NUCLEAR-PHYSICAL PROCESSES OF HYDROGENATION AND EMBRITTLEMENT OF THE ALUMINUM COATING OF A URANIUM TARGET OF A NEUTRON SOURCE

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Using the MCNPX program, the distribution of the proton beam along the depth of the aluminum coating of the uranium target of the neutron source (NI) of NSC KIPT, controlled by a high-energy electron accelerator with an energy of 100 MeV, was calculated. The contribution of the hydrogen knockout effect from the cooling water to its accumulation in the near-surface layer of aluminum was analyzed. A new kinetic effect of cross-hydrogenation of uranium by photo-protons formed in adjacent aluminum has been established. An analysis of the change in the plasticity of SAV-type alloys irradiated by high-energy electrons (200 MeV), neutrons in reactors, protons, and neutrons in ADS systems was conducted.

INTRODUCTION

The hydrogen generated in reactor structural materials under irradiation is one of the sources of embrittlement even at very low concentrations (in uranium, for example, at just a few appm). In this regard, it is necessary to conduct an analysis of its accumulation in the uranium-aluminum target of the neutron source, the physical launch of which was carried out jointly with the Argonne National Laboratory of the USA in 2021 year.

The neutron-producing target of the subcritical assembly (SCA) consists of twelve uranium plates measuring 66x66 mm with a thickness of 3 mm and more, coated on both sides with a SAV alloy (over 98 at.% Al, Mg – 0.3...0.4 at.%, Si – 1.1...1.2 at.%, Fe – 0.1...0.2 at.%) with a thickness of 0.7 mm. The gap between the plates is 1.75 mm filled with water. Water is the source of protons: electrons, photons, and neutrons, scattering on protons create recoil nuclei, which irradiate the aluminum interlayer and become trapped in it at a shallow depth.

1. CALCULATION RESULTS

Using the MCNPX program, the distribution of proton flux at the depth of the aluminum cover, which covers the front side of the second uranium plate (in the area of maximum neutron flux), was calculated within the depth range of 1.485 < h < 1.555 cm (from the beginning of the target), Fig. 1. 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 μ m, there is a sharp increase in the proton flux, and at the opposite 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 Al₂₇(γ - p) Mg₂₆, which leads to the ejection of photoprotons from aluminum nuclides. Fig. 2. shows the dependence of the cross-section of this reaction on photon energy, which was used in our calculations.



Protons also arise due to the reaction (n-p), the cross-section of which is comparable to the cross-section of the reaction $(\gamma-p)$, 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. 3 shows the profile of hydrogen deposition in aluminum and uranium, calculated with a fine step along the coordinate, and Fig. 4 presents a detailed profile of hydrogen deposition in the near-surface area of the aluminum film. As seen from Fig. 4, the concentration of deposited hydrogen obtained from the $(\gamma-p)$ 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. 3 shows that the concentration of hydrogen in aluminum at the right edge of the profile begins to drop sharply, although in Fig. 1, the proton flow in this area continues to grow. The question arises: where do the moving protons go?



Fig. 3. The profile of hydrogen deposition in an aluminum film and in a uranium foil

Let us take a closer look at the boundary region between aluminum and uranium (Fig. 5).



Fig. 4. Profile of hydrogen deposition in the aluminum film

Fig. 5 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. 3, the deficiency of hydrogen in the last two segments of aluminum (30 μ m each) is explained by the deposition of this hydrogen in the first two segments of uranium.

Fig. 3 can serve as an illustration of a new effecteffect-cross-gidrogenation of uranium with photoprotons 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 $(\gamma$ -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 hydrogen in aluminum and uranium.



Fig. 5. Distribution of proton flow in Al and U

2. DISCUSSION. THE RESULTS OF WORK ON ALUMINUM, AND ITS 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 flimes (up to 10^{23} n /cm²) [1-6].

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 VVER 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 [1], its content increased by a factor of 3.1, and in work [2], it doubled. These emissions 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 Mg₂Si particles and silicon in the aluminum matrix. Under irradiation, the tendency to intergranular corrosion may manifest, which increases with excess silicon content (compared to the ratio in the phase Mg_2Si) [4].

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

Yield strength, MPa,	Uniform elongation,	Fluence	Irradiation conditions. Reactor /	Refe-
output / irradiated	output / irradiated, %	(n/cm^2)	temperature, °C	rences
162.0 / 275.8	16.40 / 5.1	$3.48 \cdot 10^{22}$	Dimitrovgrad, (SM-2), 4050 °C	[1]
	16.50 / 2.5	$3.6 \cdot 10^{22}$	Ukraine, Kiev (VVR-2M), 4060 °C	[2]
100-106 / 135-140	18-19/ 15	10 ²²	Kazahstan, Alma-Ata (VVR-K), <100 °C	[3]

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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% [5]. At the same time, the mode of their failure during mechanical testing remained ductile.

To assess the behavior of the aluminum coating of the target NI KhFTI, the results of the work presented in [7, 8], carried out on alloy 6061 and 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 lowalloy 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 course of processes of the return 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 radiation excessive concentration of defects. characteristic of the low irradiation temperatures.

In the work [9], the irradiation of the aluminum alloy (such as SAV) was carried out by the HFIR (Oak Ridge, USA) 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 National Center for Science and Technology (NSTU) has experience studying the effects of irradiation with highenergy electron beams on both pure aluminum and aluminum alloy type AVT-1 (which has a composition quite similar to that of SAV). It has been established that only at fluences of about 10^{20} e/cm², which corresponds to a dose of $2 \cdot 10^{-2}$ dpa, begins a slight (about 1...2%) reduction in the plasticity of the alloy irradiated at a temperature of 100...120°C.

In 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. 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 $5 \cdot 10^{20} \text{ m}^{-3}$). The method of weak beams identified both stacking fault defects and perfect dislocation loops, which turned out to be interstitial.

3. ASSESSMENTS OF THE LIFE OF ALUMINUM TARGETS (OUTPUT WINDOWS ADS SYSTEMS

In the work [11] 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, $3.2 \cdot 10^{21}$ protons cm⁻², 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/cm²), 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 protons 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 concentration of hydrogen in the underlying layer of the target coating is 130 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 Institute of Nuclear Research of the National Academy of Sciences, with a damage level of approximately 1 dpa /year, may have a plasticity reserve corresponding to three years of operation at maximum power.

REFERENCES

1. V.M. Lebedev, V.T. Lebedev, S.P. Orlov, et al. Study of the nanoscale structure of the alloy SAV-1, irradiated with fast neutrons to high fluences // *Physics of Solid State*. 2014, v. 56, No. 1, p. 160-167.

2. V.S. Karasev, N.S. Zaritsky, S.D. Mamchich, L.I. Chirkov, et al. Structure and properties of the SAV-1 alloy after prolonged reactor irradiation // *Reactor Materials Science. Proceedings of the International Conference on Reactor Materials Science.* Moscow, TsNIIMF. 1990, v. 5, p. 112-117.

3. O.P. Maksimkin, A.V. Yarovchuk, L.G. Turubarova, et al. Effect of neutron irradiation on intergranular corrosion and stress corrosion cracking of SAV-1 alloy // VANT. Series "FRP and RM", 2011, No. 2, p. 108-115.

4. Fridlyander. *Aluminum deformable structural alloys*.M.: Metallurgy, 1979, 208 c.

5. Y. Dai, D. HAAMAGUCHI. Mechanical properties and Microstructure of Al Mg₃ // *Irradiated in Sinq Target-3. Journal Nucl.Materials.* 2005, v. 343, 1184-190.

6. L.S. Ozhigov, A.A. Parkhomenko, V.I. Savchenko. The influence of radiation with high-energy-electrons on the mechanical properties of aluminum alloys // PAST. 1990, Iss. 53, p. 50-55.

7. A. Munitz, A. Sctekman, C. Cotler. Mechanical properties of ntutron irradiated cold worked Al-6063 alloy // *Journal Nucl. Materials.* 1998, v. 252, 79-88.

8. R.T. King, A. Jonston // Metall. Trans. A6, 1975, 863-870.

9. M.N. Gussev, N. Shridharan, S.S. Babu, R.F. Terrani. Influence of neutron irradiation on al-6 alloy produced via ultrasonic additive manufacturing *// Journal of Nuclear Materials.* 2021, 550.

doi.org./10.116/j.nucmat.2021. 152939

10. V.V. Gann, L.S. Ozhigov, V.A. Yamnitsky, B. Sinh, T. Luffers, I. Bilde-Surensen. Porosity in pure aluminum, irrigated by the bundle of high-energy electronics of gamma-jeans // PAST. 1983, Iss. 1 (24), p. 41-46.

11. S. Meigo, M. Ooi, M. Harada, H. Kinoshita, A. Akutsu. Radiation damage and lifetime estimation of the proton beam window at the Japan Spallation Neutron Source // *Journal of Nuclear Materials*. 2014, 450, 141-146.