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RADIATION DAMAGE OF THE ALUMINUM COATING OF THE URANIUM TARGET OF A NEUTRON SOURCE

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The distribution of proton flux along the depth of the aluminum coating of the uranium target of the NSC KPTI neutron source, irradiated a high-energy electrons with an energy of 100 MeV, was calculated. A relationship has been established between the nuclear-physical characteristics of radiation damage (hydrogen, number of displacements per atom) and the physical-mechanical characteristics of samples of similar materials irradiated with neutrons in nuclear reactors and protons + neutrons in the so-called ADS systems. On this basis, an assessment of the resource of the target coating material was carried out.

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INTRODUCTION

This work is a continuation of a series of studies of the effect of high-energy electron irradiation (100 MeV) on the material properties of uranium and tungsten targets of the Kharkiv neutron source, the physical launch of which took place in 2021 [1–7].

When assessing the degree of radiation damage to materials irradiated in reactors or ADS systems, the main attention is paid to determining the concentration of radiation point defects (Frenkel`s pairs) and the content of nuclear reaction products, both in the form of gases and in the form of solid radiation-induced precepitations.

The structure of the work follows the following logic. First, we will turn to point defects, since it is their evolution under irradiation into clusters, dislocation loops, and pores that is the basis for understanding all the consequences of irradiation. In the next section, the distribution of proton flux and hydrogen concentration along the depth of the aluminum coating of the uranium target is calculated.

In the third part of the work, a relationship has been established between the nuclear-physical characteristics of radiation damage (hydrogen, number of displacements per atom) and the physical-mechanical characteristics of samples of similar materials irradiated with neutrons in nuclear reactors and protons + neutrons in the so-called ADS systems.

1. CALCULATION OF THE CONCENTRATION OF RADIATION DEFECTS

The neutron-producing target of the subcritical assembly (SCA) consists of twelve uranium plates measuring 66x66 mm with a thickness of 0.3 mm and more, coated on both sides with a SAV alloy (over 98 at. % of aluminum, Mg 0.3…0.4 at.%, Si 1.1…1.2 at.%, Fe 0.1…0.2 at.%), 0.7 mm thick. The gap between the 1.75 mm plates is filled with water. The target is described in more detail in [7]. Modeling of nuclear-physical processes of the target was carried out using the MCNPX program.

When an electron beam falls on a target, an (e-γ) shower develops in it. Fig. 1 shows the dependence of the number of electrons n_e in the (e, γ) shower on the depth z of the uranium target, which were formed by a single electron with an energy of 100 MeV. The same figure shows the dependence of the number of photons n_γ with energy of more than 5 MeV on the depth z in the shower.

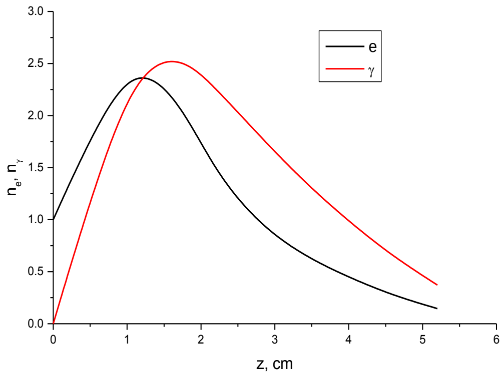

Fig. 1. Dependence of the number of electrons n_e and photons n_γ in the (e, γ)-shower on the depth z

To assess the service life of the target material, the level of radiation damage to the material during irradiation should be taken into account. Analysis shows that the largest contribution to the formation of damage in the aluminum coating is made by elastic scattering of electrons on *Al* nuclei.

The cross section $\sigma_D(E)$ of the displacement of *Al* atoms during elastic scattering of electrons with energy *E* is shown in Fig. 2. In this case, the threshold energy of the displacement of an *Al* atom from a lattice site is 27 eV.

The dose rate R(z) is determined by the expression:

$$R(z) = N_e \int \sigma_D(E) \Phi_e(E,z) dE ,$$

where $N_e = 1.9 \cdot 10^{23}$ el/year is the intensity of electron irradiation (at a beam current of 1 mA), and $\Phi_e(E,z)$ is the energy distribution of the electron flux in the shower at depth z, which is formed when a single electron falls on the target (calculated using the MCNPX program). The rate of dose accumulation of damage to the aluminum coating on the front side of the third uranium plate is 0.4 dpa/year.

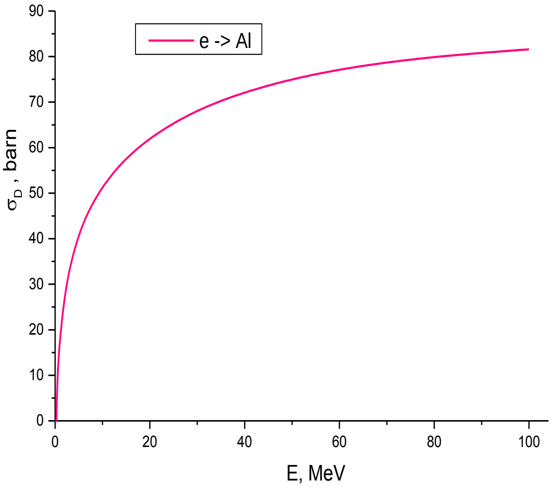

Fig. 2. The cross-section of the radiation damege in aluminum

2. DETERMINATION OF THE CONCENTRATION OF A NUCLEAR REACTION PRODUCTS

When irradiating materials in ADS systems, along with the displacement of atoms from the crystal lattice sites, transmutants accumulate, of which helium and hydrogen play a special role in the evolution of the defect structure. Their concentration in materials over a year of operation can reach values of about one percent, which is much higher than in fission reactors. This is determined by the fact that in such systems the materials are irradiated, usually with high-energy protons with energies of 600…700 MeV [6].

The formation of helium and hydrogen significantly affects the properties of all reactor materials even at very low concentrations – on the order of one appm (0.0001 at.%) [8]. Of course, the question arises about the influence of hydrogen and other nuclear reaction products (NRPs) arising in the uranium target and coating on its mechanical properties and service life.

To assess the magnitude of this impact, it is necessary to assess the concentration and distribution of NPR over the depth of the target. It should be noted that in uranium itself (more precisely, in the uranium-9% Mo alloy), burn-up is accompanied by the release of solid and gaseous fission products, and the production of helium and hydrogen. In this case, He and Kr dominate over other fission products. Their rate of formation is very high, and is about 0.2…0.3 atoms per fission. At an annual burn-up rate of $5.5 \cdot 10^{-2}$ %, the helium concentration in the uranium-molybdenum target will be about 1500 appm [3, 5]. In a non-fissile metal such as aluminum, the situation must be very different.

Fig. 3 shows the cross sections of the reactions Al (γ-p) Mg and Al (γ-α) Na as a function of the gamma-ray energy.

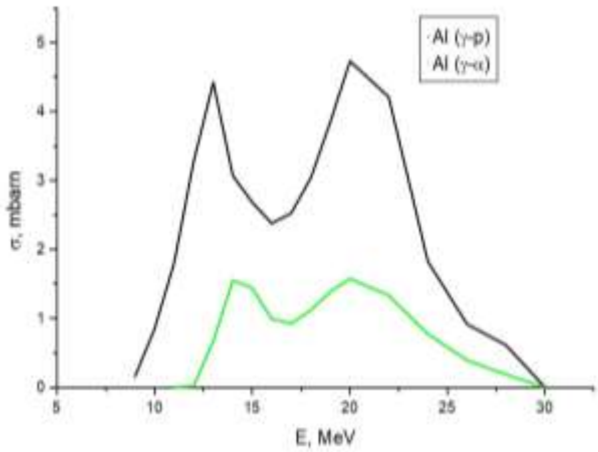

Fig. 3. The cross-section of the Al (γ–p) Mg and Al(γ–α)Na as a function of the gamma-ray energy

Using the MCNPX program, the distribution of proton flux at the depth of the aluminum cover, which covers the front side of the third uranium plate (in the area of maximum neutron flux), was calculated within the depth range of 1.485 < h < 1.555 cm (Fig. 4) (from the beginning of the target).

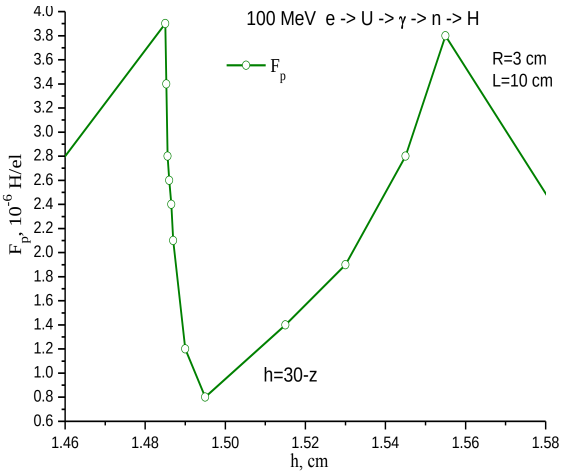

Fig. 4. Distribution of proton flow in Al

It turned out that the intensity of the proton flux incident on the aluminum cover sharply decreases at distances of about 20 μm from the surface of the cover. However, starting from a depth of 100 μm, there is a sharp increase in the proton flux, and at the oppoite surface of the cover, the flux reaches almost its initial value of magnitudes. The mechanism of this amplification is the presence of the nuclear reaction Al^{27}(γ-p) Mg^{26}, which leads to the ejection of photo-protons from aluminum nuclides.

Protons also arise due to the reaction (n-p), the cross-section of which is comparable to the cross-section of the reaction (γ-p), but since the neutron flux is 3 orders of magnitude less than the photon flux, the contribution of the reaction (n-p) can be neglected in the considered processes.

Fig. 5 shows the profile of hydrogen deposition in aluminum and uranium, calculated with a fine step along the coordinate.

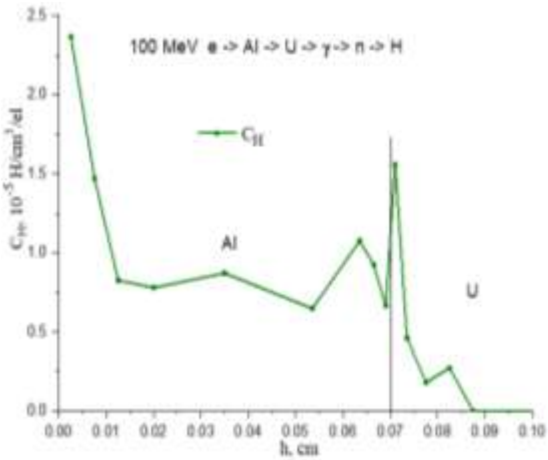

Fig. 5. The profile of hydrogen deposition in an aluminum film and in a uranium foil

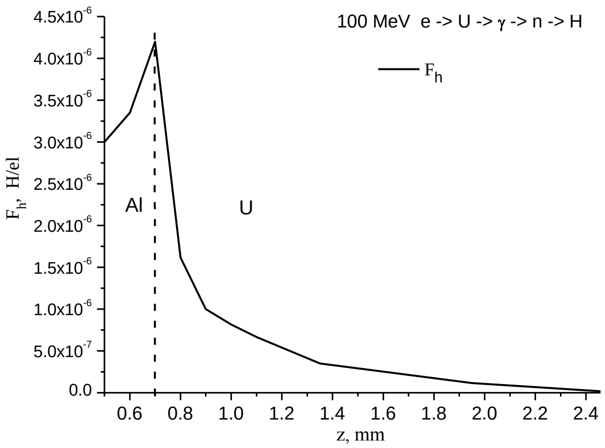

Fig. 6. Distribution of proton flow in Al and U.

As seen from Fig. 5, the concentration of deposited hydrogen obtained from the (γ-p) reaction is three times lower than the concentration of hydrogen deposited from water in the surface layer. However, the total amount of hydrogen deposited in the film via the (γ-p) mechanism exceeds the total amount of hydrogen deposited from water by four times. Fig. 5 shows that the concentration of hydrogen in aluminum at the right edge of the profile begins to drop sharply, although in Fig. 4, the proton flow in this area continues to grow. The question arises: where do the moving protons go?

Fig. 6 illustrates the change in proton flow when transitioning from aluminum to uranium. Photons generated in the aluminum film are quickly deposited in uranium. As seen from Fig. 6, the deficiency of hydrogen in the last two segments of aluminum (30 μm each) is explained by the deposition of this hydrogen in the first two segments of uranium.

Fig. 6 can serve as an illustration of a new effect – the cross-hydrogenation of uranium with photo-protons formed in neighboring aluminum. This is a purely scales, a kinetic effect associated with the interference of flowers with an energy of 10…20 MeV.

As a result, we have that the total amount of hydrogen besieged in the film according to reaction (γ-p) is fore times more than the total amount of hydrogen, besieged by the way of protons from the water. That is, photo-protons are the main source of gidrogenation in aluminum and uranium.

The concentration of helium impurities accumulated in aluminum was calculated using the MCNPX program. It is about 5 appm/year.

3. DISCUSSION. THE RESULTS OF WORKS ON ALLOYS LIKE SAV

Small-alloyed, containing silicon and magnesium, aluminum alloys, such as SAV are used in reactorous building, due to the stability of the main physical chemical properties in conditions of irradiation with high fluences (up to 10^{23} n /cm^2) [9–11, 13, 16].

Table presents the results of mechanical tests of the alloy SAV-1, which is one of the main structural elements of research reactors of the WWER type. The samples were cut from experimental channels, which are the most irradiated elements of the reactors, having operated for more than 30 years. In the radiation damage of this material, transmutation products play an important role, especially silicon, which is formed as a result of the nuclear reaction between neutrons and aluminum atoms Al(n, γ)Si. Thus, in work [9], its content increased by a factor of 3.1, and in work [10], it doubled.

These solid precepitations play an important role in the formation of the alloy's properties. The presence of silicon causes an increase in strength (radiation hardening) of the alloy due to the release of Mg_2Si particles in the aluminum matrix. Under irradiation, the tendency to intergranular corrosion may manifest, which increases with excess silicon content [11] (compared to the ratio in the phase Mg_2Si) [12].

Aluminum alloys of type SAV (AMG3) are used as structural materials for the target of the Swiss neutron source (SINQ) – the exit windows of the proton beam with an energy of 570 MeV. After seven years of operation, the plasticity of the samples cut from the exit windows has significantly decreased but remained at a fairly high level – over 8% [13]. At the same time, the mode of their failure during mechanical testing remained ductile (plastic).

To assess the behavior of the aluminum coating of the target KhFTI, the results of the work presented in [15], carried out on alloy 6063 (analogues of SAV), are of particular interest. This is due to the fact that the structure of SAV-1 KhFTI is partially cold-rolled (about 20…25%), and in these works it is precisely the effects of the influence on low-alloy aluminum alloys, which were before the state deformed by 24…30%. As a result of the implementation of studies, the fact of maintaining the high plasticity of the metal was established in all work due to the processes of the recovery of the deformation structure of the alloy under radiation, even the irradiation drama of only 40…50 °C. From our point of view, this can be explained by the values of the excessive concentration of radiation defects, characteristic of the strong irradiation temperatures.

Mechanical properties of the alloy SAV-1 irradiated to high fluences at room temperature testing

Yield strength, MPa, output / irradiated	Uniform elongation, output / irradiated, %	Fluence, n/cm^2	Irradiation conditions. Reactor / temperature, ^{o}C	Ref.
162.0 / 275.8	16.40 / 5.1	3.48 10^{22}	Dimitrovgrad, (SM-2), 40…50 °C	[9]
–	16.50 / 2.5	3.6 10^{22}	Ukraine, Kiev (VVR-2M), 40…60 °C	[10]
100…106 / 135…140	18…19/ 15	10^{22}	Kazahstan, Alma-Ata (WWR-K), <100 °C	[11]
80…85 / 255…270	17…20 / 8	3.6 dpa	SINQ , Switzerland, 70 °C	[13]
189 / 255	8 / 10	4.5 10^{21}	Beer-Sheva, Israel, <42 °C	[15]
61.3 / 253.8	20.9 / 7.7	17.3 dpa	Оак-Ridge,HFIR, USA 70 °C	[16]

In the work [16], the irradiation of the aluminum alloy (such as SAV) was carried out by the HFIR (Oak Ridge) at 70 °C. The dose amounted to 0.9…17.3 dpa. Uniform elongation was accumulated from 21% in the initial state, to 7.7% at a dose of 17.3 dpa. At the same time, the type of curve indicates a positive deformation hardening of even at maximal dose.

Unlike the tungsten-tantalum target, the NSC KIPT has experience studying the effects of irradiation with high-energy electron beams on both pure aluminum and aluminum alloy type AVT-1 (which has a composition similar to that of SAV). It has been established that only at fluences of about 10^{20} e/cm^2, which corresponds to a dose of $2{\cdot}10^{-2}$ dpa, begins a slight (about 1…3%) reduction in the plasticity of the alloy irradiated at a temperature of 100…120 °C [14].

On most detail, together with scientists from the National Center RISE (Denmark), the influence of irradiation with high-energy electron beams (200 MeV) on the radiation damage of ultra-pure aluminum (99.9999%) was studied at a dose of $4{\cdot}10^{-2}$ dpa [17]. Thus, on the irradiated samples, the concentration of helium produced due to secondary nuclear reactions was experimentally determined. At depths of about 0.6…0.9 radiation lengths, it was 0.6 appm. Electron microscopy studies revealed pores in the aluminum structure ranging in size from 5 to 40 nm, as well as so-called black dots of high density (about $5{\cdot}10^{20}$ m^{-3}). The method of weak beams identified both – stacking faults defects and perfect loops, which turned out to be interstitial.

4. ASSESMENT OF THE LIFE TIME OF ALUMINUM WINDOWS ADS SYSTEMS

In the work [18] for the strict conditions of operation of the output windows from an alloy 5083 (such as SAV) of the Japanese source of neutrons JSNS (3 GeV, flimes $3.2{\cdot}10^{21}$, a dose of 2.7 dpa, 3005 appm of hidrogen and 901 appm of He) calculations showed, that the lifetime of the window is 9500 MW-h (at 8.4 mA/cm^2), which corresponds – 1.6 years, at a capacity of 1 MW and 5000 h of work.

CONCLUSIONS

1. Using the MCNPX program, the distribution of the flow of protoned depth of the aluminum coating of the uranium target of the neutron source is calculated.

2. The contribution of the effect of knocking hydrogen out of cooling water on deposition of hydrogen in the surface layer of aluminum is analized.

3. Establishes a new kinetic effect associated with a syncering of proton streams with an energy of 10…20 MeV**:** effect-cross-gidrogenation of uranium with photo-protons formed in neighboring aluminum.

4. It was established that the main source of gidrogenation in aluminum and uranium is the maintenance of photo protons formed during the development of e-gamma shower. The calculated hydrogen deposition rate; in the near-surface layers of the aluminum coating of the target, it is 130 appm/year . and in depth ~30 appm/year.

5. A comparison of this value was made with the critical values of hydrogen concentrations and levels of damage (dpa) in the coating of the uranium target, which may lead to significant changes in its mechanical characteristics. On this basis it can be assumed that the coating of the uranium target at the NSC KIPT National Academy of Sciences, with a damage level of approximately 0.4 dpa/year, may have a plasticity reserve corresponding at least to three years of operation at maximum power.

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РАДІАЦІЙНЕ ПОШКОДЖЕННЯ АЛЮМІНІЄВОГО ПОКРИТТЯ УРАНОВОЇ МІШЕНІ ДЖЕРЕЛА НЕЙТРОНІВ

В.В. Ганн, А.В. Ганн, Б.В. Борц, О.О. Пархоменко, І.І. Карнаухов

Проведено обчислення розподілу потоку протонів по глибині алюмінієвого покриття уранової мішені нейтронного джерела ННЦ ХФТІ, керованого високоенергетичним електронним прискорювачем з енергією 100 МеВ. Встановлено зв'язок ядерно-фізичних характеристик радіаційної пошкодженності (водень, кількість зміщень на атом) та фізико-механічних характеристик зразків з подібних матеріалів, опромінених нейтронами в ядерних реакторах, і протонами + нейтронами в так званих ADS-системах. На цих засадах проведено оцінку ресурсу матеріалу покриття мішені нейтронного джерелла ННЦ ХФТІ.