a binder and small addition of a plasticizer; adding a plasticizer and preparing the molding compound; tableting/granulation of the molding mass; grinding; forming pipes from the molding material; heat treatment; siliconizing.

By means of extrusion molding, technology with high content of the finely dispersed silicon carbide with an average grain size of less than 3.5 μm was tested experimentally at the reduced content of plasticizer. Obtained compositions make it possible to form thin-walled tube made of SiC/SiC ceramics with the length of 700 mm and more in compliance with requirements in geometry and straightness (see the Figure) [1].



Sample of a thin-walled SiC/SiC ceramic pipe

At present, recommendations were proposed and requirements for additional equipment were formulated to improve technology in producing the thin-walled pipes from SiC/SiC ceramics with the length of more than 2 m in compliance with requirements in geometry and straightness [1].

The use of silicon carbide fuel element claddings in WWER and RBMK reactors would provide a radical increase in radiation safety of the modern nuclear power plants, as well as a possibility of introducing fuel assemblies in all operating WWER and RBMK reactors (water-water energetic reactor and high-power channel reactor).

At this stage of studying properties and development of technology with regard to considered materials on the tolerant fuel, it seems that it is possible to recommend composite materials for the fuel elements cladding based on the heat-resistant ceramic SiC/SiC system with the use of a reinforcing frame made of the silicon carbide fibers, and as fuel materials — materials with increased value of density, uranium capacity and thermal conductivity, i.e., nitride fuel and fuel made of uranium silicide [1, 11].

At present, these materials to the greatest extent meet requirements to the tolerant fuel, and their production technology, especially in regard to the nuclear fuel, has been worked out. More research is required on the silicon carbide to eliminate brittleness. Upon receiving positive results of experimental research under the reactor conditions, these materials could become competitive in the international market and would provide us with the future nuclear power industry.

Conclusions. The concept of tolerant fuel is considered as applied to the water-cooled power reactors.

Basic requirements for analyzing fuel safety based on the regulatory documents are provided.

Safety criteria for fuel elements and procedure for their calculated justification under the design-basis accidents are described.

Main characteristics of the new generation materials being developed for reactor cores are analyzed as applied to the tolerant fuel.

Based on comparing the presented information, composite materials based on the heat-resistant ceramic system SiC/SiC using a reinforcing cage made of silicon carbide fibers could be recommended as tolerant fuel for the fuel element claddings, and materials with increased density, uranium capacity and thermal conductivity values could be recommended as fuel materials, i.e., nitride fuel and uranium silicide fuel.

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