

RADIATION EMBRITTLEMENT OF TANTALUM COATING OF THE NEUTRON SOURCE TARGETS

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The works in the field of radiation materials science of targets for neutron sources based on subcritical assemblies driven with linear accelerators of electrons or protons, the so-called ADS systems, are presented. Currently, electronuclear ADS systems are prototypes of safe 5th-generation nuclear reactors. In connection with the physical start-up of the neutron source installation of the NSC KIPT the target of which is made of tungsten coated with tantalum, the effect of radiation on mechanical properties is considered, and the resource of the tantalum coating of the target is estimated.

Keywords: Neutron sources; Accelerators driven systems; Targets; Tantalum coating; Radiation hardening; Embrittlement; Dose dependences

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INTRODUCTION

The creation of safe nuclear facilities, designed for both electricity production and the transmutation of thermal reactor products, is one of the priorities of the world nuclear energy industry. Already in one of the first proposals for the development of such facilities, the so-called ADS systems, all the elements of the future reactors were described [1]. The basis of such facilities is a subcritical nuclear reactor using fast neutrons. It was planned to use a proton accelerator with an energy of up to 1 GeV and a current of up to 20 mA as the so-called driver of such a reactor.

Today, 30 years later, the basic structure of such systems is as follows. The accelerator of high-energy particles irradiates the material (target), as a result of which neutrons are formed, which fall on the fuel elements of the subcritical assembly (SCA), after which their multiplication occurs tens of times. The system starts working as a source of neutrons (NS).

Most ADS use neutron sources in which high-energy protons bombard targets made of heavy elements to produce so-called spallation neutrons. In 1947, the term spallation began to be used in nuclear terminology (Siborg, Serber). For nuclear fission to occur, the proton energy must be sufficiently high. Its criterion is the de Broglie wavelength λ . It should be smaller than the size of the nucleus, which will allow the proton to interact individually with the nucleons inside the nucleus. ($\lambda = h/p$, where p is the momentum of the proton), and split it.

Creating (development of) such nuclear accelerator systems, ADS, is a very difficult and challenging task. Electron accelerators can also be used to test the basic ideas of these systems. As shown in 2002 [2], to obtain flows of less than 1016 n/cm² per second, electron accelerators require significantly lower costs for installation. This is explained by the fact that photonuclear (e, γ) processes in this sense have an advantage over reactions induced by protons.

One of the most important areas of experimental work in nuclear materials science is the study of radiation damage in materials used or intended for use in nuclear reactor and ADS components. Two types of neutron-producing targets are being considered for use in future accelerator-driven safe nuclear power systems (ADS): liquid and solid metal. Among the latter, tungsten, tantalum, and their alloys have attracted considerable attention from researchers.

The purpose of the work is the analysis of radiation damage and changes (degradation) of the mechanical properties of tantalum, after irradiation in the active zone of the subcritical assembly, as a structural material (coating) of the "tungsten" target of the neutron source of the NSC KIPT.

KHARKIV NEUTRON SOURCE DRIVEN WITH HIGH-ENERGY ELECTRON ACCELERATOR

In 2021, the physical start-up of just such a facility was carried out at the NSC KIPT together with the Argonne National Laboratory of the USA. Its main functions include developing work in nuclear materials science and medical research. The facility will conduct research in reactor physics and materials science [3,4]. The first target of this installation is a tungsten target with a tantalum coating.

The purpose of the development is to create in Ukraine a safe experimental base for neutron physics research using intense neutron flows (up to 3 10¹⁴ n/sec). The main components of the installation are a linear electron accelerator, a

system for transporting the electron beam from the linear accelerator to the target, a neutron generation target (NGT), a subcritical assembly, neutron channels, biological protection, and auxiliary systems.

Primary neutrons are born in photonuclear reactions with the help of hard γ -radiation, which is formed during the scattering of electrons on the nuclei of heavy elements. Two target variants are considered: tungsten and natural uranium. The initial energy of electrons is 100 MeV.

The target is simultaneously exposed to two sources of irradiation: high-energy electron irradiation (100 MeV) and irradiation from the surrounding environment, i.e., subcritical assembly neutrons (SCA).

The subcritical assembly is designed to obtain the maximum neutron flux with a criticality of 0.98. Thus, the possibility of a chain reaction in an installation of this type is excluded. The size of the neutron flux is regulated by the beam current, and the neutron field in the source disappears after the beam is switched off.

W-Ta neutron-generating targets of ADS systems

Due to its high melting point, density, thermal conductivity, strength, and neutron interaction cross-section, tungsten has been used as a target material in ADS systems such as LANCE, KENS, etc., for many years. [5,6]. However, tungsten corrodes in water, especially under irradiated conditions. This problem can be solved by coating the tungsten with a corrosion-resistant material. These include titanium, stainless steel, and tantalum.

Targets using bimetal tantalum-tungsten were used in the ISIS and KENS installations. A significant positive metallurgical factor is the good compatibility of these materials, due to their complete solubility in the solid state.

Why tantalum? Having unique physical and metallurgical properties, tantalum is one of the most suitable materials for targets. Among the refractory metals of groups 4-6, it has the highest melting point after tungsten (3140°C) and is characterized by exceptional plasticity and viscosity in the cast and recrystallized states, so it can be deformed at room temperature to 90-95% without intermediate annealing.

Pure tantalum (99.99%) retains high plasticity and viscosity at temperatures close to absolute zero [7]. However, as with other bcc metals, it becomes sharply pronounced as the content of interstitial impurities increases. For example, [8] showed that when the oxygen content increases to 1.3 at.%, tantalum becomes brittle at room temperature.

Among all low-value materials, it has the highest corrosion resistance, approaching that of platinum at temperatures up to 150°C. However, it has low resistance to oxidation in air and other oxidizing media at temperatures above 500°C.

The radiation properties of tantalum have not been sufficiently studied and are controversial. Thus, high plasticity of tantalum irradiated to a dose of 10 dpa (displacements per atom) was reported in [9]. However, complete embrittlement of technically pure tantalum was found in [10] even with irradiation doses of only 0.14 dpa. According to one of the authors, the cause of fragility may be the material's oxygen saturation even before the start of irradiation. Alloys of tantalum with tungsten also "do not save the situation": they exhibit a complete loss of plasticity at doses below 1 dpa [7].

The disadvantages of tantalum as a target material include its relatively large interaction cross section with neutrons (especially in the superthermal energy range), which leads to its high radioactivity after irradiation. An analysis of an ESS installation (5MW) showed that after a year of exposure, tantalum exhibited induced activity 10 times greater than that of tungsten or mercury [11]. That is why the two nuclear installations KENS and ISIS immediately replaced tantalum targets with tungsten ones.

One of the main requirements for this effective anti-corrosion tantalum coating is to maintain reliable contact with the tungsten throughout the target's lifetime. The presence of a gap in the tungsten coating dramatically reduces heat removal from the target by a water stream. In turn, the tantalum coating improves heat transfer from the target to the cooling water compared to tungsten.

Reliable Ta-W contact when creating multiple targets for megawatt spallation neutron sources is achieved using the HIP (hot isostatic pressing) method. The successful use of this method was first demonstrated at the ISIS facility (Rutherford Appleton Laboratory in Great Britain) [8], and then by Japanese scientists at the KENS facility [6].

For these purposes, a high-temperature vacuum rolling method, combined with gas-phase deposition of tantalum on the target's side surfaces, has been developed at the National Research Center of KIPT.

At the same time, tantalum with a thickness of 0.25...0.3 mm is used to cover the tungsten plates, as in the LANCE installation (USA). This value is determined by a compromise between ensuring, on the one hand, the reliability of tungsten protection against water corrosion in an active environment and, on the other hand, preventing a significant decrease in neutron yield.

Analysis of the results of mechanical studies of irradiated tantalum.

To achieve the purpose of the work, regarding the assessment of the degree of radiation embrittlement and the target's resource, it was necessary to do three things.

1. To determine the tantalum radiation dose, for example, for one year of operation of the neutron source.
2. To conduct an analysis of the results of mechanical tests of samples irradiated at temperatures corresponding to the conditions of irradiation of the KIPT neutron source.
3. To mark on the dose dependences of strength and plasticity, the tantalum irradiation dose of the KIPT neutron source, and determine the corresponding values of the strength and plasticity of the irradiated material that correspond to it. Based on this, it is necessary to make an estimate of the target coverage resource.

Determination of the radiation dose in tantalum

When the target is irradiated with a beam of electrons with energy of 100 MeV, radiation defects are formed in the target material. The main sources of the formation of such defects will be recoil nuclei, which arise during the scattering of electrons and neutrons on nuclei, as well as a results of nuclear reactions involving gamma- rays.

With the help of the MCNPX program [12], the rate of creation of displacements in the coating of the tungsten target of the neutron source of the NSC KIPT was calculated when irradiated with high-energy electrons with energy of 100 MeV [13]. It was established that the largest contribution to the rate of damage is made by the elastic interaction of high-energy electrons with nuclei. The maximum dose rate is about 0.45 dpa / year, and is reached at a depth of ~ 1 cm (Figure 1).

Figure 2 shows the radiation damage dose distribution in tantalum on the surface of the second tungsten plate.

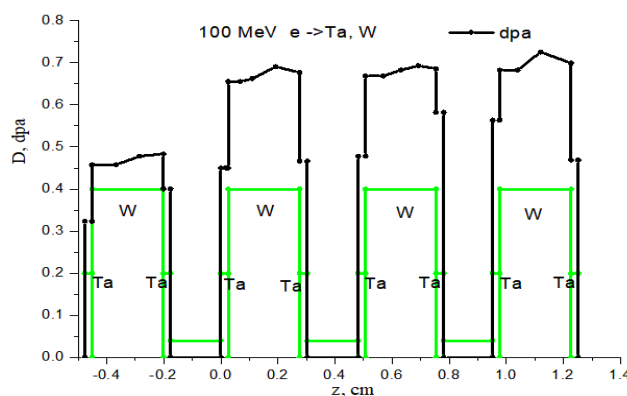


Figure 1. The doses in Ta and W accumulated during one year

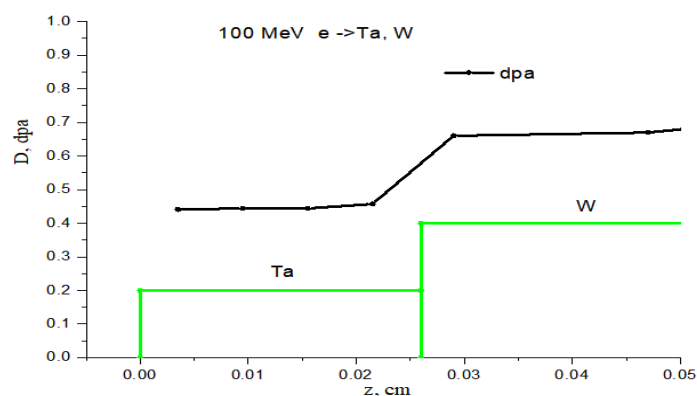


Figure 2. Distribution of radiation damage doses in Ta and tungsten that were accumulated during one year of exposure.

Consideration of damage by neutrons of a subcritical assembly. Let's not forget that the target is under the simultaneous action of two sources of radiation, high-energy electron (100 MeV) and radiation from subcritical assembly neutrons (SCA). It was shown in [13] that the contribution to the dose of irradiation of a tungsten plate from neutrons of a subcritical assembly is 0.15 dpa/year.

Tantalum nucleus differ little from tungsten, but the threshold displacement energies differ greatly. It is 70 eV for tungsten, and 90 eV for tantalum. Taking this into account, we obtain for tantalum the rate of damage by SCA neutrons equal to approximately 0.11 dpa / year. Thus, the total dose accumulation rate (electrons + SCA) in the coating of the second plate is $0.45 + 0.11 = 0.56$ dpa/year. Let's pay attention to the fact that the contribution to damage from high-energy electrons is four times greater than from neutron damage to a subcritical assembly. Is it too much or not enough? What kind of radiation risk does such a dose of exposure indicate? Let's look at the results of mechanical tests of tantalum samples for tension.

Analysis of deformation curves of irradiated tantalum samples

Tantalum turned out to be very sensitive to radiation. As can be seen from Figure 3, the decrease in its plasticity begins already at very low doses - about $4 \cdot 10^{-5}$ dpa. [10].

It is possible to distinguish 3 stages of the development of radiation embrittlement of tantalum:

1) At a dose of $4 \cdot 10^{-5}$ dpa, there is already a reduction in elongation (the strain diagram is shortened), but strain hardening still remains (after the yield point, the flow stress increases slightly).

2) At this stage, starting from $4 \cdot 10^{-4}$ to $4 \cdot 10^{-2}$ dpa, strain hardening no longer takes place. After the formation of the so-called "yield tooth" on the diagram, the stress only decreases. Deformation softening occurs.

3) At a dose of 0.14 dpa, the strain hardening stage and uniform elongation are completely absent. Immediately after reaching the maximum stress, destruction occurs. There is no plastic deformation.

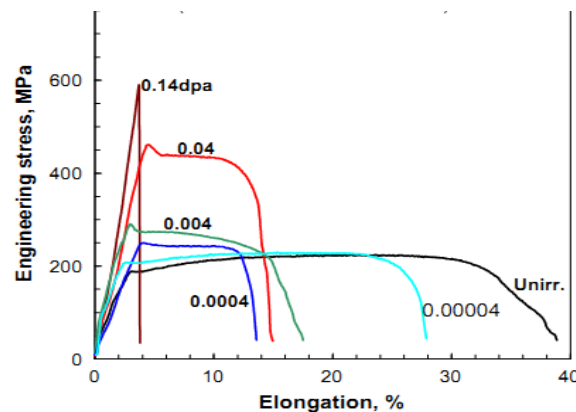


Figure 3. Deformation curves of tantalum irradiated under conditions close to the operating conditions of the neutron source of the NSC KIPT

Dose dependence of flow stress and fracture strength of irradiated tantalum

The main criteria for choosing construction materials are their strength characteristics. The material's ability to undergo uniform deformation is also of great importance. It is generally accepted that the higher these indicators are, the better the material withstands technological and operational loads. That is why, in this section, the dependencies of flow stress and uniform elongation of tantalum of different purities in the initial and irradiated states are presented.

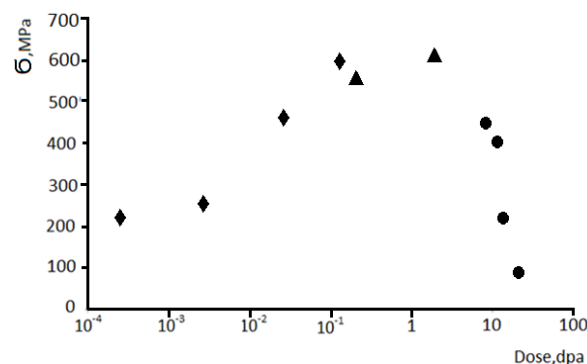


Figure 4. Dose dependence of the flow stress of irradiated tantalum. Rhombuses are work [10], balls are work [14], triangles - work [15]

During the analysis, it should be remembered that the value of the yield strength of non-irradiated samples of tantalum in these works is (depending on the purity of the metal) from 180 to 220 MPa.

Taking into account the value of the working temperature of the KIPT target, when constructing the dependence, we tried to use the results of work on tantalum irradiation at temperatures not exceeding 100°C. Currently, the most number of works on tantalum were carried out at irradiation temperatures higher than 500°C, and were aimed at the operating conditions of future thermonuclear reactors [6].

Dependence (Figure 4) can be divided into two components. At the first stage of irradiation, there is a significant effect of radiation hardening. The yield strength increases almost three times - up to 600 MPa. After reaching a dose of 1 dpa, the flow stress begins to decrease, and for doses greater than 10 dpa, the stress decreases even below the values of the yield strength in the unirradiated state. This means that the process of brittle fracture occurs in the elastic region of stresses, without the contribution of plastic deformation. This is directly evidenced by the deformation curves of material irradiated to high doses [14].

Dose dependence of plasticity (uniform elongation) of irradiated tantalum

In order to predict the degree of fragility of the tantalum coating of the target, it is necessary to know to which purity category the original tantalum belongs to. As can be seen from Figure 5, there is a significant discrepancy between the behavior of tantalum with an oxygen purity of 50 appm and "dirty" tantalum (500 appm): the latter completely loses plasticity at doses slightly above 0.1 dpa. Chemical analysis of tantalum by KIPT on the ELVAX analyzer, showed that the purity of tantalum is 98.8% (category - "dirty").

Thus, the established annual dose of 0.56 dpa for tantalum with a high concentration of oxygen (which corresponds to the operating conditions of the target coating of the NSC KPTI) corresponds to the absolute loss of plasticity.

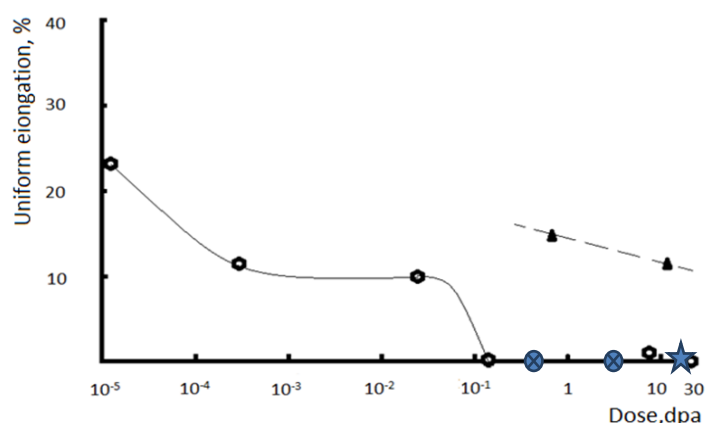


Figure 5. Dose dependence of uniform elongation of tantalum samples of different purity, irradiated at the LANCE (USA) (hexagons), SINQ (Switzerland) (circles and asterisk), triangles ISIS(UK). Triangles correspond to oxygen concentration of 50 appm ($5 \cdot 10^{-3} \%$), and hexagons to oxygen concentration of 500 appm ($5 \cdot 10^{-2} \%$). The size of the asterisk corresponds to the doses of Saito *et al.* [14], which are equal to 10...31 dpa (SINQ). Circles correspond to doses of 0.26 and 2.6 dpa [16], of irradiated “dirty” tantalum (LANCE)

The influence of the impurities and products of nuclear reactions on the development of radiation embrittlement in metals of the 5th group of the Periodic Table of Elements (tantalum). Mechanisms of embrittlement

Fleischer's theory explains the strengthening of bcc metals and alloys by the interaction of dislocations with tetragonal strain fields arising in the metal lattice in the presence of interstitial impurities [17]. Thus, the maximum interaction force of a screw dislocation with an obstacle is given by the expression $F = \mu \Delta \epsilon b^2 / 3.86$, where μ is the shear modulus, $\Delta \epsilon$ is the tetragonal strain (distortion) introduced into the metal lattice by a defect (1 for dislocation loops, either vacancy or interstitial in nature), and b is the Burgers vector.

Analysis showed that for Group 5 metals, interstitial oxygen atoms are one of the most strengthening factors at the neutron irradiation temperatures of interest, which are on the order of 100°C [18]. Specifically, the decoration of dislocations by interstitial atoms or clusters (loops) under irradiation, blocking dislocation generation by Frank-Read-type sources, causes so-called "source" strengthening (increase) of the yield strength—the formation of a "physical yield stress" As we can see from the curves shown in Figure 3, with further increase in strain, this leads to a virtually complete absence of strain hardening and the development of plastic instability.

At the mesostructural level, this is associated with the localization of plastic flow as so-called "dislocation channeling." This effect leads to the formation of defect-free regions—channels. In these channels, the first dislocations to escape from their sources, destroy defective clusters of radiation defects [19]. Dislocation channeling is responsible for the negative slope of the stress-strain curves at doses greater than 10^{-3} dpa (see Figure 3) [20]. The authors of [21] reached similar conclusions on bcc materials irradiated with high-energy protons (590 MeV) to doses ranging from 10^{-3} to 0.3 dpa, and deformed at room temperature. The first study to detect channeling in irradiated ($2.5 \cdot 10^{22}$ n/cm²) tantalum deformed at room temperature may be [22].

How might all these mechanisms influence the initiation of crack formation and propagation in irradiated materials? The absence of dislocation generation leads to suppression of the dislocation mechanism for "healing" crack nuclei, and the crack propagates freely across the entire cross-section of the specimen [23].

Impact of products of nuclear reactions. At the same time, it was established that, in addition to radiation defects and oxygen, the brittleness of materials under high doses of irradiation in ADS-systems can be associated with the influence of high concentrations of nuclear reaction products, mainly helium and hydrogen, which are formed during irradiation [24-27].

According to the authors [26], the main contribution to the sharp (from 35 to 0.5%) decrease in uniform elongation of steel during tensile tests at room temperature is made by helium and hydrogen, the total concentration of which, even at a dose of 3 dpa (in ADS), is about 0.6 at.%. At the same time, it is not related to a change in the microstructure of steel (even the formation of pores), but is determined by a combination of radiation hardening followed by a sharp deformation softening, which is associated with the localization of plastic deformation processes (see explanation to Fig. 3).

CONCLUSIONS

- A brief review of the current state of development of ADS-systems, prototypes of future 5th generation reactors, and an analysis of the use of Ta coatings in neutron-generating targets of ADS-systems was conducted. It is established that W-Ta composites occupy a prominent place in the world, among the targets of the most powerful ADS.

- An analysis of the influence of tantalum irradiation in ADS systems and reactors on its main mechanical properties was carried out. Features of the dose dependence of the plasticity of tantalum (its three-stage nature) and the fact of the determining effect of oxygen on its radiation fragility are shown.

- The dose dependence of the flow and fracture stress of irradiated Ta was established. It is shown that there is a transition from radiation strengthening (at doses of the order of 1-2 dpa) to a significant loss of tantalum strength (radiation softening), even in pure tantalum, at doses of 10 dpa. The material begins to break down in the elastic region, at load levels below the yield point, resulting in brittle failure.
- Briefly analyzed physical mechanisms of influence of radiation defects, products of nuclear reactions, interstitial impurities on strain hardening of irradiated tantalum.
- A prediction was made regarding the behavior of the tantalum coating of the neutron-generating target of the NSC of the KIPT. Taking into account the high concentration of impurities in the material (oxygen), it can be assumed that the tantalum coating of the tungsten target will practically not have a reserve of plasticity already in the first year of operation of the neutron source.

Conflict of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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РАДІАЦІЙНА КРИХКІСТЬ ТАНТАЛОВОГО ПОКРИТТЯ МІШЕНЕЙ ДЖЕРЕЛ НЕЙТРОНІВ**О.О. Пархоменко^{1,2}, В.В. Ганн¹, Б.В. Борц¹, А.Ю. Зелінський¹, І.М. Карнаухов¹, Ю.О. Марченко¹**¹ННЦ "Харківський фізико-технічний інститут", Харків, Україна²Харківський національний університет імені В.Н. Каразіна, Харків, Україна

Представлено роботу в галузі радіаційного матеріалознавства матеріалів мішеней для джерел нейтронів на основі підкритичних збірок, керованих лінійними прискорювачами електронів або протонів, – так званих ADS-систем. Наразі електроядерні ADS системи є прототипом безпечних ядерних реакторів 5-го покоління. У зв'язку з фізичним пуском установки нейтронного джерела ННЦ «ХФТІ», мішень якого виготовлена з вольфраму покритого танталом, розглянуто вплив випромінювання на механічні властивості, та оцінено ресурс танталового покриття мішені.

Ключові слова: джерело нейтронів; системи керовані лінійними прискорювачами; мішені; танталове покриття; радіаційне зміцнення; окрихчення; дозова залежність