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ASSESSMENT OF THE CORROSION RESISTANCE OF THE MAIN ALTERNATIVE MATERIALS FOR LIGHT WATER REACTORS TOLERANT FUEL ROD CLADDING

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Basic materials for nuclear fuel rod claddings (Zr+1%Nb and E110 alloys), as well as alternative materials for tolerant fuel rod claddings (Cr18Ni10Ti steel and 42CrNiMo alloy), that are able to maximally prevent the development of severe accidents at nuclear power plants were tested in the high-temperature water vapor environment. A comparative analysis of the corrosion resistance of these materials is presented, as well as the results of similar tests by the world's leading scientists. Samples of 42CrNiMo alloy revealed the highest corrosion resistance at high temperatures in a water vapor environment among the alternative materials for the fuel rod cladding considered in the study. The corrosion resistance of this alloy at a temperature of 1200 °C is approximately 40 times higher than that of Cr18Ni10Ti steel and E110 alloy. The high-temperature corrosion rate of the 42CrNiMo alloy is comparable to the corrosion rate of the Fechral alloy. The hydrogen that would be released during the oxidation of the 42CrNiMo alloy claddings would be almost forty times less compared to the zirconium alloy under the conditions of severe design accidents associated with overheating of the core.

INTRODUCTION

The Fukushima accident showed the special danger of the steam-zirconium reaction and gave impetus to R&D into the development of accident tolerant fuel (ATF). Ensuring the long-term stability of light water reactor fuel rod claddings under the design conditions and severe accidents is an important and urgent task, which is being solved by a large number of scientists from all over the world. One of the ways to ensure long-term stability, that is considered by the scientific community, is to replace the zirconium alloy for the fuel rod cladding with another, more corrosion-resistant material with a low neutron absorption cross section, which at high temperature oxidation will not result in the formation of a large amount of explosive hydrogen and the destruction of the cladding [1–3].

The ATF must be capable of operating both under normal conditions and under loss of coolant conditions according to the specification of the IAEA. Basing on this, it can be argued that one of the most important characteristics of the construction material of the fuel rod cladding is its corrosion resistance, which should maximally prevent the development of serious accidents with loss of coolant.

The advantages of steel in comparison with zirconium alloys are well-known: high corrosion resistance in steam-water coolant, absence of steam-zirconium reaction, experience of operation in various types of reactors, including light water reactors, and the availability of industrial technology for manufacturing thin-walled weld-free pipes and components.

At the beginning of atomic energy era in the first reactors of the Soviet design, steels Cr18Ni10Ti and Cr15NiMo3B were used as materials for fuel rod claddings. Austenitic steels and high-nickel alloys also were used in foreign reactors at the first stages. However, the major disadvantages of austenitic alloys, according to a number of authors, are stress corrosion cracking (SCC) as a result of radiation-induced depletion of grain boundaries by chromium [4] and a high thermal neutron capture cross section, which required either an increase in enrichment or a decrease in the wall thickness of the fuel rod cladding. Among the candidate materials for the fuel rod cladding are corrosion-resistant alloys based on iron – fechral alloys. SCC is not inherent to these materials, but radiation embrittlement is observed. At operating temperatures, the solid solution decomposes with the formation of a chromium-enriched α-phase, which is accelerated under irradiation [5].

At present, the development of ATF is a major concern in the nuclear research fields in the present time. The research concepts for enhanced ATF cladding development consists of Mo-Zr cladding [6], cladding coating [7, 8], iron-based alloy cladding [9, 10] and SiC cladding [11].

Chromium-nickel alloy 42CrNiMo is one of the promising alternative materials for fuel rod claddings. This alloy possesses high strength and plasticity, high corrosion resistance in various environments, high radiation resistance, as well as structural and mechanical stability under normal operating conditions of the WWER reactor. The advantage of the 42CrNiMo alloy is its long-term operation in the nuclear industry, where it has established itself as a reliable material for various nuclear power plants. This alloy is used for the manufacturing of fuel rod claddings of transport reactors, as well as RCCA rod claddings for WWER-1000 reactors.

There is no publicly available detailed description of the 42CrNiMo alloy corrosion resistance due to the specifics of its operation in transport nuclear power reactors. Its implementation as a material for the RCCA rod claddings, in contrast to the fuel rod claddings, does not require substantiation of its operability in a LOCA-type accidents.

The purpose of the study is to evaluate the corrosion resistance of the main alternative zirconium alloy fuel rod cladding materials in the water vapor environment for the selection of the optimal construction material for the fuel rod cladding of the tolerant fuel for light water reactors.

1. RESEARCH MATERIALS AND METHODS

Zirconium alloy E110 cladding tubes manufactured by TVEL LLC (Russia), that has been tested and certified for WWER power reactors, were used as the base material, to compare the obtained experimental results with the results of similar studies of other alternative materials.

The second base material were zirconium alloy Zr+1%Nb thin-walled weld-free tubes manufactured in Ukraine. The Zr+1%Nb alloy and the manufacturing technology of cladding tubes at the SE “Zirkoniy” [12] were developed to solve the problem of zirconium within the framework of the program to create a nuclear fuel cycle in Ukraine [13]. The alloy was obtained by the technology of calciumthermic reduction of zirconium from zirconium tetrafluoride and subsequent refining of this metal by electron beam melting [14]. The content of impurities and alloying element in alloy ingots after remelting meet the requirements of TU 95.166-98 for E110 alloy.

Thin-walled weld-free tubes manufactured of Cr18Ni10Ti stainless steel and chrome-nickel alloy 42CrNiMo were selected as alternative materials for the fuel rod cladding, that are similar in geometrical parameters to the parameters of the WWER reactor fuel rod cladding. The manufacturing of these tubes is currently mastered by the Ukrainian enterprises “OSKAR” LLC (Nikopol) and “Dniprovsky Special Pipe Plant” LLC (Dnipropetrovsk region). For many years, the enterprises have been supplying products of the 1st and 2nd safety classes for SE “NNEGC “Energoatom”. These products are implemented in the equipment of all 15 nuclear power units of Ukrainian NPPs, both in the core and in the 2nd and 3rd circuits.

The corrosion resistance of materials alternative to zirconium alloy was evaluated basing on the results of high-temperature tests at temperatures from 350 °C to 1200 °C in a water vapor environment in a tubular furnace at atmospheric pressure. The maximum isothermal exposure time was 1 h. Distilled water heated to boiling temperature was used to obtain steam. Before entering the tubular furnace, water vapor was additionally heated to a temperature of 250…300 °C, for that a certain section of the steam pipeline was equipped with additional heater. The length of the uniform temperature area in the furnace (working zone of the furnace) was 500 mm. The initial temperature of the sample was in the range from 100 °C to 300 °C. The heating rate of the sample from the initial temperature to 1000 °C was more than 20 °C per second (the temperature was reached in less than 35 s). The heating rate from 1000 to 1200 °C exceeded 2 °C per second (heating in less than 100 s).

2. RESEARCH RESULTS

2.1. HIGH-TEMPERATURE CORROSION KINETICS OF ZIRCONIUM ALLOYS

Corrosion tests of fuel rod cladding samples manufactured of zirconium alloy E110 and Ukrainian alloy Zr+1%Nb are being carried out for a long time in the NFC STE of the NSC KIPT. A large number of articles are been devoted to the study of the high-temperature corrosion kinetics of these alloys [15, 16]. The results of the study of the mass change kinetics of E110 and Zr+1%Nb alloy samples in stationary tests in a water vapor environment are given in articles [17–22]. This article summarizes the results of corrosion tests of samples of the Ukrainian alloy Zr+1%Nb at temperatures from 660 to 1200 °C when exposure time from 1 to 9 h and compares them with the results of the E110 alloy corrosion tests (Fig. 1).

The dependence of the average corrosion rate during one hour on the test temperature of samples of both zirconium alloys (E110 and Ukrainian Zr+1%Nb) in a water vapor environment (Fig. 2) showed a low rate of mass increase up to a temperature of 770 °C, that was ~ 114 mg/(dm^2·h). With an increase in the test temperature, the mass gain rate of the samples increases sharply and at a temperature of 1200 °C in 1 h test it was 3600 mg/dm^2 for the E110 alloy and 3230 mg/dm^2 for the Zr+1%Nb alloy. That is, the high-temperature corrosion rate of the Ukrainian Zr+1%Nb alloy was somewhat lower than the E110 alloy corrosion rate.

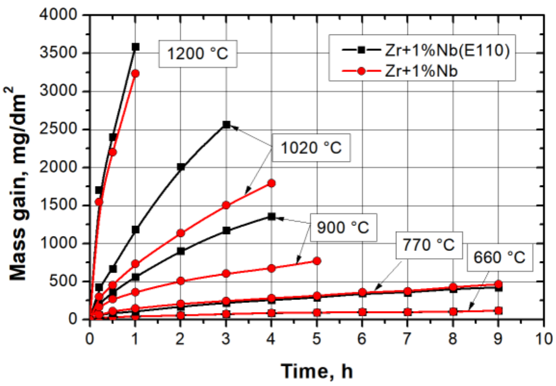


Fig. 1. Corrosion kinetics of zirconium alloy samples in a water vapor environment

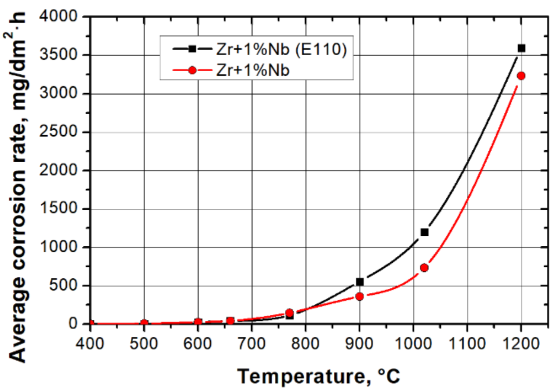


Fig. 2. The dependence of the zirconium alloy samples average corrosion rate on the test temperature in a water vapor environment

The corrosion tests results at high temperatures could vary slightly depending on the rate at which the sample is heated to a given temperature and the number of thermal cycles the sample subjected before the desired exposure time was reached. The mass change was even higher and reached 4100 mg/dm^2 for both alloys at periodical weighing (after 5, 10, 15, and 60 min of exposure) the same set of samples.

The appearance of the samples during the test is summarized in (Fig. 3). The appearance of E110 and Zr+1%Nb alloys samples was identical after all tests.

Fig. 3. Appearance of zirconium alloys samples after tests in a water vapor environment

2.2. HIGH-TEMPERATURE CORROSION KINETICS OF 08Cr18Ni10Ti STEEL

Studying of the corrosion resistance of 08Cr18Ni10Ti steel at high temperatures in a water vapor environment has been started relatively recently at the NFC STE NSC KIPT [23, 24]. A much larger number of works was previously devoted to studying the corrosion kinetics of steels grades 06Cr18Ni10Ti, 08Cr18Ni10Ti, and 12Cr18Ni10Ti in model environments at the parameters of the WWER-1000 primary coolant. Interest in this study was initiated by the search for alternative materials for nuclear fuel rod claddings resistant to accidents [25–29].

Corrosion tests of 08Cr18Ni10Ti steel samples were carried out in a water vapor environment for 1 h in the temperature range from 350 to 1200 °C as part of this study. Increasing the time of isothermal exposure results in a mass gain of samples for all temperatures. The corrosion kinetics of 08Cr18Ni10Ti steel samples at 1 h testing in a water vapor environment in the temperature range from 350 to 1200 °C is decaying (Fig. 4), i.e. the curve can be characterized as a power-law dependence.

The dependence of the average corrosion rate for 1 h on the test temperature of 08Cr18Ni10Ti steel samples in water vapor (Fig. 5) reveals a low rate of mass gain for the samples up to a temperature of 800 °C. At the same time, the mass gain does not exceed 100 mg/dm^2, a sharp mass gain is observed at a temperature above 800 °C. Mass gain for samples that were exposed for 1 h at a temperature of 1200 °C was almost 4000 mg/dm^2.

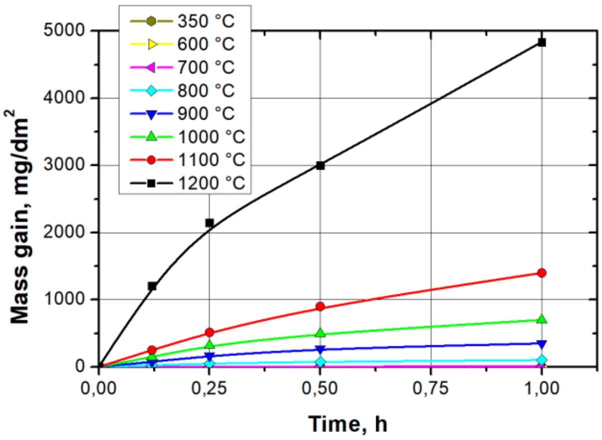


Fig. 4. Corrosion kinetics of 08Cr18Ni10Ti steel samples in a water vapor environment

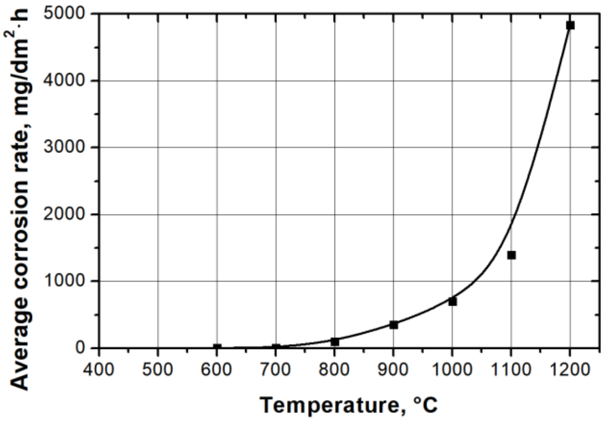


Fig. 5. Dependence of the average corrosion rate of 08Cr18Ni10Ti steel samples on the test temperature in a water vapor environment

700 °C 900 °C 1000 °C 1100 °C 1200 °C

Fig. 6. Appearance of 08Cr18Ni10Ti steel samples after testing in a water vapor environment

The appearance of the surface changed during the test from dark gray color over gray and brown to gray color with a metallic luster after test at a temperature of 1200 °C (Fig. 6).

2.3. HIGH-TEMPERATURE CORROSION KINETICS OF THE 42CrNiMo ALLOY

The corrosion tests results of 42CrNiMo samples in a water vapor environment for 1 hour in the temperature range from 350 to 1200 °C revealed that the corrosion kinetics of this alloy is decaying (Fig. 7). This indicates to the protective properties of the oxide film, formed during corrosion testing in a water vapor environment.

The rate of mass gain increased with increasing the test temperature, as evidenced by the dependence of the average corrosion rate during 1 h on the test temperature (Fig. 8). A characteristic feature of the corrosion resistance of the 42CrNiMo alloy samples in the water vapor environment is that the corrosion rate is much lower compared to the Zr+1Nb alloy and the 08Cr18Ni10Ti steel. The dependence curve of the average corrosion rate of the 42CrNiMo samples alloy is similar to the curves for the Zr+1Nb alloy and the 08Cr18Ni10Ti steel. A sharp mass gain of the samples was observed at a temperature above 800 °C, and at a temperature of 1200 °C in 1 h of testing it was 120 mg/dm^2.

The appearance of the surface changed from brown over black and dark green to gray color at the end of the test at a temperature of 1200 °C (Fig. 9).

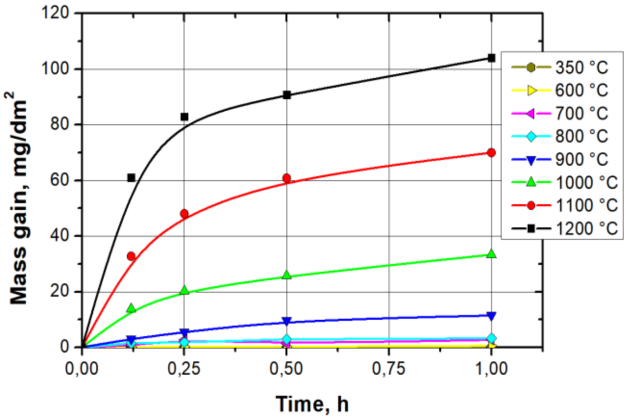


Fig. 7. Corrosion kinetics of the 42CrNiMo alloy samples in a water vapor environment

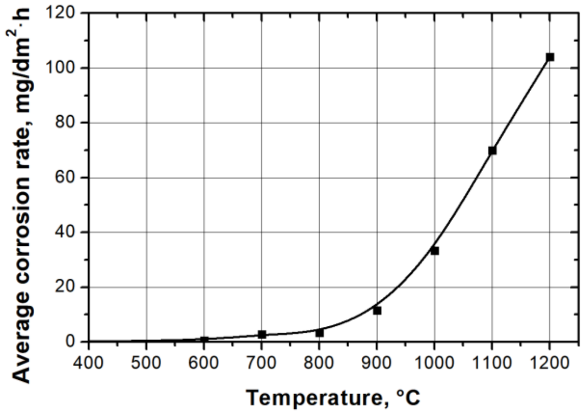
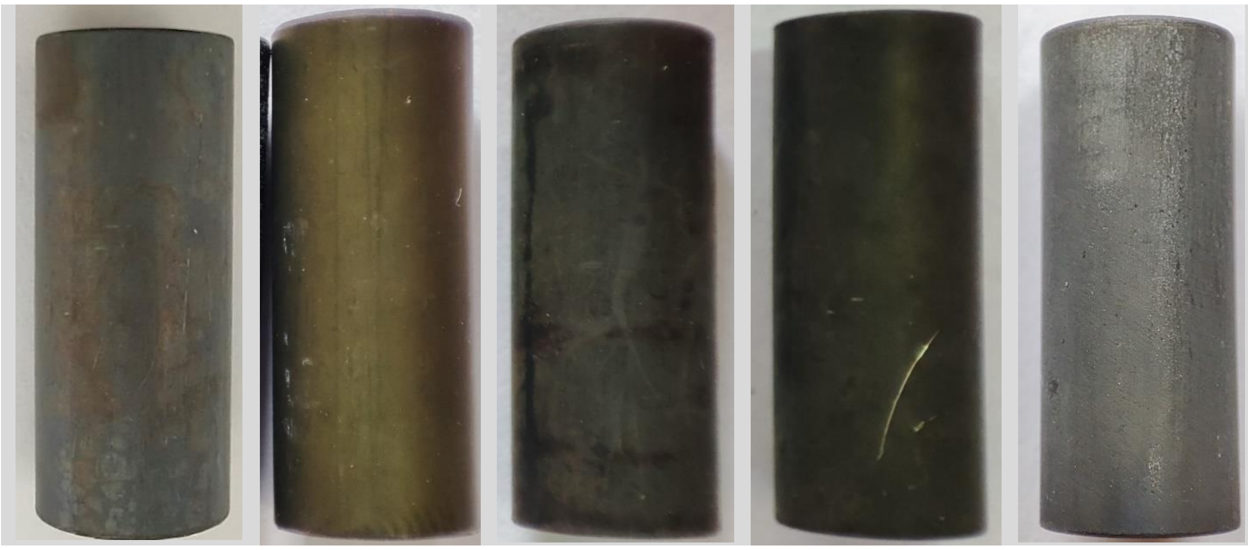


Fig. 8. Dependence of the average corrosion rate of 42CrNiMo alloy samples in 1 h on the test temperature in a water vapor environment



700 °C 900 °C 1000 °C 1100 °C 1200 °C

Fig. 9. Appearance of 42CrNiMo alloy samples after testing in a water vapor environment

3. DISCUSSION OF RESULTS

The paper presents the research results of corrosion resistance in the water vapor environment of the basic materials for WWER nuclear fuel rod claddings and alternative materials of tolerant fuel rod claddings. Austenitic stainless steel Cr18Ni10Ti and chrome-nickel alloy 42CrNiMo were tested as alternative materials. These materials are able to maximally prevent the development of severe accidents at nuclear power plants according to the world community [1–3], and are produced at Ukrainian enterprises.

The kinetics of the mass change of tube samples manufactured of all studied materials is decaying (Figs. 1, 4, 7) and can be characterized by a power-law dependence, that indicates the protective properties of oxide films that are formed under high-temperature corrosion testing in a water vapor environment.

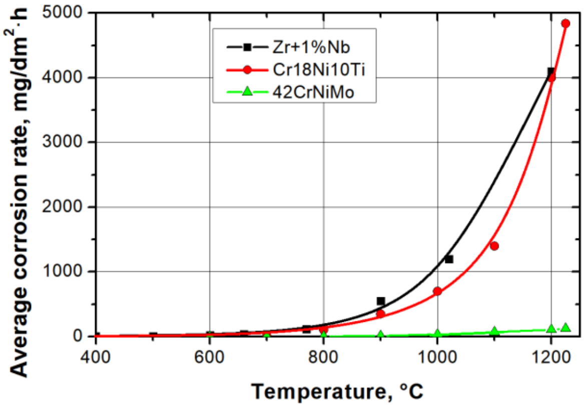


Fig. 10. Dependence of the average corrosion rate of Zr+1%Nb, Cr18Ni10Ti and 42CrNiMo samples on the test temperature in a water vapor environment

High-temperature corrosion tests of tube samples manufactured of 08Cr18Ni10Ti steel revealed higher corrosion resistance, but up to temperatures of 1100 °C. The corrosion rate increased at higher temperatures, and at a temperature of 1200 °C it was comparable to the corrosion rate of zirconium alloy E110 (Fig. 10), and the mass gain was almost 4100 mg/dm^2 after exposure for about 1 h.

The high-temperature corrosion rate of the 42CrNiMo alloy samples was significantly lower than that of the E110 alloy and stainless steel Cr18Ni10Ti (see Fig. 10). Products manufactured of 42CrNiMo alloy revealed high corrosion resistance even at a temperature of 1200 °C. The corrosion resistance of the 42CrNiMo in a water vapor environment at a temperature of 1200 °C was approximately 40 times higher than that of the Cr18Ni10Ti steel and the E110 alloy (see Fig. 10). There is lack of information on the corrosion resistance of the 42CrNiMo alloy under core operating conditions due to the specifics of this alloy implementation in nuclear power reactors. In paper [30] it is stated that the difference in the amount of material that has turned into an oxide can reach 90 times. But the paper does not specify the parameters of corrosion tests that can affect the oxidation kinetics, such as the heating rate, the corrosion environment (water vapor or air), the state of the samples surface (mechanical, chemical, electrochemical treatment). But the comparative data obtained in this study do not contradict the literature data given in [30] and confirm the significantly higher corrosion resistance of the 42CrNiMo alloy compared to the E110 alloy.

The question arises as to how much the corrosion resistance of 42CrNiMo alloy tubes is higher compared to other metal materials suitable for high-temperature operation. High-temperature tests of Fechral alloy samples were carried out for comparison, despite the fact that this material was not in the form of tubes and that it was not implemented in nuclear power industry at all. Fechral alloy is the material for the manufacture of heating elements of powerful electronic heating devices and operates at temperatures up to 1400 °C due to its high corrosion resistance in air at high temperature.

Fig. 11 shows the dependence of the average corrosion rate for 1 h of the 42CrNiMo alloy tube samples and industrially manufactured Fechral alloy plates on the test temperature in a water vapor environment. The high-temperature corrosion rate of the 42CrNiMo alloy was comparable to the corrosion rate of the Fechral alloy basing on the obtained results. The mass gain of both alloys was 100...105 mg/dm^2.

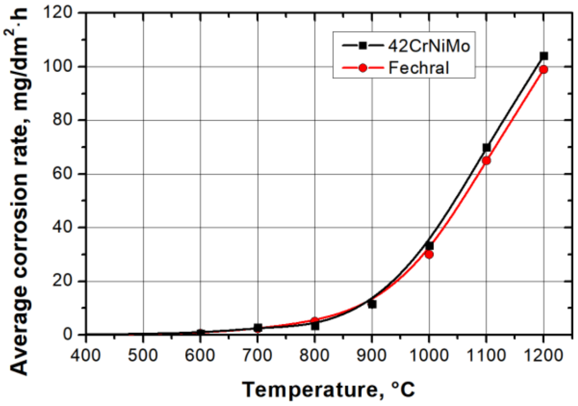


Fig. 11. Dependence of the average corrosion rate of the 42CrNiMo and Fechral alloy samples on the test temperature in a water vapor environment

This indicates that 42CrNiMo alloy tubes reveal the highest corrosion resistance among metal materials, manufactured in Ukraine. Therefore, this alloy can be recommended for implementation as an alternative material for rod claddings of tolerant fuel. But it should be noted that the justification of this material operation in the core of a light water reactor has great difficulties, which are associated with a sharp change in mechanical characteristics at high temperature and significant parasitic capture of thermal neutrons (~ 21%). In order to reduce the parasitic capture of thermal neutrons, adaptation of claddings manufactured of this alloy for operation in WWER is required, and consists in reducing the wall thickness of the tube, if it does not result in a loss of mechanical stability.

CONCLUSIONS

1. Basic materials for nuclear fuel rod claddings (Zr+1%Nb and E110 alloys), as well as alternative materials for tolerant fuel rod claddings (Cr18Ni10Ti steel and 42CrNiMo alloy), that are able to maximally prevent the development of severe accidents at nuclear power plants were tested in the high-temperature water vapor environment. A comparative analysis of the corrosion resistance of these materials is presented, as well as the results of similar tests by the world's leading scientists.

2. An increase in the corrosion rate of zirconium alloys of both brands at a test temperature above 700...750 °C in a water vapor environment was observed. The high-temperature oxidation kinetics of the Ukrainian Zr+1%Nb alloy and the E110 alloy have a similar character, with a difference of up to 40% in the temperature range of 900…1000 °C, probably related to the higher oxygen content in the Zr+1%Nb alloy. Testing at a temperature of 1200 °C resulted in a mass gain of both zirconium alloys, that was almost 4100 mg/dm^2 after 1 h exposure.

3. The mass change kinetics of 08Cr18Ni10Ti steel samples is decaying and can be characterized by a power-law dependence, which indicates the protective properties of oxide films. The dependence of the average corrosion rate in 1 h on the test temperature of the steel samples in water vapor revealed a low rate of mass gain up to a temperature of 700 °C. A sharp mass gain was observed at a temperature above 800 °C, and after 1 hour exposure at a temperature of 1200 °C it was almost 4000 mg/dm^2.

4. The mass change kinetics of chromium-nickel alloy 42CrNiMo samples is decaying and characterized by a power-law dependence. A sharp mass gain rate with an increase in the test temperature to 1200 °C was not observed. After 1 h exposure at a temperature of 1200 °C the mass gain was only 105 mg/dm^2.

5. Samples of 42CrNiMo alloy revealed the highest corrosion resistance at high temperatures in a water vapor environment among the alternative materials for the fuel rod cladding considered in the study. The corrosion resistance of this alloy at a temperature of 1200 °C is approximately 40 times higher than that of Cr18Ni10Ti steel and E110 alloy. The high-temperature corrosion rate of the 42CrNiMo alloy is comparable to the corrosion rate of the Fechral alloy. The hydrogen that would be released during the oxidation of the 42CrNiMo alloy claddings would be almost forty times less compared to the zirconium alloy under the conditions of severe design accidents associated with overheating of the core.

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ОЦІНКА КОРОЗІЙНОЇ СТІЙКОСТІ ОСНОВНИХ АЛЬТЕРНАТИВНИХ МАТЕРІАЛІВ ОБОЛОНКИ ТОЛЕРАНТНОГО ПАЛИВА ЛЕГКОВОДНИХ РЕАКТОРІВ

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Проведено високотемпературні дослідження в середовищі водяної пари базових матеріалів оболонок ядерного палива (сплави Zr+1%Nb та Е110), а також альтернативних матеріалів оболонок толерантного палива (сталі Х18Н10Т та 42ХНМ), які здатні максимально перешкоджати розвитку важких аварій на АЕС. Представлено порівняльний аналіз корозійної стійкості цих матеріалів, а також результати подібних випробувань світових провідних вчених. Із розглянутих у роботі альтернативних матеріалів оболонки твел найбільш високу корозійну стійкість при високих температурах у середовищі водяної пари показали зразки сплаву 42ХНМ. Корозійна стійкість цього сплаву при температурі 1200 °С приблизно в 40 разів вища, ніж сталі Х18Н10Т та сплаву Е110. Швидкість високотемпературної корозії сплаву 42ХНМ співставна зі швидкістю корозії сплаву фехраль. В умовах максимальних проектних аварій, пов'язаних з перегрівом активної зони, кількість водню, який виділиться при окисненні оболонок, виготовлених зі сплаву 42ХНМ, буде майже в 40 разів менше в порівнянні з цирконієвим сплавом.