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THERMOPHYSICAL PROPERTIES OF FUEL CLADDING WITH VARIOUS VACUUM-ARC COATINGS

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The developed research technique for determining emissivity of thin-walled cylindrical samples was used to investigate, in the mode of heat transfer by radiation, thermophysical properties of Zr1Nb fuel cladding samples with different coatings deposited by vacuum-arc method. It has been shown that CrN coating, compared to Zr1Nb-base alloy and Cr coating, can ensure a more effective heat expulsion by radiation in the event of loss-of-coolant accident and high temperature excursion This occurs according to the Stefan-Boltzmann radiation law and due to a higher value of total emissivity for CrN.

INTRODUCTION

Heat transfer from fuel pellets to fuel cladding and then into the coolant is an important stage of energy transformation in a thermal-neutron nuclear reactor. The heat transfer phenomenon generally is associated with the processes of heat conductivity, convection and radiation. In the case of normal reactor operation the radiation process makes an insignificant contribution to the heat transfer. In particular, according to [1] a fraction of heat radiation in thermal conductivity of the fuel-cladding contact at temperatures from 300 to 500°C is ~ 2%. The situation will cardinally change during severe accident scenarios. A maximum design accident with loss of coolant has been imitated during high-temperature testing of the WWER-1000 fuel element in the reactor MIR and by computer simulation [2]. It has been shown that at cladding temperature of about 1000°C the ratio of heating, convection and heat radiation in the released power is 52, 15 and 33 %, respectively. The fuel cladding temperature can be decreased by increasing the emissive power that will prevent the zirconium steam reaction (zirconium-produced gaseous hydrogen) and loss of tightness. And the emissive power of the fuel element can be increased by depositing coatings having radiative characteristics exceeding these of Zr1Nb. The deposited coating should also ensure a high resistant property of the fuel cladding in the case of the zirconium steam reaction.

Note, that for the gas-cooled very high-temperature reactors (VHTR) the heat radiation becomes an important channels of heat energy dissipation and in this connection a considerable attention has been given to the emissive power of candidate materials with various coatings [3].

In view of the aforesaid there is a need for studying thermal characteristics and emissive power of various coatings on structural materials applied in the nuclear energy engineering. The goal of this study was to investigate fuel cladding samples (of Zr1Nb alloy) with different oxidation-protective zirconium coatings. Experimental data on the heart-transfer properties should be taken into account when using modified fuel elements.

SAMPLES AND RESEARCH TECHNIQUES

The technique of vacuum-arc coating deposition on the samples, cut from the fuel cladding (diameter 9.2 mm, wall thickness 0.65 mm), is described in [4, 5]. In experiments we used the samples having different surface states: 1) initial state, 2) ion-cleaned surface (ICS), 3-5) surface layer oxidized during different exposure time, 6–7) coated with a Cr layer of different thickness, 8) coated with a CrN layer, 9) coated with a Cu layer.

The technique applied for thermophysical studies was based on the use of a heating element (HE), providing a preset power-level maintenance with a high degree of accuracy (up to 0.1%) in all experiments. A schematic diagram of the experimental device is shown in Fig. 1.

A heating element comprises a cylindrical copper case of 50 mm length and an internal resistance furnace made from wire of 0.15 mm diameter bifilarly wound on a ceramic pipe. Insulation between the heater spiral coils and on their outer surface was made of the mix of beryllium oxide powder with “liquid glass” which after drying was backed at temperature of 600 °C.

The outer diameter of the heater working part corresponded to the internal diameter of the fuel rod that ensured a reliable thermal contact between them. The length of initial samples and samples with different coatings was 30 mm.

The heater was powered by alternating-current mains. In the course of experiment the constant power level of the heater was maintained using two series-connected current stabilizers. Temperature measurements on the outer surface of the heater and samples were carried out in a vacuum chamber under residual gas pressure of about $(7…8)\cdot10^{-6}$ Torr in the state of stable thermal equilibrium. Temperature was measured accurate to 0.1 K. using a differential chromel-alumel thermocouple. Thermophysical characteristics of the samples of various coating material were determined under the same conditions as for heater calibration.

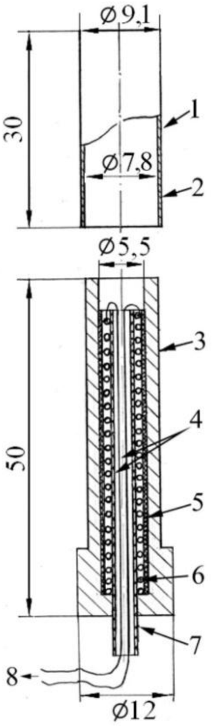

Fig. 1. Diagram of thermophysical measurements of samples: 1 – thermocouple farening point; 2 – sample; 3 – copper block; 4 –- heater terminals; 5 – electrical insulation; 6 – filament heater; 7 – ceramic pipe; 8 – wires connecting the power source

RESULTS AND DISCUSSION

Fig. 2 shows the kinetics of thermal equilibrium attainment in the fuel cladding samples with different surface states (initial surface state, ion-cleaned surface, oxidized surface, chrome coated surface, chrome nitride) after heating element turning on. One can see that for a Zr1Nb sample with initial surface state and for a sample with ion-etched surface the curves $T(t)$ differ slightly and attain the saturation (T_{HE}=267 and 270 °C) at t_s≈50 min.

The curves $T(t)$ of the samples with oxidized surface (oxidation for τ=6 and 11 h at 650 °C) and with the CrN coating of 5μm thickness are well coinciding and attain the saturation at lower temperature ($T_s \approx 225$ °C, t_s≈40 min). The sample oxidized for $\tau = 5$ h is characterized by $T_s = 207$ K and t≈60 min. Samples coated with Cu and Cr are characterized by t_s≈60…70 min and significantly higher values of saturation temperature. For the coating of Cu, the T_s = 339 °C, for the coating of Cr with a thickness of 8.8 and 4.5 μm, the T_s = 313 and 305°C, respectively.

The histogram in Fig.3 gives a notion about the relation T_s for the fuel cladding samples with different surface states and coatings, and about the spread of T_s values for different samples. It is seen that for oxide and nitride coatings lower values of T_s are characteristic.

As is known, CrN is a semiconductor [6] and pure zirconium dioxide is a good insulator – in it an ionic conduction is almost absent [7]. Metal coatings show higher values of $T_н$. A copper coating, compared to other metal coating (Cr of 4.5 μm and Cr of 8.8 μm) are characterized by a minimum value of $\Delta T = T_{HE} - T_s$; copper has a higher heat- and electric conduction [8].

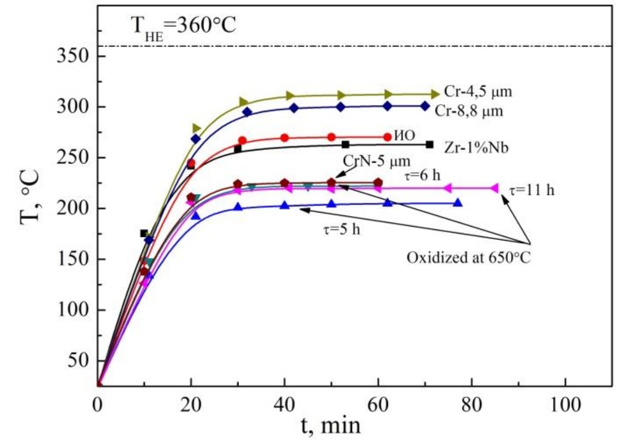

Fig. 2. Kinetics of attainment of thermal equilibrium in the samples with different surface states. The dashed line corresponds to temperature T_{HE} of the surface of the heating element without a fuel cladding sample

Below we consider the heat transfer peculiarities in our experiments. For a cylindrical wall with a symmetric central heat source the constant linear power is characterized by a constant value of the linear heat flow in the radial direction Q_R (see [9]). The applied heating element design and the ratio of its dimensions to fuel cladding sample dimensions allow us to realize the condition Q_R = const for the initial sample and for the coatings in the form of coaxial cylinders on the fuel cladding surface.

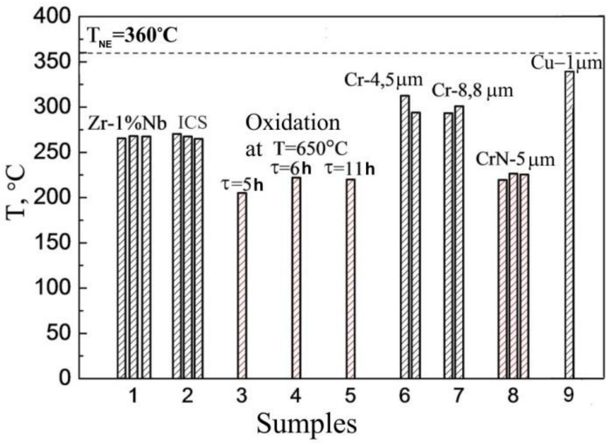

Fig. 3. Histogram of thermal equilibrium temperature values for samples with different coatings radiating in vacuum The dashed line corresponds to temperature T_{HE} on the surface of the heating element without a fuel cladding sample

The condition of absence of other channels of heat transfer from the heating element, besides thermal radiation, permits to estimate the heat radiation characteristics for different fuel cladding coatings.

To calculate the heat transfer we use the Stefan-Boltzman equation for gray bodies [9]

$$Q_R = \varepsilon c_0 \left[\left(\frac{T_S}{100} \right)^4 - \left(\frac{T_0}{100} \right)^4 \right] \times \pi d. \qquad (1)$$

The radiation heat-transfer coefficient can be written in the following form [9]

$$\alpha_R = \frac{Q_R}{\left(T_s - T_0\right)}. \qquad (2)$$

In the above equations d –outer diameter of sample; c_0=5.76·10^{-8} W·m^{-2}·K^{-4} – the Stefan-Boltzman constant; T_s– radiating surface temperature and T_0 – ambient temperature, ε – total emissivity of radiating surface which is the ratio between the gray body total self-radiation density and absolute black body total radiation density at the same temperature. The total emissivity is the material characteristic depending of both its surface temperature and surface state.

The normal total emissivity of metals can be optimized using the formula obtained by improved electromagnetic theory [10]

$$\varepsilon(T) = 5,76(\rho T)^{1/2} - 17,9\rho T + 58,6(\rho T^{3/2} - \\ -2\pi c\tau(870\rho)^{1/2} - 5900\rho T^2 + 3250\rho^{3/2}T^{5/2}), \qquad (3)$$

where $\tau = \dfrac{m}{\rho N e^2}$ is the relaxation parameter, (m and e – mass and charge of electron, N – number of free electrons, c – velocity of light, ρ - resistivity).

In work [11], in Fig. 3, for the Zr1Nb alloy, the temperature dependence of the electrical resistance in a wide temperature range (20…700 ºC) and in table 2 the specific electrical resistance ρ=59 μΩ·cm for T = 20 ºC is given. From these data it is easy to estimate the resistivity $\rho\approx$98 μΩ·cm at T=T_s=541 K.

For Zr1Nb alloy at T_s=541 K and ρ=98 μΩ·cm [11], with taking into account the three first terms of formula (3), we have $\varepsilon\approx$0.20. Then for Zr1Nb we obtain from formula (1) Q_{RZr1Nb}=26 W·m^{-1}.

To obtain an estimate of the degree of emissivity ε of Zr1Nb fuel cladding samples, we substitute ρ=98 μΩ·cm in the formula (3) and the experimental value T_H = 541 K. To obtain dimensionless ρT, $(\rho T)^{1/2}$ and $(\rho T)^{3/2}$ in the formula, we transform the dimension of each multiplier so as to make the reduction. We use the following ratios: 1 Ω·cm = 1.11·10^{-12} units of CGS with dimension [s]; 1 K = k_B/h = 0.846·10^{11} with dimension [s^{-1}], where k_B is the Boltzmann constant, h is the Planck constant. As a result, limited to 3 terms in formula (3), we have ε≈0.33. Then from formula (1) we obtain for Zr1Nb the value of Q_{RZr1Nb} = 41.9 W m^{-1}.

Note that, according to the data of [12], Zr-2.5Nb alloy is characterized by an increase in ε from ~ 0.28 to ~ 0.6 with an increase in the thickness of the oxide layer to ~ 0.1 μm.

Considering that the linear radiation heat flow value is a constant in the case of constant power of the central heat source and using the measured surface temperature values T_s by formula (1) we obtain the estimate of emissivity for all the investigated coatings (see Table).

The corresponding values of the radiation heat transfer coefficient obtained by formula (2) are given in Table. In calculations the value of T_0 = 293 K (ambient temperature) was used.

Thermal and physical characteristics of the sample

Sample	Temperature of heating element surface T_{HE}, K	Temperature of sample outer surface T_s, K	Emissivity, ε	Radiation heat transfer coefficient α, W·m^{-2}·K^{-1}
Zr1Nb	633	541	0.20	3.64
Cr coating, 4.5 μm	633	586	0.14	3.04
CrN coating, 5 μm	633	500	0.28	4.29
Cu coating, 1 μm	643	612	0.12	2.87

From Table it follows that the highest value of the radiation heat transfer demonstrates the CrN coating (α=4.29 W·m^{-2}·K^{-1}), which is characterized by the highest emissivity.

So, the fuel elements with CrN coating having a high resistivity relatively to the zirconium-steam reaction [4] can provide an effective heat transfer due to the thermal radiation in the event of loss-of-coolant accident.

CONCLUSIONS

1. The method for studies on the radiation heat transfer from thin-wall cylindrical solids is developed.
2. Emissivity of fuel cladding samples with different coatings deposited using the vacuum-arc technique is investigated.
3. It is shown that the CrN coating, compared to the base Zr1Nb alloy and Cr coating can provide a more effective heat expulsion by radiation in the event of loss-of-coolant accident and high temperature rise. This occurs according to the Stefan-Boltzmann radiation law and due to a higher value of the total emissivity for CrN.

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ТЕПЛОФИЗИЧЕСКИЕ СВОЙСТВА ОБОЛОЧЕК ТВЭЛОВ С РАЗЛИЧНЫМИ ВАКУУМНО-ДУГОВЫМИ ПОКРЫТИЯМИ

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Используя разработанную методику определения излучательной способности тонкостенных цилиндрических образцов, исследовали теплофизические свойства в режиме теплопереноса излучением образцов оболочки твэла из сплава Zr1Nb с различными покрытиями, нанесенными вакуумно-дуговым методом. Показано, что покрытие CrN, по сравнению с базовым сплавом Zr1Nb и покрытием Cr, может обеспечить более эффективный съем тепла излучением в случае аварии с потерей теплоносителя и повышением температуры, что связывается с действием закона Стефана-Больцмана и более высоким значением интегральной степени черноты CrN.

ТЕПЛОФІЗИЧНІ ВЛАСТИВОСТІ ОБОЛОНОК ТВЕЛІВ З РІЗНИМИ ВАКУУМНО-ДУГОВИМИ ПОКРИТТЯМИ

В.А. Білоус, В.І. Соколенко, А.О. Чупіков, О.С. Купрін, О.П. Леденьов, В.Д. Овчаренко

З використанням розробленої методики визначення випромінювальної здатності тонкостінних циліндричних зразків досліджено теплофізичні властивості в режимі теплопереносу випромінюванням зразків оболонки твела зі сплаву Zr1Nb з різними покриттями, нанесеними вакуумно-дуговим методом. Показано, що покриття CrN, в порівнянні з базовим сплавом Zr1Nb і покриттям Cr, може забезпечити більш ефективне знімання тепла випромінюванням у разі аварії з втратою теплоносія і підвищенням температури, що пов'язується з дією закону Стефана-Больцмана і більш високим значенням інтегрального ступеня чорноти CrN.