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BURNUP AND RADIATION EMBRITTLEMENT OF THE U-Mo NEUTRON SOURCE TARGET

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The burnup of uranium target plates of the NSC KIPT neutron source was studied. An analysis of experimental work on the effect of neutron irradiation on the strength and plastic properties of uranium and U-Mo alloys under conditions close to the operation of the target has been carried out. A description of the processes of radiation embrittlement is presented, taking into account the deformation and porosity of materials at various levels of burnup. An estimate of the expected service life of a uranium target under irradiation has been carried out.

INTRODUCTION

Recently, at the NSC KIPT, together with the Argonne National Laboratory of the USA, the physical launch of the research nuclear installation “Neutron source based on a subcritical assembly controlled by an electron accelerator” was carried out as a stage in the creation of new generation nuclear power plants [1].

Two types of neutron producing targets (NPT) are being considered for use: depleted uranium (U-Mo) based alloy and tungsten. Previously, an analysis was made of nuclear-physical processes occurring in uranium and tungsten targets under irradiation [2, 3]. It was found that the greatest contribution to the rate of damage formation in a tungsten target is made by the elastic interaction of high-energy electrons with nuclei, and the maximum rate of dose accumulation is about 0.83 dpa/year and is achieved in the second plate of the tungsten target (at a depth of ~ 1 cm). An analysis of the nuclear-physical processes taking place in a uranium target under irradiation has shown that the greatest contribution to the damage rate is made by uranium photofission fragments, as a result of which the maximum defect creation rate will be 100 dpa/year.

The physical and mechanical properties, and hence the service life of a uranium target, directly depend on the level of uranium burnup, the value of which can have a great effect on the structure, radiation growth, radiation embrittlement, and other service characteristics of the target. Burnup, i.e. the relative change in the concentration of uranium atoms due to fission processes will be $0.55\ 10^{-3}$ per year, which approximately corresponds to the values in thermal reactors of the WWER type [2].

The purpose of this work was to conduct a comparative analysis of the strength and plastic characteristics (radiation embrittlement and hardening) of uranium and U-(7…9%) Mo alloys, as the material of the target plates of the NSC KIPT under different level of burnup.

Interest in the radiation resistance of these alloys is also due to the fact that they are today one of the main candidates for the material of monolithic nuclear fuel for experimental and research reactors of a new generation [4].

1. BURNUP OF THE URANIUM TARGET OF A NEUTRON SOURCE

The studies of nuclear-physical processes occurring in uranium and tungsten targets under electron irradiation and leading to radiation defect formation and burnup are also the subject of [2, 3] and earlier works on mathematical modeling. It was shown that, at a depth of about 1 cm an electromagnetic shower develops in the target, which causes photofission of U-238 nuclei, while the fission fragments create a large number of radiation defects in the uranium target. It was found that the highest level of burnup is achieved at a depth of ~ 1.4 cm and is $5.5\ 10^{-2}$ % per year of continuous operation (Fig. 1). In this case, the maximum rate of creation of radiation defects in uranium reaches 100 dpa/year.

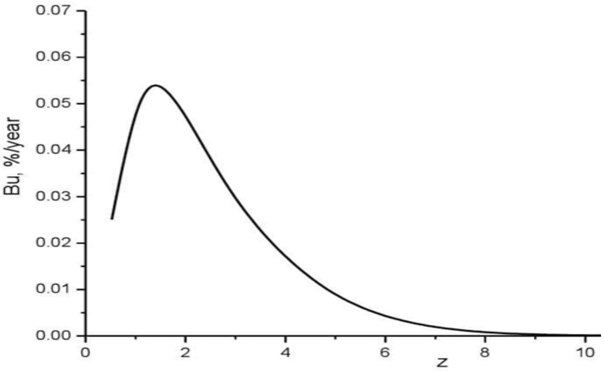

Fig. 1. Burnup rate U along the length of the neutron source target; z – depth in cm

Uranium burnup is accompanied by the release of solid and gaseous fission products and the production of helium, while Xe and Kr dominate over other fission products. The rate of their formation is very high, and is about 0.2…0.3 atoms per fission. With an annual burnup level of $5.5\ 10^{-2}$ %, the helium concentration in the uranium target will be about 1500 appm. These processes lead to the fact that gas nanobubbles with high internal pressure can form in the uranium-molybdenum target even at burnup of the order of one percent [5].

When analyzing the experimental work performed on samples for mechanical testing irradiated in reactors, we use the following ratios between the values a dose of

1 dpa corresponds to: 1...2 10^{-4} % uranium burnup, and a neutron fluency of about $2 \cdot 10^{17}$ n/cm [2, 6, 7].

2. COMPARATIVE ANALYSIS OF DATA ON RADIATION EMBRITTLEMENT AND HARDENING OF URANIUM AND URANIUM-MOLYBDENUM ALLO1YS

The results of analysis of the strain-to-fracture dependence during tensile tests of natural uranium specimens and U-(7...9%)Mo alloy specimens irradiated in reactors are shown in Fig. 2,a. As follows from the figure, the value of deformation before destruction of all irradiated samples lies below one percent.

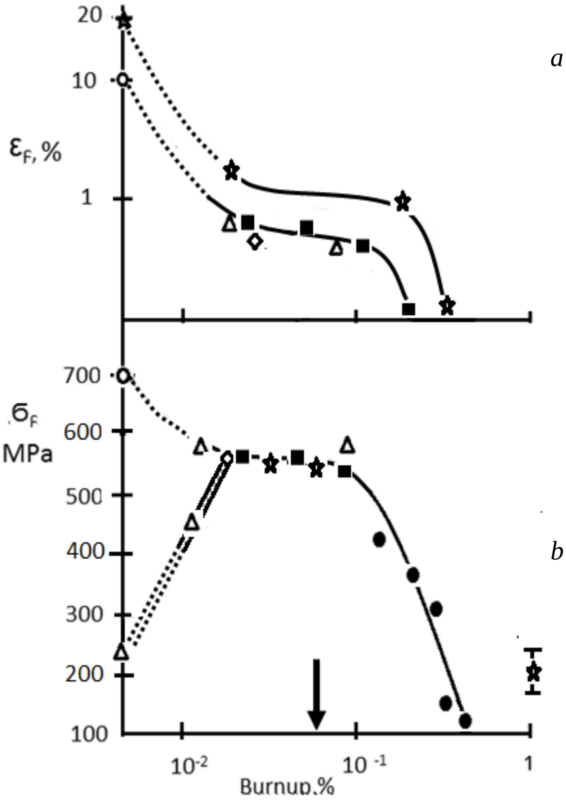

Fig. 2. Influence of the degree of burnupt under the neutron irradiation on the deformation to failure (a) and tensile strength (b) for natural uranium: ● – [7]; Δ – [8]; ◇ – [9]; ■ – [10, 11] *and alloys U (9…10)% Mo;* ★ – [12–14]. *The irradiation temperature of all types of uranium samples did not exceed 140 °C. The test temperature is room temperature. The arrow indicates the maximum burnup level of the NI NSC KIPT uranium target for a year of operation at full power. The level of deformation of non-irradiated samples is in the range of 9…19%*

The following regularities can be noted:

a) The greatest change in ductility to failure is observed when samples from natural uranium and uranium-molybdenum alloys burnup by less than 0.02%. At the same time, their plasticity at room temperature decreases from 11...19 to 0.4...0.5%, i.e. significant radiation embrittlement is observed.

b) Having a significant advantage over natural uranium in a number of characteristics – structural stability, resistance to swelling, recrystallization under irradiation [15], uranium-molybdenum alloys exhibit practically the same reduction in deformation before failure, as uranium (difference in fractions of a percent).

The plates of the NPT KIPT uranium target will receive a maximum dose of about 100 dpa/year [2], and have a burnup level of about 0.055% during the same time (shown by the arrow in Fig. 2). This can lead to almost complete embrittlement of the target material, associated with a high level of damage to the uranium alloy: – in a single act of uranium fission, several hundred thousand point radiation defects are formed.

Up to burnup levels of the order of 0.02% for any materials irradiated and tested at low temperature < 0.3 T_m radiation hardening is observed – an increase in the yield strength, while reducing the tensile strength of uranium (shown by a double dashed line in Fig. 2,b).

A comparison of the results shown in Fig. 2 shows that at burnup levels of more than 0.1%, the destruction of natural uranium samples is accompanied by a significant decrease in the ultimate strength, and also, as shown in [7, 13, 16], a decrease in its density (by 5%). Fracture occurs in the elastic region, without traces of plastic flow.

The dependence of the ultimate strength of polycrystalline uranium on the level of burnup is shown in Fig. 2,b. Like the analogous dependence for the fracture strain, it has a stage nature: the first stage of strength reduction occurs at burnup of less than 0.01%. Further, up to burnup of approximately 0.1%, there is a small plateau, followed by final radiation softening associated with complete embrittlement of the material.

The dependence of the tensile strength on burnup for the U-Mo alloy largely coincides with the same dependence for natural uranium. This is especially pronounced in the range of 0.01…0.1% – in the “plateau” section.

At a burnup level of about a percent, the tensile strength decreased from 1050...1100 MPa to 250...399 MPa (see Fig. 2,b). Structural studies have shown that the fracture occurred without any signs of plasticity, at the elastic part of the stress-strain curve [13, 14]. However, it should be noted that the data for this alloy were obtained under bending deformation, which is considered a “softer” type of test.

In the same works, it was found that at this level of burnup, the value of the elastic modulus decreases significantly due to the presence of porosity, which was 3…5%. The development of porosity in uranium is stimulated by the formation of gaseous fission products in the form of inert gases: helium, argon, xenon, etc. Having a low solubility in metals, they form helium-vacancy bubbles. In [5], it was found that in the U-Mo alloy, in the absence of recrystallization under irradiation, in bubbles with an average size of about 2 nm, the internal pressure can reach 800 MPa. Comparison of these values with the tensile strength of the uranium-molybdenum alloy, taking into account its sharp decrease after irradiation, makes the situation rather alarming from the point of view of embrittlement.

3. MECHANISMS OF RADIATION EMBRITTLEMENT

If we assume (taking into account Fig. 2) the presence of three stages of embrittlement depending on burnup: 1 – burnup level up to 0.02%, when plasticity is

0.4...0.8%; 2 – “plato” for plasticity and 3 – “zero” plasticity when burnup is over 0.1%, – then the failure mechanisms can be represented as follows.

Stage 1 embrittlement. The level of burnup is up to 0.02%, corresponds to doses of the order of tens and hundreds of dpa. Under these conditions, the deformation of irradiated materials with different types of crystal lattice takes place under conditions of plastic instability associated with the effect of dislocation channeling – slip localization in narrow regions less than a hundred angstroms wide [17–19]. The mechanism of crack initiation is related to the concentration of stresses (**σ**) generated by the interaction of localized slip bands having a large number of similar dislocations (**n**) with grain boundaries (**σ = nb**, where **b** is the value of the Burgers vector) (Fig. 3).

According to [15], the decisive factor in the embrittlement of such uranium alloys at low temperatures is the decrease in cohesive strength associated with a change in the state of grain boundaries. In turn, the main type of grain-boundary precipitates in uranium-molybdenum alloys are carbides.

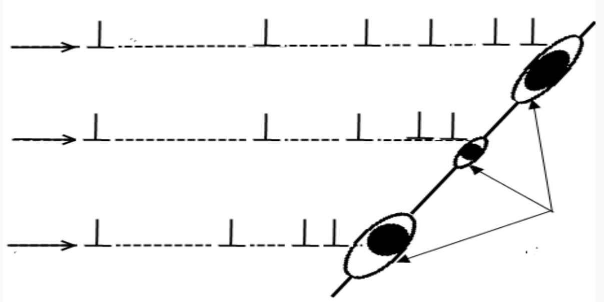

Fig. 3. Pile-up process of interaction of localized slip bands with precipitates at grain boundaries at burnup levels corresponding to stage 1 of embrittlement. The arrows mark the places of formation of vacancy-helium pores on the surface of precipitates

In real U-Mo alloys, even in those smelted under the conditions of vacuum arc remelting, the carbon content in the matrix corresponds to values of the order of (1...7) 10^{-2}% [20]. Taking into account the important role of carbon in the formation of the structure and properties of uranium and its alloys, its concentration was determined corresponding to the formation of a monolayer at the grain boundaries. It was assumed that the shape of the grains is octahedral, and their size is at the level of 130…170 μm, which corresponds to the conditions for optimal heat treatment [20]. As a result, it was found that this concentration is at the level of (5…6) 10^{-3} %. Thus, at the carbon concentrations indicated above, not only a carbon monolayer can be located at the boundaries of uranium grains, but carbides will also form, as noted, for example, in [21], and as shown in Fig. 3.

The interface (uranium carbide)-U-Mo matrix has a great influence on the mechanical stability of the material under irradiation due to a significant difference in the thermophysical characteristics of the carbide and alloy. Thus, it was established that the interatomic bond formed at the interface is rather weak, as a result of which it becomes the predominant site of nucleation for vacancy-helium bubbles [22].

These processes can lead to a sharp decrease in the effective surface energy of the boundaries and, as a consequence, to a decrease in the critical fracture stress (according to the modified Straw criterion [16]). The same patterns are observed in U-(7…9%) Mo alloys in the non-irradiated state: intercrystalline fracture associated with grain-boundary carbon segregations, which reduce its cohesive strength [15].

The second – transitional stage – (“plateau”) is a kind of incubation period for the origin of grain-boundary porosity on carbides. Under these conditions, when the fracture stress (up to 500 MPa, Fig. 2,b) is significantly higher than the yield strength, plastic deformation can take place, which means that there should not be complete embrittlement

At the 3 – “zero” stage, embrittlement is determined by a significant loss of strength Fig. 2,b, and destruction in the elastic region associated with a change in volume due to an increase in volume ΔV/V, (especially due to its local change at the grain boundaries in where they interact with localized slip bands). The expression for the experimentally determined tensile strength of samples σ_f of the irradiated material, taking into account Hooke's law, can be represented in the following form:

$$\sigma_f = \sigma_o - \frac{E}{3(1-2\upsilon)}\,\Delta V/V,$$

where σ_o is the tensile strength of the non-irradiated material, υ is Poisson's ratio.

With an increase in the level of burnup, and the corresponding concentration of vacancies and gas impurities, the process of combining cavities associated with grain boundary carbides (the so-called debonding) (see Fig. 3) can occur, under the action of applied stresses, due to the deformation elimination of bridges between the pores, followed by formation of microcrack nuclei. Thus, the ultimate tensile strength under irradiation decreases as internal stresses increase, proportional to the expansion of the crystal lattice, and as the stress concentration at the grain boundaries increases.

Where does the increase in the volume of uranium and its alloys come from? Usually, the volume of nuclei of fission products is greater than the volume of uranium nuclei from which they were formed [16]. This circumstance leads to the so-called “solid” swelling. The effect does not depend on the irradiation temperature and for alpha-uranium gives an increase in volume of the order of 3…4% per 1 atomic percent burnup [23].

What values of ΔV/V are necessary to create brittle fracture conditions for reactor materials irradiated to high doses? The answer to this question is the results of [16], in which it was found that a volume change of about 7% leads to the destruction of irradiated austenitic stainless steels during mechanical tensile tests at room temperatures in the elastic region of deformations.

A comparative analysis of the dependence of radiation hardening on the fluence and dose of irradiation of uranium and structural materials showed that despite the large difference in the values of neutron fluences (respectively, $10^{15}…10^{16}$ n/cm^2 for uranium [15], and about 10^{20} n/cm^2 for structural materials [18]), the saturation level of radiation hardening (at test temperatures not exceeding 0.25 Tm) corresponds to doses of the order of 0.1 dpa. The same regularity is also observed during mechanical testing of structural materials irradiated with high-energy electrons with an energy of the order of several hundred MeV, i.e. under conditions close to the operating parameters of the target NPT NSC KIPT [24].

For natural uranium irradiated in reactors, doses of the order of 0.1 dpa correspond to a burnup of $2{\cdot}10^{-6}\ldots10^{-5}$ %. According to the results of [25], it is these values that correspond to the effect of saturation of the density of clusters of radiation defects, which have a decisive effect on the radiation hardening of the material. This confirms the decisive role in hardening (and hence in embrittlement, given their direct relationship), namely the level of damage to the material - the concentration of point defects and their complexes.

It was found in [26, 27] that in plate nuclear fuel for experimental and research reactors made of U-10% Mo alloy, surrounded by a coating of 6061 alloy (analogous to SAV), even at extremely high (for this type of fuel) burnup levels (tens of percent) and gas swelling (up to 76%!), despite the destruction of uranium fuel plates, due to the aluminum coating, the mechanical integrity of the fuel elements is maintained.

The situation when the structural materials of nuclear installations operate practically in a brittle state is not new for reactor materials science. So, for example, solid-state targets of existing megawatt ADSs – such as ISIS [28], LANCE [29], the facility under construction in Lund (Sweden) [30], as well as the facility controlled by the high-energy electron accelerator (100 MeV) 100 kW at NSC KIPT, from the very beginning work uses tungsten targets at operating temperatures well below the brittle-ductile transition temperature of these materials.

Under these conditions, of great importance is the analysis of the parameters of high-cycle fatigue of the target plates under conditions close to the parameters of the NPT KIPT target. Such an analysis was carried out in [31]. It was shown that for uranium the endurance limit under high-cycle fatigue (up to $2\ 10^{7}$ cycles) is on the order of 100 MPa. Comparison of this value with the fracture stress of uranium and uranium-molybdenum alloy (see Fig. 2,b) shows that the service life of a uranium target cannot exceed 9 years of continuous operation. In addition to the high-cycle fatigue limit, one should also subtract the "third stage" of the unstable dependence of embrittlement at burnups greater than 0.2%. This gives such a target a service life of 3 years of continuous irradiation.

CONCLUSION

1. Analysis of the results of mechanical testing of uranium and uranium-molybdenum alloys showed that such levels of damage (burnup) can lead to almost complete embrittlement of the target material. At the same time, the advantages of the uranium-molybdenum alloy over natural uranium in terms of plastic characteristics are practically not observed.

2. The dependence of the degradation of strength and plastic properties on the level of uranium burnup is constructed, which consists of three stages: the first – up to burnup of the order of 0.01%, when plastic flow occurs in the irradiated material and an increase in the yield strength is observed, the second – at burnup of 0.01...0.1%, ("plateau") mechanical properties are weakly dependent on burnup, and the third stage is at burnup of more than 0.1%, when softening is observed and a transition to fracture occurs in the elastic region of deformations.

3. A description of the processes of radiation embrittlement is presented, taking into account the deformation and porosity of materials at various levels of burnout.

4. A sharp decrease in the characteristics of plasticity and strength, uranium and uranium-molybdenum alloys, even at burnup levels of several tenths of a percent, can cause a violation of the mechanical integrity of the target plates. However, under conditions of maintaining the continuity of the coating of aluminum alloy SAV-1, this effect will not lead to a change in the performance of the target.

5. An estimate of the expected service life of a uranium target under irradiation has been carried out. Taking into account the influence of high-cycle fatigue and the dependence of embrittlement at burnup greater than 0.2%, gives the value of the target's service life of 3 years at full accelerator power.

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ВИГОРЯННЯ ТА РАДІАЦІЙНА КРИХКІСТЬ U-Mo-МІШЕНІ ДЖЕРЕЛА НЕЙТРОНІВ

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Досліджено вигоряння уранових мішеней джерела нейтронів ННЦ ХФТІ. Проведено аналіз експериментальних робіт по впливу нейтронного опромінення на міцнісні та пластичні властивості урану і сплаву U-Mo в умовах, наближених до умов роботи мішені. Проведена оцінка очікуємого ресурсу роботи пластин уранової мішені під опроміненням.

ВЫГОРАНИЕ И РАДИАЦИОННОЕ ОХРУПЧИВАНИЕ U-Mo-МИШЕНИ НЕЙТРОННОГО ИСТОЧНИКА

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Исследовано выгорание урановых мишеней источника нейтронов ННЦ ХФТИ. Проведен анализ экспериментальных работ по влиянию нейтронного облучения на прочностные и пластические свойства урана и сплава U-Mo в условиях, приближенных к условиям работы мишени. Проведена оценка ожидаемого ресурса работы пластин урановой мишени под облучением.