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NANO-MECHANISMS OF CONNECTION IN THE SOLID PHASE OF TUNGSTEN AND TANTALUM IN THE MANUFACTURE OF A NEUTRON SOURCE TARGET

B.V. Borts[1], I.A. Vorobyov[1], A.Yu. Zelinsky[1], I.M. Karnaukhov[1], A.A. Lopata[1], A.A. Parkhomenko[1,2], I.V. Patochkin[1], V.I. Tkachenko[1,2]
[1]National Science Center "Kharkov Institute of Physics and Technology", Kharkiv, Ukraine;
[2]V.N. Karazin Kharkiv National University, Kharkiv, Ukraine
E-mail: borts@kipt.kharkov.ua

Nano-mechanisms of solid-phase bonding of tungsten and tantalum, which are implemented in the manufacture of a neutron source target for the research nuclear facility (RNF) of NSC KIPT, are considered. This facility is a new type of nuclear reactor, in the core of which the intensity of the nuclear fission reaction of the uranium isotope ^{235}U is controlled by an electron accelerator. An installation for joining metals in the solid phase is described, its parameters are given, as well as the parameters of the joined metals. An electron-probe X-ray spectral analysis of the interface between the samples was carried out. The physical foundations of the nano-mechanism of bonding in the solid phase of tungsten and tantalum at the dynamic and isostatic stages at hot vacuum pressing are formulated. Experimental data on the relative temperature convergence of rolls at the dynamic and isostatic stages of hot vacuum pressing have been obtained.

INTRODUCTION

The nuclear research facility (NRF) under development at NSC KIPT is a new type of nuclear facility, in which the intensity of the nuclear fission reaction of the uranium isotope ^{235}U in the core is controlled by an electron accelerator [1, 2].

The IAEA international classification of such installations is ADS-systems (Accelerator Driven Systems). At present, all concepts of such systems being developed at CERN, LANL, KENS are based on the spectrum of fast neutrons in a subcritical assembly (SCA).

Target-converters of such generators, which convert accelerated charged particles into high-intensity neutron fluxes, are a very important unit, they have a number of requirements and limitations, primarily due to the purpose directions and technological features of the installations. Neutrons are produced as a result of photoneutron reactions (γ, xn) from bremsstrahlung of electrons incident on a target of heavy nuclei. On Fig. 1 shows the dependence of the neutron yield per unit power [3]. In the system of NRF NSC KIPT, 2 types of targets-converters are being developed: 1) based on tungsten and 2) based on uranium. They are used to fabricate target plates with high heat flux and as structural materials with high density in the radiation environment. The NSC KIPT carried out research work to determine and develop the optimal, from the point of view of materials science, target material for a 100 MeV source, a current of 1 mA and a power of 100 kW.

However, it has been found that tungsten has poor corrosion resistance to aqueous coolants due to the formation of fragile tungsten hydroxide [4–6] and high susceptibility to radiation embrittlement [7]. One way to eliminate these problems is to clad the tungsten with a corrosion resistant material such as tantalum, titanium, stainless steel, zircalloy, niobium, or gold.

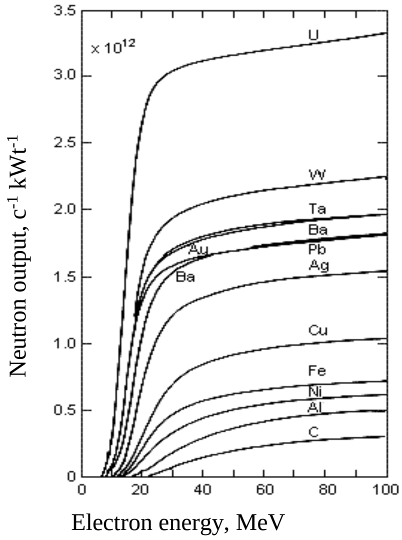

Fig. 1. Dependence of the neutron yield per unit power of the incident electron beam

The result of this action is an increase in resistance to radiation damage, a decrease in low-temperature recrystallization and radiation embrittlement. To create a tungsten target for a neutron source, tantalum was chosen as a protective material. In this regard, a new task arose – to develop a technology for the production of a target resistant to long-term exposure to an electron beam with an energy of 100 MeV, a power of up to 100 kW and resistant to corrosion in a cooling aqueous medium.

Previously, our studies were aimed at fabricating a target from pure cast tungsten with a fine-grained structure. However, they were unsuccessful, due to intergranular cracking of the target material. The solution to the problem was found by manufacturing target plates from pure tungsten powder by hot vacuum pressing.

To protect the tungsten, tantalum was chosen as one of the best materials resistants to corrosion under irradiation in the aquatic environment.

High-purity tantalum powder was deposited on the side surface of tungsten plates, 1.2 mm thick, from $TaCl_5$ powder, using the CVD method, on a specially designed “Termit” installation.

The connection in the solid phase of flat protective surfaces of tungsten and tantalum plates, 250 μm thick, was carried out using an intermediate damping layer of titanium by the following method:

a) dynamic hot rolling in vacuum;

b) isostatic pressing method using a vacuum rolling mill.

Rolling of dissimilar metals in vacuum and at high temperature was carried out on a vacuum rolling mill DUO-175 [8–11]. The schematic diagram of the installation is shown in Fig. 2.

The installation consists of a vacuum system 6 providing a vacuum $p = 10^{-2} \ldots 10^{-3}$ Pa, a furnace 2 for heating samples to a temperature of $T \approx 1300$ °C and a chamber with rolls 4 providing a rolling speed $V_0 = 0.03$ m/s and compression force $P = (2 \ldots 32) \cdot 10^2$ MPa. The radius of heat-resistant steel rolls simultaneously rotating with the same angular velocity in oppos.

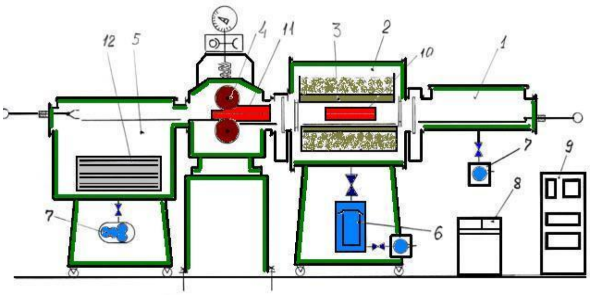

Fig. 2. Vacuum rolling mill for joining dissimilar materials in the solid phase: 1 – loading chamber; 2 – vacuum furnace; 3 – ceramic insulator; 4 – rolls; 5 – chamber for unloading and collecting finished products; 6 – vacuum system consisting of a diffusion pump 6 and fore vacuum pumps; 7, 8 – racks control of the technological process of rolling; 9 – automated control system and control of the technological process; 10 – weldable plate pack in the furnace; 11 – weldable plate pack in the roller chamber

To ensure the process of joining Ta-Ti-W-Ti-Ta metals, a batch scheme for joining metals of different hardness and plasticity was used. The rolled metals were placed in a strong Nb alloy mandrel in the following sequence: a W plate of different thicknesses from 2.5 to 9.5 mm is in contact on both sides with Ti plates 30…40 μm thick. In turn, Ti plates are in contact with Ta plates with a thickness of 240…250 μm. The surface W had irregularities in the form of protrusions and recesses, obtained during processing on an electroerosive machine. The height of the plates varied in the range of 1.5…10 μm, and the transverse dimensions corresponded to the length and width of the tungsten target. The tungsten plate had the shape of a square with a side of 65.8 mm.

The mandrel made of N holds the metals to be joined in the solid phase with the set order Ta-Ti-W-Ti-Ta was placed in a rolling mill according to the scheme shown in Fig. 3.

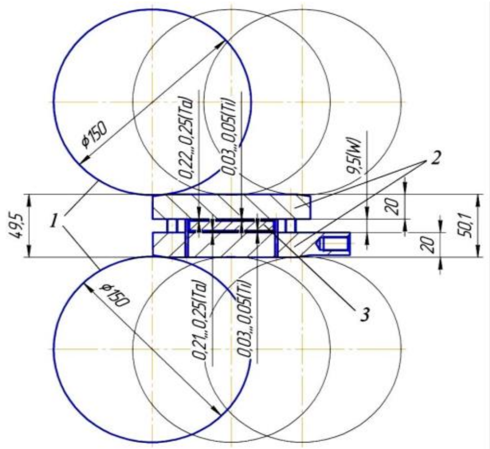

Fig. 3. Layout of the rolls of the rolling mill and rolled metals: 1 – rolls; 2 – Nb alloy mandrel; 3 – joined metals in the solid phase, arranged in the order Ta-Ti-W-Ti-Ta

MODEL OF THE COMPOUND OF TUNGSTEN, TITANIUM AND TANTALUM BY ROLLING IN VACUUM

In the experiment, the connection of Ta-Ti-W-Ti-Ta by hot rolling in vacuum is carried out in two stages, preliminarily by heating the package in a furnace to 1300 °C with an exposure of about 1 h and feeding it under the rolls.

At the first stage of rolling, which we will call **dynamic**, Ta-Ti-W-Ti-Ta layers are installed rigidly and symmetrically with respect to the tungsten layer in a strong Nb alloy mandrel. The mandrel is moved by rolls. The movement of the mandrel is carried out in the forward, reverse and again in the forward direction. Rolling ends when the rolls are in the middle of the mandrel.

In the second stage of rolling, the rolls remain in a position in the middle of the mandrel, and this state is maintained for several hours. This stage of rolling will be called **isostatic**.

The experimental conditions indicate the absence of relative motion of the rolled metals: a special mandrel does not allow the metals to move relative to each other. However, it transmits the force of the rolls in the direction transverse to the speed of movement of the metals. Due to the high pressures and rather high temperature of the samples, Ti can pass into a quasi-liquid state [12], while the Nb, Ta, and W alloys remain in the solid phase.

In a microscopic measurement on a thin section depicting the interface between W and Ti after the dynamic stage, one can see the incomplete connection of W and Ta through the Ti interlayer, as shown in Fig. 4.

It follows from Fig. 4 that, immediately after the dynamic stage of rolling, an incomplete penetration of Ti into W occurs. An increase in the load does not lead to complete filling of the cavities with titanium, but only to uncontrolled deformation of the mandrel and the entire package.

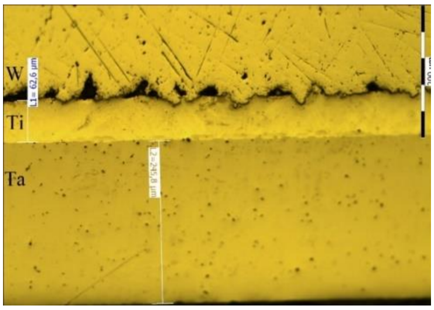

Fig. 4. Microphoto of the boundaries of the W, Ti, Ta compound after the first dynamic stage. At the interface between W and Ti, one can see unfilled cavities in W

The cavity filling condition W was found as a result of a series of experiments. To do this, it is necessary to leave the package under load in a hot state, followed by cooling. In the process of isostatic loading and when the temperature drops from 1200...1300 to 882 °C, within 10…12 h, quasi-liquid Ti fills all depressions in W (Fig. 5).

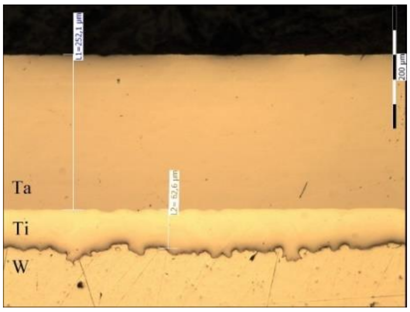

Fig. 5. Microphoto of the boundary of the W+Ti+Ta compound after the second, isostatic stage of pressing. Filled cavities in W are visible at the interface between W and Ti

PHYSICAL MODEL OF W, Ta, AND Ti COMPOUND BY THE METHOD OF ISOSTATIC HOT VACUUM PRESSING

PHYSICAL BACKGROUND TO FORMULATION OF THE COMPOUND MODEL

The properties of titanium $\alpha+\beta$-alloys are such that, above a temperature of 882 °C, it has superplasticity and fluidity in the region of the β-phase [13]. On Fig. 6 shows the phase diagram of the state of a binary Ti-Ta mixture as a function of temperature.

An analysis of the phase diagram indicates that at the boundary of the solid-phase Ti-Ta compound under equilibrium conditions, the sample should consist of the α-phase of titanium and pass into the tantalum region through the two-phase region $\alpha+\beta$, which provides superfluidity when all irregularities are filled in the solid-phase connection.

The study by scanning electron microscopy of the microstructure of the zones of the W-Ti and Ti-Ta joints (Fig. 7), as well as in the body of the constituent samples, showed the absence of defects in the form of pores, delaminations, cracks or inclusions. An X-ray electron probe microanalysis made it possible to determine the widths of the diffusion zones for Ta-Ti and Ti-W equal to 2 and 3 μm, respectively.

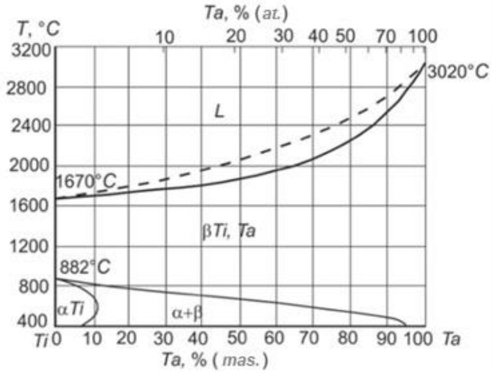

Fig. 6. Phase diagram of the state of the binary mixture Ti-Ta as a function of temperature

The nature of the slope of the concentration curves at the boundaries of Ta-Ti and Ti-W indicates the predominant penetration of quasi-liquid titanium into both tantalum and tungsten, which is explained by the high mobility of titanium.

When an isostatic load occurs, Ti penetrates into micro-depressions in W, and the connection in the solid phase of the surface of Ti and W occurs along the entire interface.

In addition, at this stage, we will assume that, in view of the quasi-liquid state of Ti, the metal bonding process occurs as a result of the penetration of Ti into the roughness of the tungsten boundary. Since the irregularities of tungsten are in a vacuum, the joining process will be associated with the penetration of a heavier substance (quasi-liquid Ti) into a lighter substance (vacuum). The whole system is placed in a force field, the effective acceleration of which g^* is determined by the force of volumetric compression Ti with the Ta+Nb alloy on the one hand, and W on the other, due to different coefficients of their thermal expansion.

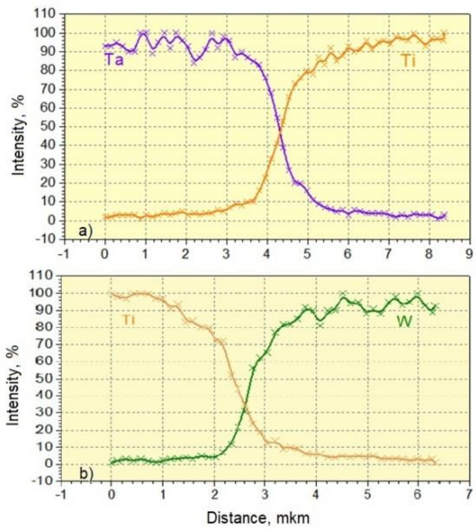

Fig. 7. Electron probe X-ray spectral analysis of Ta-W-Ta samples (Ti interlayers) through the boundaries of the connection: a – tantalum-titanium and b – titanium-tungsten

Thus, we come to the Rayleigh-Taylor instability problem, which describes the motion of the interface between liquid media in a gravitational or other [14] force field. In media with dissipation, which is the viscosity of liquids, such instabilities are called dissipative Rayleigh-Taylor instabilities (DRTI) [15].

To describe the process of combining titanium with tungsten, a model description was proposed in [16]. It is based on the use of the DRTI theory. An estimate of the values of the parameters of a binary metal system subjected to DRTI is given.

EXPERIMENTAL RESULTS AND DISCUSSION

Based on the use of the DRTI model, the estimate of the characteristic connection time turns out to be about 40 s. The validity of the model is confirmed by the experimentally measured relative change in the diameter of the rolls at the dynamic and isostatic stages of rolling. The relative change in the roll gap is the difference between the current convergence of the rolls and the initial one, which in the experiment was chosen to be 100 μm. The initial convergence of the rolls during rolling or isostatic hot vacuum pressing is determined by the difference between the thickness of the Nb – $H_{As}(t)$ and the vertical distance between the generatrixes of the upper and lower rolls – $H_{Rolls}(t)$, where *t* is time.

Due to the small, applied stresses during rolling, we will assume that all metals, except for Ti, obey Hooke's law, i. e. when stress is removed, they return to their original shape. Therefore, a change in the dimensions of rolled metals can occur as a result of linear expansion / contraction of the samples when their temperature changes.

The experimentally obtained relative temperature change in the gap between the rolls at the dynamic and isostatic stages of rolling is shown in Table.

Relative temperature convergence rolls at dynamic and isostatic stages

	Dynamic stage									
Time, s	0	8…9	–	–	–	–	–	–	–	–
Relative discrepancy, μm	0	20	–	–	–	–	–	–	–	–
Phase of the stage	a)		–	–	–	–	–	–	–	–
			Isostatic stage							
Time, s	–	–	0	10	25	40	50	80	200	510
Relative discrepancy, μm	–	–	20	0	-20	-5	0	15	75	85
Phase of the stage	–	–	b)			c)		d)		

a) the mandrel with metals is heated, and its thickness increases, and the relative divergence of the rolls increases from zero to 20 μm. The distance between the rolls does not change due to a small change in their temperature;

b) the assembly cools down, the rolls heat up, and Ti is still in a quasi-liquid state;

c) with a decrease in the assembly temperature, Ti passes from a quasi-liquid state to a solid state. The temperature difference between the rolls and assembly remains quite high. Therefore, the cooling of the assembly and the heating of the rolls can be considered as heat exchange between the heated and cold solids. Currently interval, the heating of the assembly occurs due to the work of viscous forces arising from the thermal expansion of the assembly and rolls;

d) the temperature gradient between the rolls and the assembly decreases so much that the entire assembly of metals St.s+Nb+Ta+Ti+W+Ti+Ta+Nb+St.s cools over time as a whole, due to radiant heat transfer. In this case, the relative divergence of the rolls increases.

From Table, it follows that during the dynamic stage, during rolling in the forward and reverse directions for about 9 s, the massive rolls were in contact along the generatrix line with the surface of the heated assembly over a rolling length of 10 cm, which corresponds to the rolling of the rolls clockwise and in the opposite direction by angle 67.44º. As a result of such contact, the rolls heat up slightly, and the assembly, in preparation for rolling, is preheated in the furnace to 1300 °C. During rolling, the rolls are heated, because of plastic deformation of the rolled package, so that the relative convergence with an external increase in the diameters of the rolls at the isostatic stage will increase to 20 μm. The increase in the assembly temperature in this case is estimated at about 25…30 ºC [17]. Then, at the isostatic stage, the rolls are heated through the area of contact with the package assembly, and the assembly of Nb+Ta+Ti+W+Ti+Ta+Nb metals is cooled in accordance with their thermal conductivity coefficients.

Thus, because of the cooling process of the assembly and heating of the rolls and, accordingly, their increase in diameter, during the isostatic stage, volumetric thermal compression of the assembly is observed, and a decrease in pressure in Ti, which is still in a quasi-liquid state for a certain time. This stage, as shown by experiment and numerical evaluation, lasts on the order of several tens of seconds. During this time, the tungsten cavities are filled with quasi-liquid titanium because of the development of DRTI [16].

After that, the rolls continue to heat up, and as a result of their volumetric thermal expansion, the force of isostatic vacuum pressing of the package assembly is increased. After a long period of cooling (about 12 h) of the entire system, Ti passes into the solid phase, and the process of joining Ti+W is completed.

On Fig. 8 shows a graph of the relative change in the diameter of the rolls from the time of rolling, in the dynamic and isostatic stages. It follows from this figure that the experimentally measured time for changing the gap between the rolls and decreasing the pressure in Ti corresponds to the characteristic time for the development of DRTI.

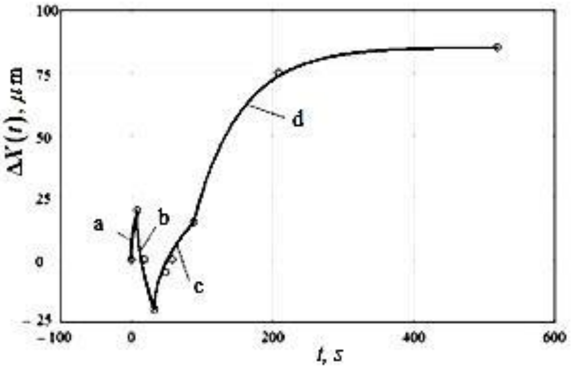

Fig. 8. Dependence of the relative change in the diameter of the rolls $\Delta X(t)$ on the rolling time: ○ – experimental points, the solid broken line consists of dynamic and isostatic stages with stages: a; b; c; d

CONCLUSIONS

The article deals with the manufacture of a target-converter for the RNF under development at NSC KIPT, which is a new type of nuclear reactor, in the core of which the intensity of the nuclear fission of the uranium ^{235}U isotope is controlled by an electron accelerator. The process of joining W with Ta in the solid phase with the help of a Ti interlayer by the vacuum rolling method is considered. To facilitate the process of joining these metals, it is proposed to use the previously developed batch scheme for joining metals of different hardness and plasticity. The combination of W and Ta by rolling in a vacuum is carried out in two stages. The first is the dynamic stage of rolling. The second is the isostatic stage of pressing the W and Ta compound by rolling in vacuum, controlled by nano-mechanisms with the appropriate temperature-force parameters. A scheme has been developed for rolling a mandrel made of niobium with joined metals in the solid phase. An electron-probe X-ray spectral analysis of the boundary between the titanium-tungsten and titanium-tantalum junctions has been carried out. The predominant diffusion penetration of titanium into tantalum and tungsten is shown. The stages of the physical foundations of nano-mechanisms of joining in the solid phase of tungsten and tantalum at the dynamic and isostatic stages of hot vacuum pressing are determined. Experimental data on the relative temperature convergence of the rolls at these stages have been obtained.

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НАНОМЕХАНІЗМИ З'ЄДНАННЯ У ТВЕРДІЙ ФАЗІ ВОЛЬФРАМУ І ТАНТАЛУ ПРИ ВИГОТОВЛЕННІ МІШЕНІ НЕЙТРОННОГО ДЖЕРЕЛА

Б.В. Борц, І.А. Воробйов, А.Ю. Зелінській, І.М. Карнаухов, А.А. Лопата, О.О. Пархоменко, І.В. Паточкін, В.І. Ткаченко

Розглянуто наномеханізми з'єднання у твердій фазі вольфраму та танталу, що реалізуються під час виготовлення мішені нейтронного джерела для дослідницької ядерної установки НДК ПЯС ННЦ ХФТІ. Ця установка є новим типом ядерного реактора, в активній зоні якого інтенсивність протікання ядерної реакції поділу ізотопу урану ^{235}U керується прискорювачем електронів. Описано установку для з'єднання металів у твердій фазі, наведено її параметри, так само як і параметри металів, що з'єднуються. Проведено електронно-зондовий рентгеноспектральний аналіз межі з'єднання зразків. Отримано експериментальні дані щодо температурного сходження валків на стадіях: динамічній і ізостатичного гарячого вакуумного пресування.