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The interaction between nitride uranium and stainless steel

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Abstract. Uranium nitride is most popular nuclear fuel for Fast Breeder Reactor New Generation. In-pile experiments at reactor BOR-60 was shown an interaction between nitride fuel and stainless steel in the range of 8-11% burn up (HA). In order to investigate this interaction has been done diffusion tests of 200 h and has been shown that the reaction occurs in the temperature range 1000-1100 ° C. UN interacted with steel in case of high pollution oxygen (1000-2000 ppm). Also has been shown to increase interaction UN with EP-823 steel in the presence of cesium. In this case the interaction layer had a thickness about 2-3 μm. Has been shown minimal interaction with new ODS steel EP-450. The interaction layer had a thickness less then 2 μm. Did not reveal the influence of tellurium and iodine increased interaction. It was show compatibility at 1000 °C between UN and EP-450 ODS steel, chrome steel, alloying aluminium and silicium.

1. Introduction

Currently, one of the directions of development of nuclear power is a widespread introduction of fast breeder reactors. At the same time as the fuel is considered to use a nitride mixed uranium-plutonium fuel, which has high thermophysical and radiation properties [1]. As construction materials (fuel cladding and fuel assemblies) are treated corrosion-resistant steel (CRS) different classes of EP-823, EP-450/450 ODS, EK-164, etc. The most important aspect of the nuclear fuel is its compatibility with structural materials in a range of operating temperatures characteristic for fast neutron reactors, ie 600-1000 °C. For a long time it was believed that the interaction of nitride fuel and steel shell, up to 1000 °C is absent, and the main cause of damage to the fuel element is associated with swelling of the fuel, offensive of thermomechanical contact with the cladding and damage the latter [2-4]. However, the test reactor in recent years of fuel assemblies with nitride fuel in the experiment Bora-Bora, and their subsequent postirradiation research in "hot" cells using the methods of electro-probe microanalysis showed the presence of the interaction with the fuel cladding.

2. Experimental method

To obtain preliminary data on the interaction of the fuel-cladding was used method manufacturing composite ie co-pressing and sintering powders Mononitride uranium and corrosion-resistant steel. UN powder obtained by the hydrogenation-dehydrogenation-nitriding has a particle size of 10-20 micrometers, and the powder of corrosion-resistant steel, obtained by mechanical grinding of ingot and subsequent milling with a ball mill into powder particles had a size range of 20-30 micrometers. Subsequently, the powders UN and corrosion-resistant steel are mixed in a ratio of 20:80 or 30:70, respectively. Sample compacts in the form of plates and sintered at 1000 °C in vacuum furnace for 5 hours. Due to the large volume of content steel powder provided its sintering, ie the sample is a dispersion composition, wherein as the dispersed phase acts mononitride uranium. The significant disadvantages of this method is the impossibility of introducing chemically active elements (cesium, iodine and tellurium), since through the pass-through porosity said elements vaporized in the annealing process.

In this regard, was developed a method to test the compatibility of the powder of nuclear fuel with cladding material, consisting in the fact that the preliminary from the cladding-material is made crucible with a lid, wherein the inner surface of the crucible is polished to roughness values of Ra = 0.16-0.125 mm, and the lid is made in the form of a truncated cone, after which the crucible filled with the powder of the test fuel, then added simulators chemically active fission products and sealing of the crucible is carried out in inert gas atmosphere, followed by annealing in the temperature range 600-1000 °C.

After a series of diffusion annealing conducted metallographic preparation of the samples to determine the magnitude of the interaction layer. Initially, all the samples (crucibles) were cut into two halves along the cut portion the generatrix part of the diameter of the crucible. Cuts on the diamond saw Buehler IsoMet, as well as the coolant used isopropyl alcohol (IPA), which reduces oxidation of uranium nitride. Metallographic preparation of thin sections was performed according to standard procedures, a grinding carried out on sandpaper with fineness 1200 and then polished on a cloth alternately with diamond paste of size 10, 3 and 1 micrometers. To prevent oxidation, in all cases using IPA.

Investigation of the microstructure of diffusion couples UN / SS carried by the scanning electron microscope JEOL 6610 LV. As a further method for determining the thicker layer of interaction used method of measuring microhardness. The measurement was performed by pressing a diamond tetrahedral pyramid with an apex angle of 136°, and then determines the size of the diagonal and perform calculations microhardness. The purpose of this work is determined by the values of the microhardness near the border "nitride - CRS", as well as an array of CRS. Any deviation microhardness of the border array clearly indicates diffusion processes or the formation of intermetallic compounds. Measurements were performed at a load of 100 g, loading time was 10 s.

3. Experimental results

3.1. Features of interaction between the UN and the CRS

Analysis of the sintered powders UN and CRS, implemented by the X-ray phase analysis, is highly sensitive and allows to detect interactions at the initial stage. According to the results presented above, we can conclude that there is no interaction up to 1000 °C and only at exposure 200 hours were found traces of interaction products. According to test results, may be noted the good compatibility of the steel EP-823 and EP-450 ODS, also need to pay attention to chromium steels, alloyed with aluminum. Table 1 shows the results of X-ray phase analysis of powder compacts. From the data in the table it can be seen that during the annealing at 1000 °C compacts powders mononitride uranium and CRS begin to appear traces of oxide and nitride phases (Fe_3N, Fe_2O_3, (Fe, Mo) $_3O_4$) and ternary phase U_2CrN_3, indicating that beginning of the reaction. With regard to the diffusion couples with steel EP-450 it is possible to note the presence of the interaction, as evidenced by the presence of the triple phase U_2CrN_3. Otherwise, there is a presence of mononitride uranium ferrite and uranium dioxide. It is also considered the interaction with uranium nitride dispersion-strengthened steel EP-450 ODS.

Analysis of the diffraction pattern shows a lower level of interaction, the presence of triple-phase have not been identified, apparently this could be explained by the presence of particles of yttrium oxide that is effective diffusion barrier. Steel EP-823 also interacts with uranium nitride to form iron nitride, positive aspects may include less, compared with the EP450, peak height, what indicating a very weak interaction. However, in this case was found the traces of oxide and iron nitride.

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Table 1. Results of X-ray phase analysis of diffusion couples «UN-CRS» manufactured from a mixture of powders.

Labelling	Sample status	Detected phases	Extras. phases
UN-EP450	Sintered	UN, α-Fe, UO_2, U_2CrN_3	
UN-EP450 ODS	Sintered	UN, α-Fe, Y_2O_3, UO_2	tracks: $(Fe,Mo)_3O_4$
UN-EP823	Sintered	UN, α-Fe, UO_2	tracks Fe_3N and Fe_2O_3
UN-H18N10T	Sintered	UN, α-Fe, UO_2, U_2CrN_3, γ-(Fe,Ni)	
UN-(Fe, Cr, Al)	Sintered	UN, α-Fe, UO_2, U_2CrN_3	tracks U_2N_3

Austenitic steels X18H10T also interacts with nitride fuel to form a ternary phase. The powder compacts UN- (Fe, Cr, Al) traces ternary phase, but its amount is small. Also found a number of one and a half nitride, the appearance of which can be attributed to exchange reactions between the aluminum nitride. A similar situation exists for steels alloyed with silicium, but in this case no phase one and a half nitride. This was followed by an analysis of the microstructure and MXSA samples.

3.2. The influence of oxygen on the interaction between the UN and CRS

One reason for the strengthening of interaction between the UN and the steel can be increased content of oxygen in mononitrides uranium. To verify this fact made UN powders with different oxygen content: № 1 - Powder UN, containing ~ 100-200 ppm oxygen; №2 - Powder UN, containing ~ 1000-2000 ppm oxygen. Each of these powders was charged in a steel crucible EP-450, EP-823, as well as chrome steel crucibles with additions of silicium and aluminum.

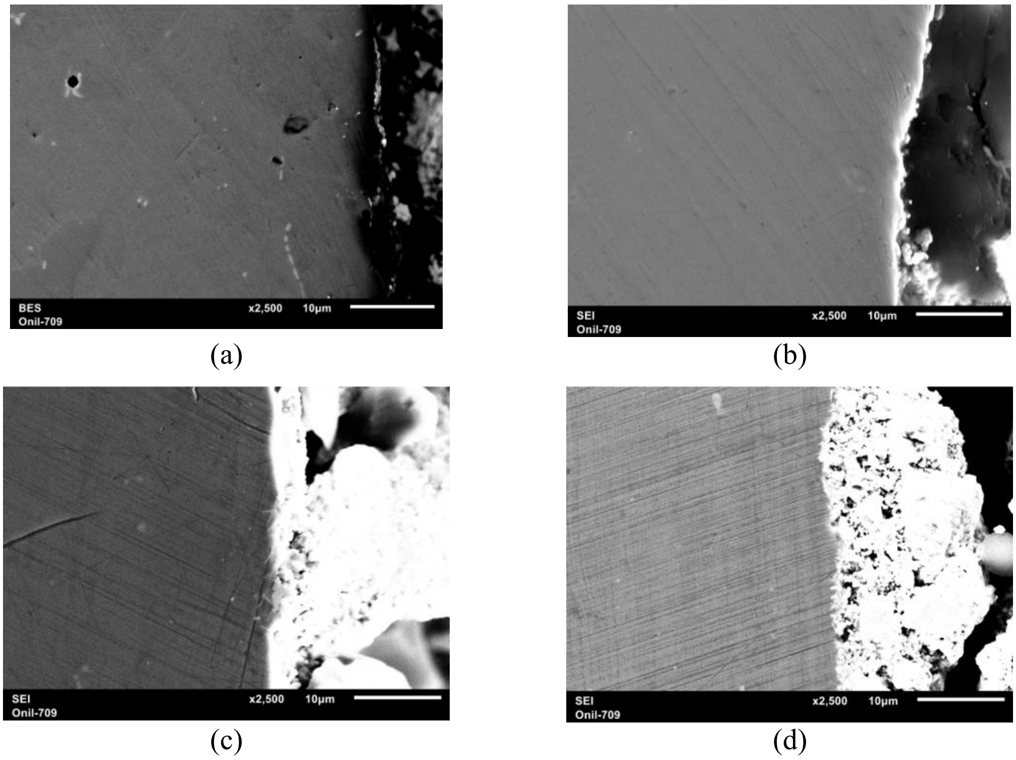
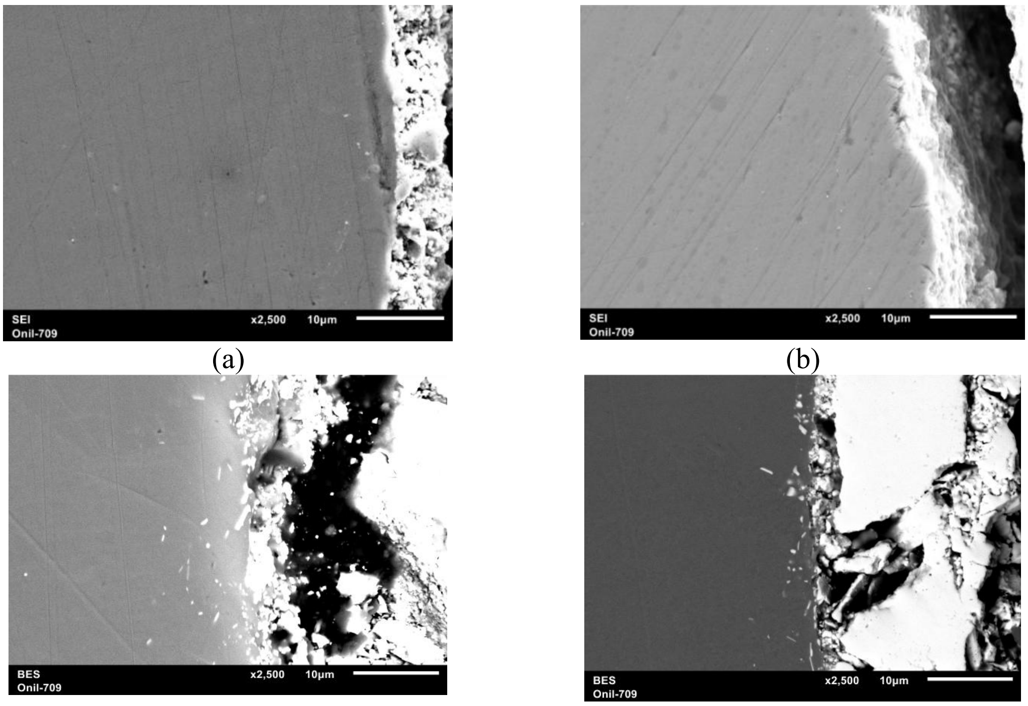


Figure 1. (a) The microstructure of the the interaction layer diffusion couples UN-EP450, the oxygen content of 100 ppm; (b) diffusion couples UN-EP450, the oxygen content of 1000 ppm; (c) UN (100)-EP823; (d) UN (1000)-EP823.

Metallographic analysis of diffusion couples UN (100 ppm) with steel EP-450 allow note the very small width of the interaction zone (not more than 2 micrometers), as well as the emergence of low porosity. This fact is illustrated in figure 1 (a, b). A similar situation exists for the steel EP-823, on the border observed porosity what, apparently, due to the Kirkendall effect. Also not found significant difference for diffusion couples nitride-EP823 with different oxygen content (100 and 1000 ppm), the width of the interaction zone differs slightly and varies in the range of 2-5 micrometers. This fact can be explained by the low partial pressure of oxygen (over the oxide phase) as shown in figure 1 (c, d).

3.3. Influence of chemically active fission products on interaction between the UN and the CRS

Accelerate the interaction may also presence of corrosive fission products (cesium, iodine and tellurium). In this paper, added a chemically-active products in an amount equivalent to 10.2% of HA burnout (Cs 12.017, I 0.899, Te 1.661 g / kg of irradiated fuel), which is due to the fact that this value is the threshold at which there is damage to the cladding. These elements were introduced into the box with an inert atmosphere, due to the high activity of cesium and tellurium toxicity. The uranium nitride powder poured alternately cesium, iodine and tellurium, mixed in an agate mortar and pressed into the crucible of the investigated steel. In this work, mainly conducted corrosion tests have become EP823 in the presence of cesium, iodine and tellurium, due to its high potential as a fuel rod cladding material. Photograph of the microstructure shown in figure 2 a and b (the oxygen content was 1000 ppm). From the analysis of the microstructure, it follows that the interaction layer has a width of about 5-9 micrometers, which is somewhat higher than in the case of a pure nitride or in case of couples diffusion test without CAPD. Also, with CAFP were tested the samples chromium steel (oxygen content was 1000 ppm), doped with 2 wt. % of aluminum and silicium, which is shown in figure 2 c, and d.



(a) (b) (c) (d)

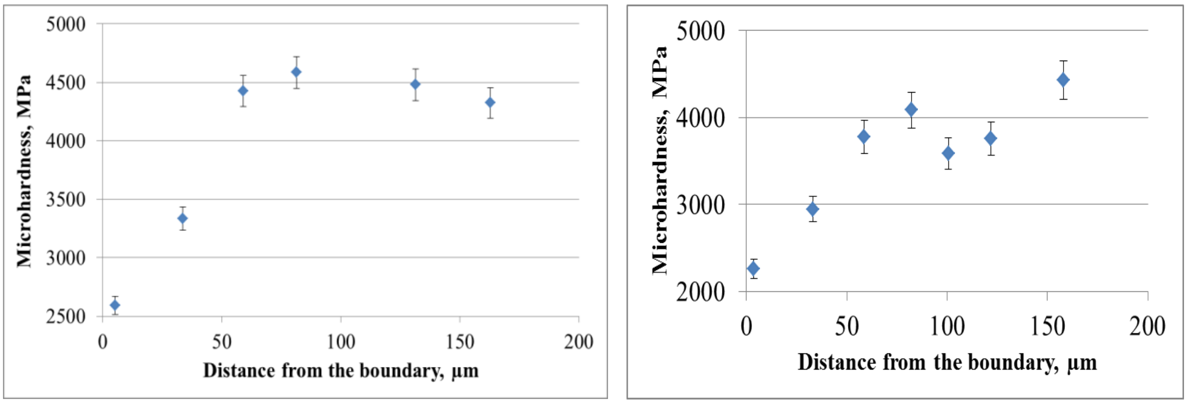
Figure 2. (a) The microstructure of the the interaction layer diffusion couples UN-EP823 with oxygen content 1000 ppm; (b) diffusion couples UN-EP823; (c) diffusion couples UN with FeCr-2% Al; (d) diffusion couples UN with FeCr-2% Si.

In this case, the interaction is almost undetectable in some cases there is a weak interaction, wherein the the interaction layer thickness was not greater than 1 micrometers. Note that such a value is very low, even with the error and resolution scanning microscope one can speak about a significant reduction in the interaction layer for steels alloyed with aluminum and silicium, even in the presence of CAFP.

Based on the concepts of a parabolic law of growth of the layer, the parabolic growth constants. The constants for similar compositions UN (100) + EP450, UN (100,1000) + EP823, UN (1000) + 2% FeCr-Al, of the order of 10-17 m^2/s. For the composition of UN (1000) + EP823 + CAFP there is an increase the kinetics of the interaction with the UN on the order of CRS. The activation energy of the interaction is not possible due to lack of cooperation with UN CRS at 600 °C.

3.4. Evaluation microhardness of diffusion couples

Studies microhardness of diffusion couples UN-CRS allows us to conclude that the microhardness of areas CRS near the border and in the array varies. Basically microhardness boundaries is 10-15% lower compared to the array, which can be explained by the weak interaction CRS with nitride fuel microhardness decrease due to the appearance of porosity and intergranular corrosion. Also, from the obtained data it is clear that such samples UN (1000 ppm O2) + FeCr-2% Al and UN (1000 ppm O2) + FeCr-2% Si difference between microhardness array and the boundary is within measurement error, which may be indicative the lack of interaction. Strong distinction between an array of micro-hardness and abroad observed for samples UN (1000) -EP823 and UN (1000) -EP823-Cs. Microhardness of distance samples UN (1000) + EP823 and UN-EP823-Cs is shown in figure 3.



a b

Figure 3. (a) Microhardness diffusion couples UN (1000) - EP 823; (b) diffusion couples UN-EP823-Cs.

From these dependencies can be seen that the graphs of microhardness similar, which suggests about the similarity of corrosion damage to the contact area in the presence of high oxygen content and in the presence of cesium.

4. Conclusions

Developed two methods of diffusion test compatibility nitride fuel and CRS. One of which is annealed and pressed powders UN CRS followed by X-ray phase analysis. It is shown that the time of occurrence of nitrides or triple phase clearly indicates the beginning of the interaction. Another technique is based on the study of the filling of the crucible steel powder with mononitride uranium, the process allows you to enter chemically-active fission products, as well as uniquely determine the value of the interaction layer.

According to the results of diffusion tests for 200 hours showed no interaction at a temperature of 600 ° C nitride fuel with the CRS. At the same time shows the appearance of the interaction layer thickness of 2-3 micrometers in the temperature range 1000-1100 ° C for 200 hours.

Noted increased interaction and thus increase the interaction layer to 5-7 micrometers in the case of tests Mononitride with increased oxygen content (1000-2000 ppm). During the diffusion test steels EP-450 and 823 and 18Cr10NiTi shown appearance iron nitride Fe3N phase and ternary phase U2CrN3, the presence of which is uniquely enabled to determine the beginning of interaction. It showed good compatibility of the steel EP-450 ODS and chromium steel, alloy aluminum and silicium at 1000 ° C. Analysis of the boundaries of the "nitride - steel", as well as microhardness revealed no occurrence of the interaction layer.

Shown increased interaction UN and steel EP823 in the presence of cesium, in this case, the interaction layer was 15 micrometers. There was no impact of tellurium and iodine to enhance interaction.

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