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DEVELOPMENT OF STRUCTURAL STEEL FOR FUEL ELEMENTS AND FUEL ASSEMBLIES OF SODIUM-COOLED FAST REACTORS

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The main results of development work and post-reactor studies of different structural materials for fuel-element cladding and hexahedral fuel-assembly jackets for sodium-cooled fast reactors are examined. Austenitic and ferritic-martensitic steels, including steel obtained by powder metallurgy, are examined as promising materials for fuel-element cladding for staged increase of fuel burnup.

Successful work on and development of fast reactors are largely determined by the functional properties of the structural materials at high damaging dose. The most important characteristics are the swelling and creep, which determine the shape change of articles, and the degradation of the mechanical properties.

The Bochnar All-Russia Research Institute for Inorganic Materials (VNIINM) is developing fuel elements and structural materials in close cooperation with Afrikantov Experimental Design Bureau of Mechanical Engineering (OKBM), which is the main contractor, and the Leipunskii Institute for Physics and Power Engineering (FEI), which is the scientific direactor for fast reactors. The results of post-reactor studies, which are being conducted at the Beloyarskaya nuclear power plant, at IRM, the Research Institute for Atomic Reactors (NIIAR), and the FEI, are very important for making decisions. The objects of post-reactor studies are materials-engineering, experimental, reference, and standard fuel assemblies.

The objective of the first comprehensive program for the development of radiation-resistance materials was to develop steel that would give burnup of at least 10% h.a [1]. In this program, the following steels were tested: austenitic steel in various states of mechanical-heat treatment – 08Kh16N11M3, 08Kh16N11M3T, 06Kh16N15M3B (EI-847), 06Kh16N15M3BR (EP-172), 06Kh16N15M2G2TFR (ChS-68); ferritic-martensitic steel Kh13M2BFR (EP-450), 05Kh12N2M, Kh12MSFBR (EP-823); and high-nickel alloys with 30–40% nickel. In 1987, it was decided on the basis of the results of the tests to use in BN-600 ChS-68 c.d. and EP-450 steels, respectively, as the standard materials for the fuel-element cladding and fuel-assem-

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TABLE 1. Chemical Composition of Austenitic Steel, wt %

Element	EI-847	EP-172	ChS-68	EK-164
C	0.04–0.06	0.04–0.07	0.05–0.08	0.05–0.09
Si	<0.4	<0.6	0.3–0.6	0.3–0.6
Mn	0.4–0.8	0.5–0.9	1.3–2	1.5–2
Cr	15–16	15–16.5	15.5–17	15–16.5
Ni	15–16	14.5–16	14–15.5	18–19.5
Mo	2.7–3.2	2.5–3	1.9–2.5	2–2.5
Nb	<0.9	0.35–0.9	–	0.1–0.4
Ti	–	–	0.2–0.5	0.25–0.45
V	–	–	0.1–0.3	0.15
B	–	0.003–0.008	0.002–0.005	0.001–0.005
P	<0.02	<0.02	<0.02	0.01–0.03
Ce	–	–	–	0.15

bly jackets. The steel developed provided failure-free operation of BN-600 with maximum burnup 11.2% h.a. and damaging dose 82 dpa. These same materials were adopted for BN-800 fuel-element cladding and fuel-assembly jackets.

Austenitic and martensitic steels, including steel dispersion-hardened by oxides and obtained by powder metallurgy, are being considered as promising materials for fuel-element cladding for staged increase of the fuel burnup of sodium-cooled fast reactors.

Steel for Fuel-Assembly Jackets

A fundamental solution to the problem the shape-change of fuel-assembly jackets was found by using ferritic-martensitic steel. Representative post-reactor studies showed that with this steel the shape-change of the fuel-assembly jackets is small: the degree of deformation along the entire length of a hexahedral tube is within 1% and depends weakly on the damaging dose, at least, to 100 dpa. Thus, for EP-450 steel the modulus of the radiation creep does not exceed $0.4 \cdot 10^{-6}$ $(\text{MPa}\cdot\text{dpa})^{-1}$, and the swelling at damaging dose 160 dpa does not exceed 0.4% [2]. The mechanical properties of the EP-450 steel, irradiated in BN-350 and -600, as fuel-assembly jackets were studied [3, 4]. The results were as follows:

1) the maximum hardening of the steel under irradiation is observed at low temperatures 290–370°C even with a low damaging dose 2–10 dpa; as the dose increases, the degree of radiation hardening decreases and above 60 dpa equals the characteristic value for material irradiated at higher temperature; the dose dependence at moderate and elevated irradiation temperatures (370–450 and 450–560°C) is characterized by saturation even when the dose reaches 10–15 dpa, the level remaining unchanged to high damaging dose;

2) the residual plasticity of EP-450 steel irradiated at >370°C is at least 3% at the appropriate working temperature and at least 1% at room temperature; and

3) the maximum decrease of the toughness is observed at low irradiation temperature; for dose above 10–20 dpa, the properties are restored and for 80–90 dpa the toughness of steel irradiated at different temperatures is approximately the same; the temperature of the brittle-ductile transition does not exceed 130°C for the most dangerous section of the fuel-assembly jacket, and in all other sections the temperature of the brittle-ductile transition is lower.

EP-450 steel has been adopted as the material for fuel-assembly jackets in the BN-800 under construction, and there is every reason to believe that the fuel-assembly jackets made of this steel will not prevent attaining higher fuel burnup.

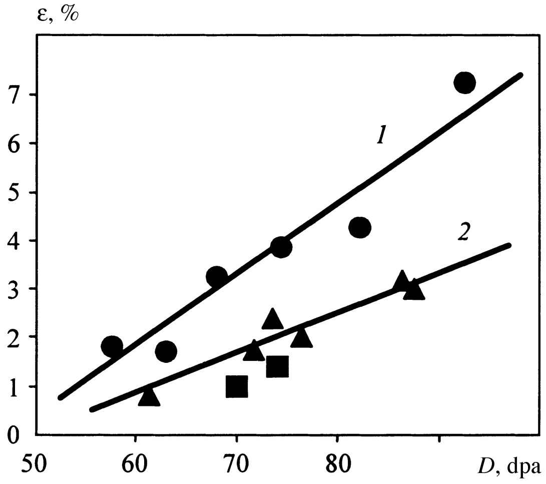

Fig. 1. Shape-change of BN-600 fuel-element cladding made of ChS-68 c.d. steel versus the damaging dose for experimental (*1*) and standard (*2*) fuel assemblies.

Steel for Fuel-Element Cladding

Austenitic chromium-nickel steel. The previously developed and well-studied steel EI-847 (Table 1) was taken as the base steel for developing a new austenitic steel; the new steel EP-172 was obtained by modifying EI-847 with boron (0.003–0.008%) [5]. EI-847 was also used to develop one other steel – ChS-68, doped with (aside from boron) silicon and titanium. The new austenitic steel EK-164 has a higher content of nickel and is doped with titanium, niobium, vanadium, boron, silicon, phosphorus, and cerium.

Radiation-resistance steel is developed taking account of the following structural factors with control swelling [6, 7]:

1) solid-solution determined by the concentration of the matrix of dopants (first and foremost, nickel) and impurity elements (C, Nb, Ti, B, Si, rare-earths, and other elements), which form complexes of point defects – impurity with altered diffusion characteristics;

2) phase instability, manifested in the formation of particles of precipitate, whose nature, composition, volume fraction, morphology, and location largely determine the process of nucleation and pore growth; and

3) dislocations, for which cold deformation (15–25%) increases the density of dislocation sinks of point defects and makes it possible to delay considerably the onset of intense pore formation.

The structural factors controlling swelling are interrelated in a definite manner and influence one another. For this reason, increasing the radiation resistance of austenitic steel is an optimization problem. The data obtained from post-reactor studies showed that the approach based on the use of the indicated factors made it possible to decrease substantially the swelling of austenitic steel [8]. On the basis of the results of such studies, the steel ChS-68 c.d. was recommended as the standard material for the cladding of BN-600 fuel elements.

Analysis has shown that two groups of results of profilometry performed on fuel-element cladding made of ChS-68 c.d. steel can be singled out at different stages of the mastery of the steel, which were obtained on experimental fuel assemblies and are standard fuel assemblies with a much higher rate of swelling (Fig. 1). The manufacturer changed three times during the production of fuel-element tubes made of ChS-68 steel: first the tube works in Dnepropetrovsk, then PNTZ company in Pervouralsk, and the current manufacturer is MSZ company in Elektrostal. The equipment for rolling and heat-treatment as well as the schemes for fabricating tubes are different in these plants. On the basis of studies, a decision was made to use diffusion annealing to improve the quality of the tubes; this made it possible to increase the structural uniformity of the material considerably [9].

The main requirements for fuel-element cladding for fast reactors are high durability and plasticity. Tubular samples made of ChS-68 c.d. steel with different technological variants were made for comparative pre-reactor durability tests [9].

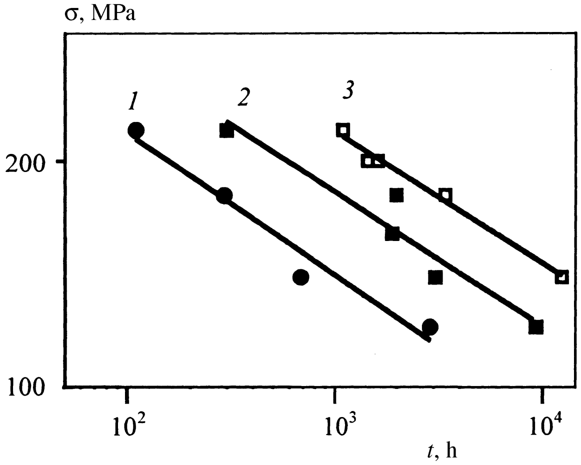

Fig. 2. Durability with uniaxial stretching of 6.9 × 0.4 mm tubes made of ChS-68 c.d. steel and fabricated at PNTZ (*1*), MSZ (*2*), and MSZ with diffusion annealing and plug drawing (*3*).

As one can see in Fig. 2, tubes with diffusion annealing, optimized regime of austenitizing annealing, and plug drawing had the highest durability at 700°C.

The effectiveness of the methods proposed for optimizing the structure of fuel-element cladding made of ChS-68 c.d. steel had to be confirmed experimentally by irradiating reference fuel assemblies. As post-reactor studies showed, the cladding fabricated by means of the improved technology has on average 1.5 times smaller shape-change than previously provided cladding. The investigations of the strength and plastic properties of ring-shaped samples of the fuel-element cladding of a reference fuel assembly irradiated to damaging dose 74 dpa showed that the material maintained its deformation capability and high strength. Computational estimates of the stress–strain state made it possible to predict fuel-element serviceability for reference fuel assemblies to dose 95 dpa.

Validation of the serviceability of austenitic cladding steel – ChS-68 to 95 dpa and EK-164 to110 dpa – is currently continuing on BN-600 using experimental, reference, and standard fuel assemblies. To attain higher burnup, the austenitic steel may turn out to be unsuitable because of swelling at a higher damaging dose. For this reason, the main efforts are being directed at investigating low-swelling ferritic-martensitic steel as the material for fuel-element cladding.

Ferritic-martensitic steel. Its advantage is a higher, as compared with austenitic steel, resistance to swelling, high embrittlement temperature, and radiation creep; its disadvantages are lower heat-resistance, proneness to low-temperature radiation embrittlement, and unsatisfactory corrosion resistance during long-term storage in cooling-pond water.

Post-reactor studies of EP-450 steel fuel-element cladding irradiated in BOR-60, BN-350, and BN-600 demonstrate the relative structural stability of the steel and high resistance to vacancy swelling to damaging dose 160 dpa. The main factor limiting the use of EP-450 steel for fuel-element cladding in fast reactors is its heat-resistance, which is too low. To prevent damage to fuel elements due to low heat-resistance of the cladding and intra-fuel-element corrosion processes, the maximum irradiation temperature must be limited to 650°C.

Two cladding materials, which permit fuel-element operation to 140 dpa and are now under consideration, are the heat-resistant complexly doped steel EK-181 (16Kh12V2FTaR) and the steel ChS-139 (20Kh12NMVBFAR) [10, 11]. The EK-181 and ChS-139 steels have been mastered in metallurgical industry; mastering the production of especially thin-walled tubes has begun. These steels differ from EP-450 by the additional doping with carbon, nitrogen, tungsten, and tantalum and a somewhat lower content of chromium (Table 2). Such doping ensures stability of the hardening phases and resistance to recrystallization processes and increases the heat-resistance (Fig. 3). Thus, the time to failure at test temperature 700°C with biaxial stretching of tubes made of EK-181 and ChS-139 steel is 1.5–2 times greater than for EP-450 steel under the same testing conditions.

TABLE 2. Chemical Composition of Ferritic-Martensitic Steel, wt %

Element	EP-450	EK-181	ChS-139
C	0.1–0.15	0.1–0.2	0.18–0.2
Si	<0.6	0.3–0.5	0.2–0.3
Mn	<0.6	0.5–0.8	0.5–0.8
Cr	12–14	10–12	11–12.5
Ni	<0.3	<0.1	0.5–0.8
Mo	1.2–2.8	<0.01	0.4–0.6
Nb	0.25–0.55	<0.01	0.2–0.3
V	0.1–0.3	0.2–1	0.2–0.3
B	0.004	0.003–0.006	0.003–0.006
W	–	1–2	1–1.5
Ti	–	0.03–0.3	0.03–0.3
Ta	–	0.05–0.2	–

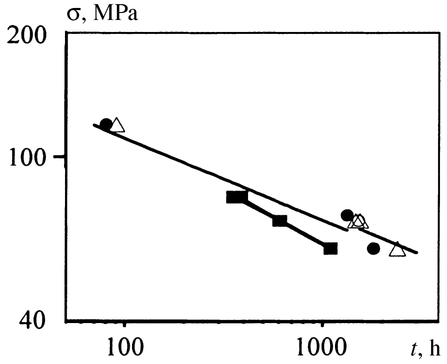

Fig. 3. Durability of fuel-element tubes made of ferritic-martensitic steels EP-450 (■), EK-181 (△), and ChS-139 (●) with biaxial stretching by internal pressure at test temperature 700°C.

The results of studies of EK-181 steel samples irradiated in BOR-60 at 320°C to damaging dose 8 dpa show that the toughness and plasticity remain at acceptable levels [11].

The results obtained thus far show the promise of using heat-resistant steels of the type EK-181 and ChS-139 as the material for the fuel-element cladding in fast reactors. For experimental validation, there are plans to irradiate, starting in 2010, in BOR-60 one fuel assembly with fuel-element cladding and in BN-600 two materials-engineering fuel assemblies with EK-181 and ChS-139 steel samples to maximum damaging dose 140 dpa.

To attain a damaging dose of 180 dpa, a ferritic-martensitic steel dispersion hardened by oxides is being developed by power metallurgy methods at the VNIINM. The required increase of the characteristics of creep and toughness is attained by hardening the matrix with disperse particles of yttrium and titanium oxides. A technological scheme for obtaining steels dispersion hardened by oxides on the basis of EP-450 steel has been tested on the basis of world and domestic experience [12]. Evaluation of the deformation capability has shown that the steel retains high elongation after cold deformation to 60%.

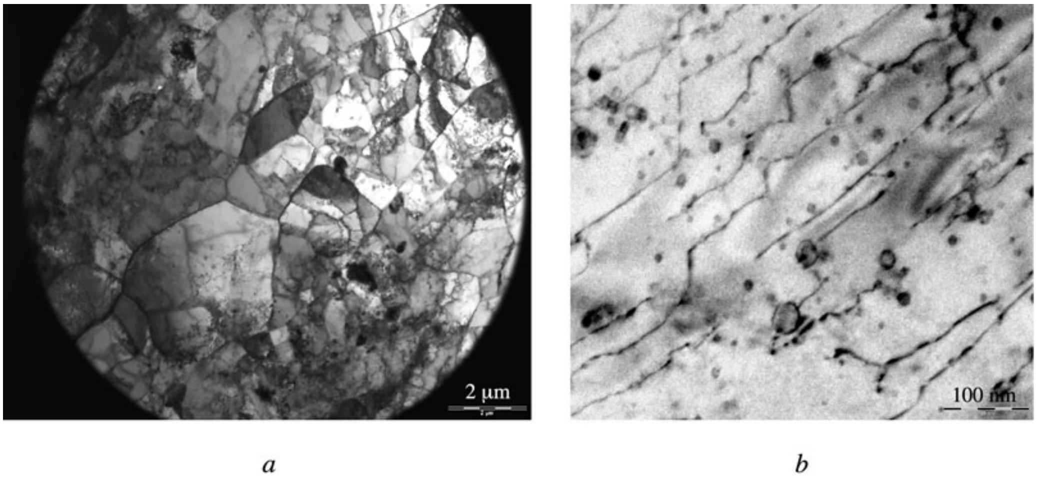

Fig. 4. Grain structure (*a*) and oxides (*b*) in a casing tube made of EP-450 steel dispersion-hardened by oxides.

On this basis, a regime has been chosen for rolling tubes with intermediate heat treatments; this regime is being implemented at the MSZ.

Tests for thermal creep have been performed on flat samples cut from a casing tube (EP-450 steel) and a hot-rolled plate (EP-450 steel, dispersion-hardened by oxides). Even though there is some difference in granularity, the nonuniformity of the distribution of the oxide particles and the presence of a small fraction of quite large insoluble oxides, the rate of thermal creep of EP-450 steel, dispersion hardened by oxides, was 100 times lower than the rate of thermal creep of EP-450 steel [12]. The first experimental batch of especially thin-walled tubes with dimensions 6.9 × 0.4 mm from EP-450 steel dispersion-hardened by oxides was obtained at the VNIINM in 2006. The structure of the tubes obtained consists of regions with recrystallized grains ranging in size from 10 to 25 μm and subgrains ranging in size from 0.1 to 3 μm (Fig. 4*a*). Uniformly distributed oxides are observed inside grains and subgrains (Fig. 4*b*). The average size of the oxide particles is ~7 nm and the concentration is $\sim 10^{22}$ m^{-3}.

Substantial efforts are now being made to develop a fabrication technology (powder metallurgy and tube production) and to perform complex pre-reactor studies of fuel-element tubes made of ferrite (based on EP-450 steel) and martensite (based on EK-181 steel), which are dispersion hardened by oxides, and their weld seams. Samples of these steels are also present in the composition of two materials-engineering fuel assemblies, whose irradiation in BN-600 will begin in 2010.

Conclusion

Increasing fuel burnup in sodium-cooled fast reactors is being held back by the swelling of fuel-element cladding, consisting of austenitic steel, at high damaging doses. Studies now being conducted in BN-600 show that damaging doses 95 and 110 dpa can be attained for ChS-68 and EK-164 steel, respectively, cladding.

An increase of the serviceability of fuel-element cladding of fast reactors to higher damaging dose 140 dpa is solved by using new ferritic-martensitic steels EK-181 and ChS-139. Heat-resistant steels, dispersion-hardened by oxides, based on EP-450 and EK-181 are being developed in order to ensure fuel-element cladding serviceability to damaging dose 180 dpa.

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