PROBLEMS OF CREATING FUEL ELEMENTS FOR FAST GAS-COOLED REACTORS WORKING ON N_2O_4-DISSOCIATING COOLANT

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A variant of fast gas-cooled reactors is one using dissociating N_2O_4 nitrogen tetroxide as a coolant. This type of reactors is promising because of great thermal effects of dissociation reactions while heating and recombination while cooling; small latent heat of evaporation; high heat transfer coefficient owing to additional heat transfer in a chemical reaction; high N_2O_4 density in a gas state at operation parameters. The mentioned advantages give possibility to create a small turbine, heat exchange apparatus and to get high heat production in the active zone. All this opens new ways to increase power plants effectiveness.

Embodiment of the ideas concerning effective use of N_2O_4 dissociating gas in fast reactors demands a great number of theoretical and experimental research, development and creation of new constructions and apparatus; determination of their utmost operation parameters under simultaneous influence of in-pile irradiation and coolant medium; and confirmation of their workability in concrete atomic power station (APS) operating conditions.

Rather hard operating conditions are typical for fuel elements of gas-cooled reactors with N_2O_4 dissociating coolant. The cladding and fuel centre temperatures are 950 K and

1400-1600 K respectively. Fluence and heat flux from the fuel elements surface are 2.10^{23} n/cm^2 and $2,8.10^6$ w/m^2, and the burnup value of the nuclear fuel is 10% Fima. A specific feature of fuel elements operation in gas-cooled reactors with N_2O_4 dissociating gas is the contact of the chemically active coolant with the cladding material and, in the case of its seal failure with the nuclear fuel. That is why, besides general demands /2/ to fuel elements operating in reactors with N_2O_4 coolant, there is a demand to provide compatibility of the nuclear fuel and the coolant. This demand is of great importance, as schemes of being developed APS operating on dissociating gas are one-circuit; their radiation safety will greatly depend on corrosion resistance of the nuclear fuel in the coolant.

The problem of corrosion resistance of the nuclear fuel to the coolant was solved by development and creation of dispersion nuclear fuel using N_2O_4 corrosion resistant materials as a matrix; and also some new chemical uranium compounds such as UO_2CrO_3, $UO_2Nb_2O_5$ that have high compatibility with the N_2O_4 coolant /3/.

Dispersion nuclear fuel on the base of uranium dioxide in a chromium matrix is the most suitable for fuel elements of fast gas-cooled reactors active zone with N_2O_4 dissociating coolant. This fuel at uranium dioxide density of 10 g/sm^3 ensures high uranium charge per a volume unit. Preparation technology of this kind of nuclear fuel includes the following main steps /2,4,5/:

- production of uranium dioxide microspheres;
- covering them with a corrosion resistant chromium coating;
- pressing chromium coated microspheres into fuel pins and their subsequent shaping;
- thermal treatment for elimination of residual stresses.

Prepared by this way dispersion compositions with 30 vol. % chromium matrix have the structure of uniformly distributed particles of a heat generation phase from uranium dioxide in the chromium matrix along the whole volume of the fuel pin

(Fig. 1) and have rather high thermophysical and strength characteristics. Thermoconductivity of dispersion nuclear fuel on the base of uranium dioxide in the chromium matrix is 5-8 times higher than that of uranium dioxide and equals 20-25W/mK /4/.

The study of dispersion compositions behaviour on the base of uranium dioxide with 30 vol. % chromium matrix indicates their satisfactory corrosion resistance in N_2O_4 coolant. Carried out research of UO_2-Cr dispersion composition oxidation kinetics during 360 hours at different temperatures and pressures of the coolant shows that while increasing the test temperature from 520 to 1020 K and N_2O_4 pressure from 0.1 to 16.0 MPa corrosion rate rises propertionally to pressure to 0.3-0.5 power, and one can see the maximum weight gain of samples during the first 100 hours of the test. The following prolongation of the test leads to the dispersion fuel oxidation rate decrease.

The investigation of chromium matrix volume content in the dispersion composition shows that its content decrease till 20% leads to sharp deterioration of the dispersion nuclear fuel corrosion resistance in N_2O_4.

Carried out corrosion tests of the dispersion nuclear fuel on the base of uranium dioxide in N_2O_4 medium showed, that such types of compositions having heat generation phase volume content not more than 70% are sufficiently corrosion resistant. Corrosion rate is 10^{-1} $g/m^2 \cdot h$ at the temperature of 1020 K and the pressure of 16.0 MPa.

Nuclear fuel corrosion tests in statical conditions of N_2O_4 medium allowed to get quantative characteristics of the given material corrosion resistance. But in the case of in-pile seal failure of the fuel element cladding the corrosion resistance of the coolant and the nuclear fuel will be different. High coolant circulation rate with defects being present in the fuel element cladding can lead to corrosion and erosion processes evolution, to appearance of nuclear fuel corrosion products in the loop, and to its irradiation contamination. Dispersion nuclear fuel corrosion rate is greatly influenced by neutron irradiation, which at high burn-ups results in damages and cracks of the matrix material. The structure with a great number of microcracks and macrocracks in the dispersion number fuel chromium matrix can lead to corrosion nonresistance of the matrix in the N_2O_4 coolant.

Investigation of models behavior of dispersion nuclear fuel elements with artificial defects in N_2O_4 was carried out at the temperature of 820 K, the coolant pressure of 16.0 MPa during 360 hrs. The coolant flow rate in the thermophysical gap between the casing tube and the fuel element cladding was 45-65 kg/hr. The tests were performed on a high pressure installation,its scheme and operation principle are given in /6/.

It is shown, that the contact of the fuel pin with N_2O_4 through the hole of 0.3 mm diameter in the fuel element cladding results in the pin oxidation to the depth of 600-900 mkm, in its volume increase at the expense of reconstruction of UO_2 crystal lattice into U_3O_8 lattice and, respectively, in the cladding deformation. The maximum cladding deformation after 360 hrs of testing was 0,065 the creep rate was 2.10^{-4} hr^{-1}. It was noted that damage evolution and uranium dioxide corrosion products in the coolant circuct had not been registered while testing.

Corrosion resistance investigations of 09X16H15M3B type stainless steel showed that the fuel element cladding corrosion rate in the temperature range of 800-1000K is governed besides chemical kinetics by the whole complex of mechanical stresses, which the cladding experiences, and their relaxation rate /7/. In the stressed state of the cladding phase stresses aroused by crystal lattice reconstruction during oxidation, thermal stresses caused by large thermal flux from the pin, and stresses caused by fluctuations of the temperature, rate, oxidizer turbulent flow pressure play the main role. Ceramic oxide films on the fuel element cladding surface have sufficiently high compression strength and low tensile strength. The scope of turbulent fluctuations providing fatique load of the oxide films depends not only upon the linear rate of the N_2O_4 flow but also upon the characteristic size of the gap /8/. In this case the flow temperature fluctuations can result in oxide surface layer damage (the level of corresponding stresses of 10 MPa at the flow having Re-number of 10^4) and rate and flow pressure fluctuations (the stress level of 10^3 - 10^4 Pa) lead to erosion of the damaged layers. It is found that, when the hydraulic gap size is equal to 0.5-0.7 mm, stress fluctuations prevail at oxide film thickness being more 10-30 mkm and so one can get high corrosion resistance of the fuel element cladding if the oxide film thickness is not more 10-30 mkm in the period of operation.

A great number of pre-reactor experimental and theoretical research of the nuclear fuel and fuel elements with the dispersion fuel allowed to prognose the possibility of reliable fuel elements creation for atomic power stations working on dissociating N_2O_4 coolant, however, the final choice of a nuclear fuel, cladding material and fuel element construction can be done only after in-pile tests.

Radiation stability and workability of fuel elements is governed both by radiation stability of their main components (nuclear fuel and fuel element cladding material) and the fuel element construction . Workability estimation of fuel elements according to the investigation results of irradiated nuclear fuel and cladding material for the given structure of the fuel element must be confirmed by in-pile life tests of fuel elements.

In-pile tests of the nuclear fuel and the fuel elements with UO_2-Cr dispersion fuel were carried out in BSSR АН ИЯЭ ИРТ[1)] reactor working on thermal neutrons in ampoule and loop channels of gas-cooled loop istallation (Fig. 2) with dissociating N_2O_4 coolant /9, 10/.

Carried out in-pile test and post-reactor investigations allowed to establish main behaviour regularities of UO_2-Cr dispersion-type nuclear fuel and fuel elements in the conditions of neutron irradiation and N_2O_4 dissociating coolant influence.

Microctructural investigations of the irradiated nuclear fuel reveal its high dimension and structure stability. There are no processes of components mass transfer in the fuel element rod volume and interaction between them at the burnup of 10% heavy atoms in the dispersion type nuclear fuel.

I) ИРТ - reactor's name; АН - Academy of sciences;
ИЯЭ - Institute of nuclear energetics;

The dispersion nuclear fuel structure optimisation shows, that optimum size of fuel kernels is less than 600 mkm. It is necessary to note that the probability of UO_2 microspheres and matrix material cracking increases with the UO_2 fissile phase particles size growth (Fig. 3). On the other hand fuel particles size decrease increases probability of formation of contacting fuel particles chains at rod pressing.

Obtained experimental data agree well with calculations of stress-strained state of dispersion systems under the influence of three-axis tensile stress /8/. The maximum breaking stress value is shown to localize in the places where the distance between kernels at the surface or between the kernel and the surface of the rod is minimum.

As a rule irradiation leads to mechanical characteristics changing of the fuel element cladding material. Simultaneous action of n , γ - irradiation and chemically active N_2O_4 atmosphere can have more noticeable influence on mechanical properties of structural materials than each action separately. The main characteristics of strength and plasticity σ_B $\sigma_{0,2}$ δ_{equal} δ_{total} of 09X16H15M3B steel investigated as a fuel element cladding material of gas-cooled reactors with dissociating N_2O_4 coolant, irradiated in N_2O_4 till the fluence of $2,4 \cdot 10^{20}$ n/cm^2 (E 0,1 Mev) show that there is growth of ultimate strength and yield limit and reduction of plasticity in the temperature range of 293-673K. At the temperature growth till 725K and higner δ_{total} decreases till 10% and δ_{equal} decreases till 3,5-4% simultaniously σ_B and $\sigma_{0.2}$ values become lower than initial ones /12/. Received results of the investigation of fuel element cladding material corrosion mechanical properties of 09X16H15M3B steel allow to make conclusions about sufficient store of strength and plasticity characteristics of this steel, being irradiated for more than 1000 hours in the atmosphere of dissociating N_2O_4 coolant.

Received experimental results, confirming high irradiation resistance of dispersion-type nuclear fuel on the dioxide uranium base, its compatibility with the fuel element cladding material (09X16H15M3B steel), low value of gaseous fission products (gfp) release under fuel element cladding (less 10% of gfp formed) and satisfactory corrosion mechanical characteristics of the cladding material allow to make the conclusion about the possibility of creation of fuel elements for gas-cooled reactors with dissociating N_2O_4 coolant.

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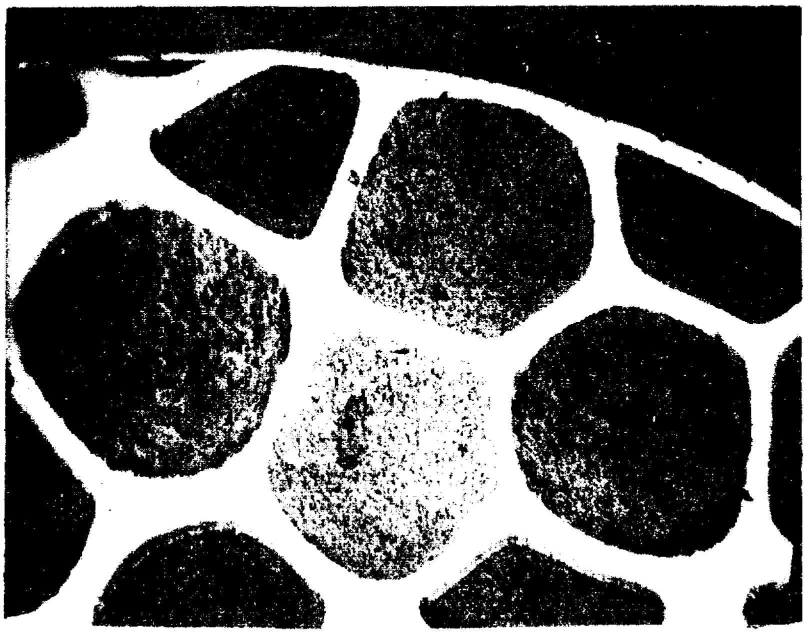


Fig.1. The microstructure of dispersion nuclear fuel on UO_2 base with 30 vol % chromium matrix (x80).

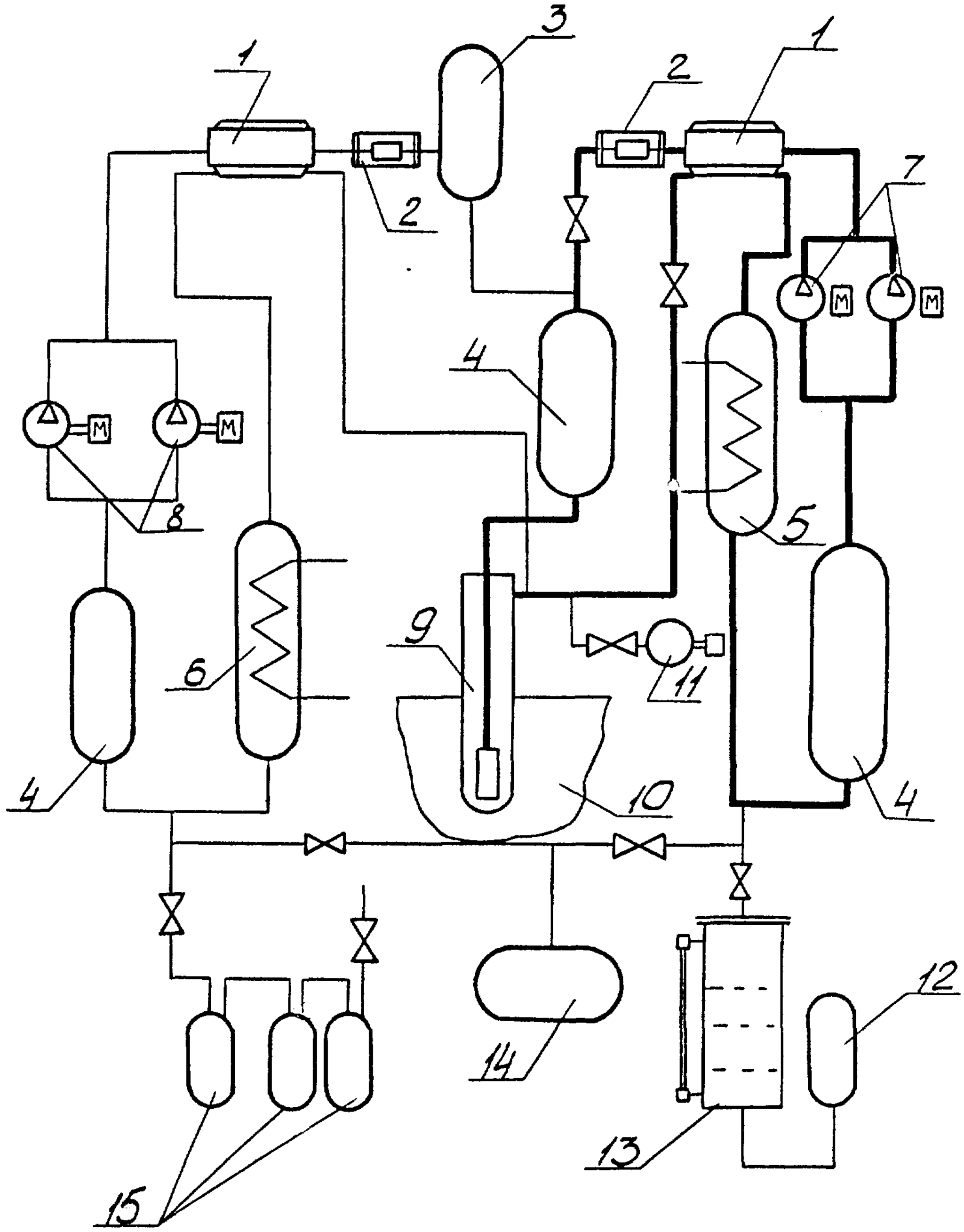
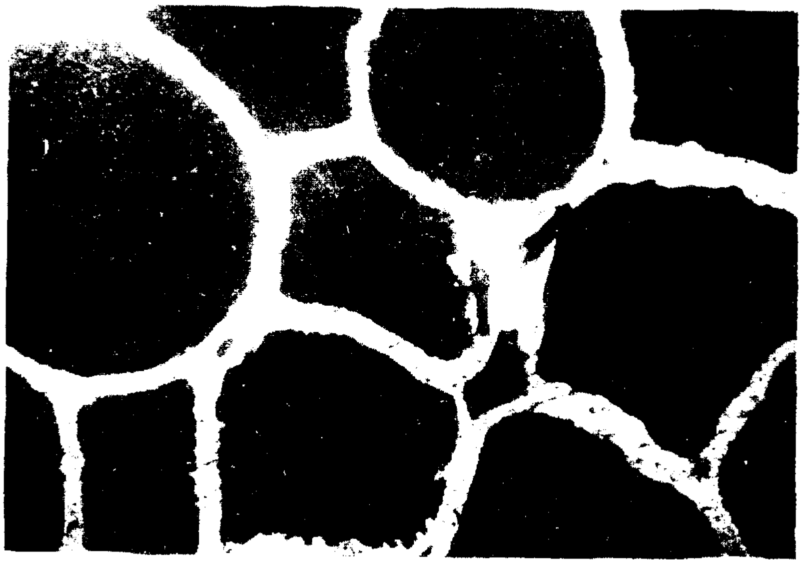


Fig 2. Principle scheme of a gas-cooled loop plant working on dissociating N_2O_4 coolant for investigation of fuel element workability.

1 - evaporator, 2 - electrical heater, 3 - pressure accumulator, 4 - filter, 5 - main circuit condensator, 6 - CAPX condensator circuit, 7,8 - valves, 9 - loop canal, 10 - active zone of the reactor, 11 - vaccuum pump, 12 - pH-meter, 13 - reservoir-neutralizer, 14 - feeding reservoir, 15 - exposure volume.

a



b

Fig 3. The microstructure of dispersion nuclear fuel on the UO_2 base with 30 vol % chromium matrix after irradiation (x80).

a) UO_2 fissile phase particle size 400-600 mkm;

b) UO_2 fissile phase particle size 600-900 mkm