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THERMAL ANNEALING EFFECTS ON MICROSTRUCTURE AND MECHANICAL PROPERTIES OF THE Ta-W-Ta SOLID PHASE JOINTS FOR NEUTRON PRODUCING TARGETS OF THE RESEARCH NUCLEAR FACILITY “SOURCE OF NEUTRONS”

A.A. Lopata*[1]*, M.P. Domnich*[1]*, B.V. Borts*[1]*, A.Yu. Zelinsky*[1]*, A.A. Parkhomenko*[1]*, V.I. Sytin*[1]*,
***V.I. Tkachenko*[1]*, I.A. Vorobyov*[1]*, A.D. Pidlisnyi*[2]*, I.V. Patochkin*[1]*, L.I. Gluschenko*[1]**
[1]*National Science Center “Kharkov Institute of Physics and Technology”, Kharkiv, Ukraine;*
[2]*Kharkiv National University of the Air Force named after Ivan Kozhedub, Kharkiv, Ukraine*
E-mail: borts@kipt.kharkov.ua

The paper considers the neutron-producing tantalum-tungsten-tantalum target using a niobium interlayer manufacturing variant by means of vacuum hot roll bonding for the research nuclear facility «Source of Neutrons». The metallography results and the Ta-W-Ta solid-phase junction specimens mechanical testing ones obtained by hot roll bonding in vacuum, both immediately after production and after the heat treatment in the temperature range of 900…1300°C after roll bonding, are presented. The influence of thermal annealing on the interaction nature of the clad layers with each other and with the core tungsten material of the Ta-W-Ta solid-phase junctions with Nb interlayers obtained by vacuum hot roll bonding has been studied.

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INTRODUCTION

In the developing research nuclear facility (RNF) ‘Neutron Source Based on a Subcritical Assembly Controlled by the Linear Electron Accelerator’, the neutron source is equipped with a neutron-producing target device, the main unit of which is made of tungsten plates. The set of target plates is located inside the target assembly and is located in the center of the active zone. The electron beam with the energy of 100 MeV, the power of 100 kW, the beam current of 1 mA, and the power emitting density of 2.5 kW/cm^2 released from the target is incident on the water cooled assembly of these plates generating a neutron flux.

The main technological difficulties are connected with the manufacturing of the target plate assembly itself, since the target plates stand under intense fields of hard γ-radiation. As a result of radiation processes the physical and the mechanical properties of tungsten are being changed and the corrosion processes are being intensified when interacting with water. This circumstance requires the tungsten target plates protection with a corrosion-resistant protective layer of clad material among which the tantalum is the most suitable [1, 2].

The laminates are a specific type of junctions and play an important role in today's innovative solutions where multitask solutions are required. The creation of the refractory metals solid-phase junctions with high tightness which can prevent the radioactive products release from tungsten into the cooling water is possible by using the vacuum hot roll bonding (VHRB) [3-9]. The point of this method is heating and subsequent entire batch deformation consisting of materials layers to be joined by means of roll bonding under the vacuum conditions. Due to the melting and intensive oxidation absence of the materials to be joined, the vacuum hot roll bonding method provides the high strength, ductility and tightness of the dissimilar metals junction boundaries.

1. MATERIALS AND METHODS

The following materials of high purity were used in this work: tungsten, niobium and tantalum. The Ta-W-Ta solid-phase junctions specimens were obtained via hot roll bonding in vacuum at the temperature of 1300°C using the laboratory roll bonding equipment DUO-175 mill, designed and built at NSC KIPT.

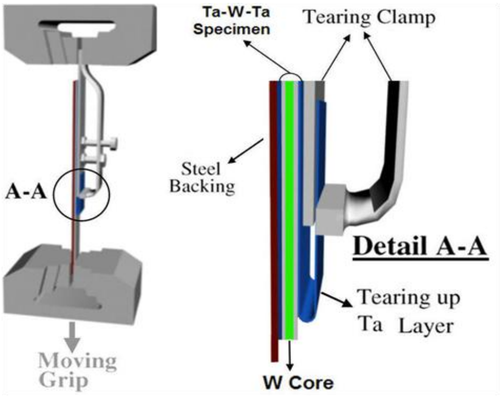


Fig. 1. The peeling test scheme

In order to do this tungsten and tantalum plates were cut out by the electric spark method on a ZAPbp BP-96ds machine to the dimensions of 65×65×3 mm, after which they were washed with acetone and subsequently chemically etched in the nitric and hydrofluoric acids solution. After that, the plates were assembled into a layered batch and the entire specimen was placed in a vacuum chamber for a subsequent heating to predetermined temperature. After reaching it, the specimen was put under the rolls by manipulator where the roll bonding process took place. The chamber pressure did not exceed $1 \cdot 10^{-6}$ mm Hg.

The layers adhesion quality was studied using the tantalum clad layer peeling tests according to the scheme used in [10] and presented in Fig. 1. The ‘Bi-00-201 Nano’ servo-hydraulic testing machine was used for this. The peeling strength was calculated as a load divided by the bond width – N/mm.

2. RESULTS AND DISCUSSION

2.1. THE INTERLAYERS APPLICATION

A common technological technique for the vacuum hot roll bonding, is the possibility of influencing the transition zone formation process by introducing of a intermediate layer material into the contact zone [11] in order to reduce the rolling temperature, the reduction pressure and accelerate the high-quality solid-phase junctions formation when dissimilar materials joining is taking place. From this point of view it is appropriate to use intermediate layers that facilitate the volumetric interaction processes development in the contact zone and activate the joining process that is the materials with high plastic properties possessing a lower plastic deformation resistance comparing to those of the base metals [4-7, 12-14]. As a rule the main requirement for such layers materials is the ability to form the solid solutions with both joining materials [9, 15]. The intermediate layer role is also to provide a strong junction due to partial compensation of internal stresses that arise in adjacent to the contact surface areas of the materials being joined as a result of their linear thermal expansion coefficients mismatch. When joining the dissimilar metals using soft interlayers the strength of a solid-phase joint, in the absence of brittle phases, depends on the base and intermediate materials mechanical properties, as well as on the geometric dimensions of the interlayer. The strength of a solid-state joint, in this case, can significantly exceed that of the intermediate layer material due to the so-called contact hardening, when the plastic deformations of the interlayer are restrained by stronger neighboring metals [16-18].

An approximate strength calculation of a solid-phase joint and the intermediate layer thickness choice can be carried out according to the formula presented in [19]:

$$\sigma_b = \sigma_b^{bm} - K \cdot \delta_M \cdot \gamma_M \cdot \chi \cdot 10^2, \qquad (1)$$

where ${\sigma_b}^{bm}$ – tensile strength of the base metal (less durable of the connected ones); K – depending on γ_M coefficient; ($\gamma_M = \dfrac{{\sigma_b}^M}{{\sigma_H}^M}$ – layer hardening parameter during the roll bonding process for a certain material layer with a strength of ${\sigma_H}^M$; at $\gamma_M \le 1.5$, K =2+4; at $\gamma_M > 1.5$, K =1+2); δ_M – the layer material relative elongation; $\chi = \dfrac{h}{b}$ – the intermediate layer relative thickness (h – the intermediate layer thickness; b – the laminate total thickness). Both the level of mechanical bonds in the interlayer and its joint strength with the base metal increase with the relative thickness are decreasing (χ).

2.2. SPECIMENS MICROSTRUCTURE

The positive result of the niobium interlayer using is explained by the fact that niobium possesses a tendency to form junctions with both tantalum and tungsten, and a wide solubility range both in tungsten and tantalum. Thermal and physical properties of these materials are close in a wide temperature range. The good compatibility of these metals makes it possible to obtain the multilayer materials by hot roll bonding in vacuum, which have the layers high bonding strength.

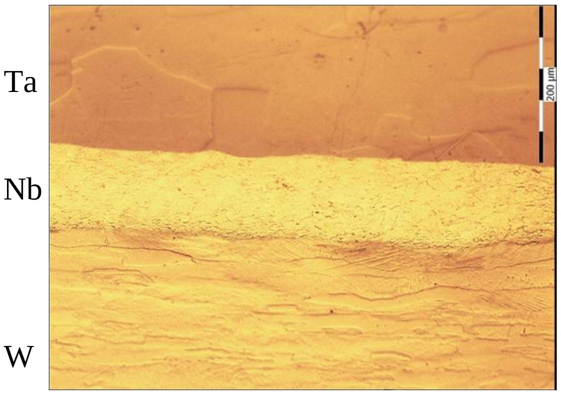


Fig. 2. Microstructure of the Ta-W-Ta (Nb interlayers) solid-phase joints specimens ×200

When using a niobium interlayer in multilayer solid-phase joints, metallographic studies revealed the absence of rolled samples interlayers breaks or gaps (Fig. 2). The absence of discontinuities in the niobium interlayer does not allow the tungsten and tantalum layers to come into direct contact, but makes them interact through an intermediate buffer zone, which is the niobium interlayer, that makes it expedient to use such an interlayer. This is due to the high ductility and low brittle fracture tendency of the niobium sheets and foils, especially in the annealed state.

2.3. STRENGTH CHARACTERISTICS

The peeling tests for the Ta-W-Ta specimens joined directly without interlayers, the result of which is shown in Fig. 3, showed the presence of a solid-phase bond at the Ta-W interface.

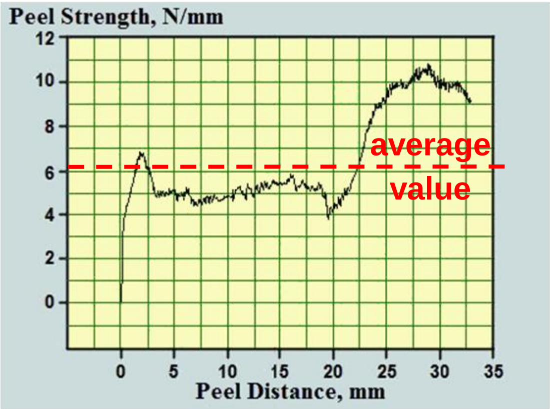


Fig. 3. Ta-W-Ta specimens peeling test result

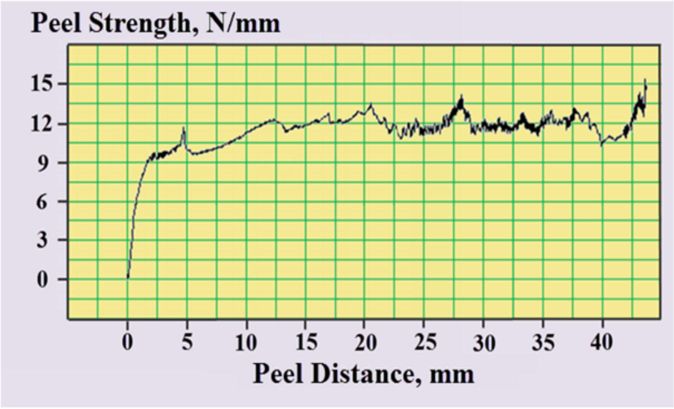


Fig. 4. Ta-W-Ta (with Nb interlayers) specimens peeling test result

The clad layer detachment passed through the tungsten layer. The average strength value is 6 N/mm. The diagram bursts indicate the uneven layers connection reaching a strength of 11 N/mm, indicating the areas of joined layers full-fledged interaction. The most part of the specimen has the strength value of 4 N/mm. Apparently this result is a consequence of the internal stresses occurrence arising at the tantalum-tungsten interface.

According to the clad layer peeling tests results for the specimens using niobium interlayers (Fig. 4), it can be seen that the bonding nature is uniform along the entire separation line. The average strength is approximately 12 N/mm, which exceeds the maximum strength values bursts of Ta-W-Ta specimens roll bonded directly without interlayers. This is explained by the high plasticity of the solid-phase joints boundaries when using a niobium interlayer in such laminates during the VHRB experiments due to the high plasticity of the niobium interlayer itself.

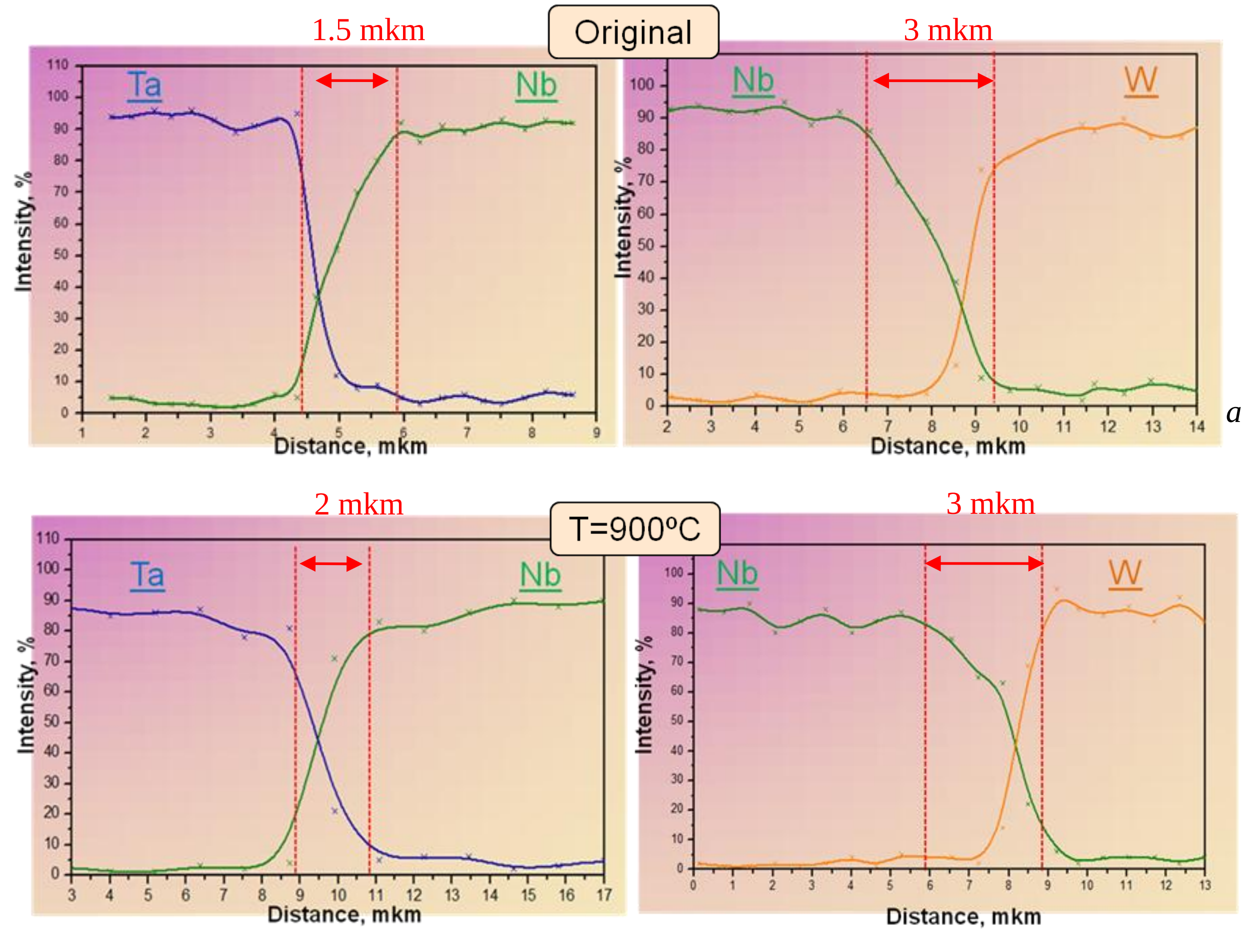
2.4. THERMAL ANNEALING EFFECT

The width of the solid-phase interaction zone (the atoms penetration of one material into another one) for each specific material under the certain conditions can be determined experimentally or estimated by calculation, knowing the mutual diffusion parameters and setting the temperature and the holding time of the material at the given temperature. Mutual movement of the laminate components masses also causes the changes in structure and properties of the bonding area. The width of such an interaction zone can be described by the expression [20], knowing the annealing time and temperature, and if it is known the mutual diffusion coefficient D dependency on the solid solution concentration for this temperature:

$$D(C,T) = D_0(C) = exp\left(-\frac{Q(C_0)}{RT}\right), \qquad (2)$$

here T – temperature by Kelvin; Q – diffusion process activation energy; D_0 – pre-exponential multiplier; R is the universal gas constant.

Evaluating the electron probe X-ray spectral analysis results of the original Ta-W-Ta specimens with Nb interlayers (Fig. 5) even before the thermal annealing it can be observed a significant difference in the concentration curves slopes through the interfaces: the niobium curve slope is less comparing to both the tungsten and the tantalum ones. This fact is the evidence of the preferential niobium advance through the interfaces even at the stage of joining materials due to its increased diffusion mobility in comparison with neighboring materials. The same effect was also experimentally observed in such material pairs like Cu-Pb and stainless steel-Cu and others and described in [5, 6, 9]. With the annealing temperature increasing this effect intensifies (Fig. 5), the niobium curves slope becomes smaller and consequently this result is connected with the transition zones widths increasing that is reflected in Table 1. It should be noted that the tungsten diffusion mobility increases slightly with the temperature increasing.



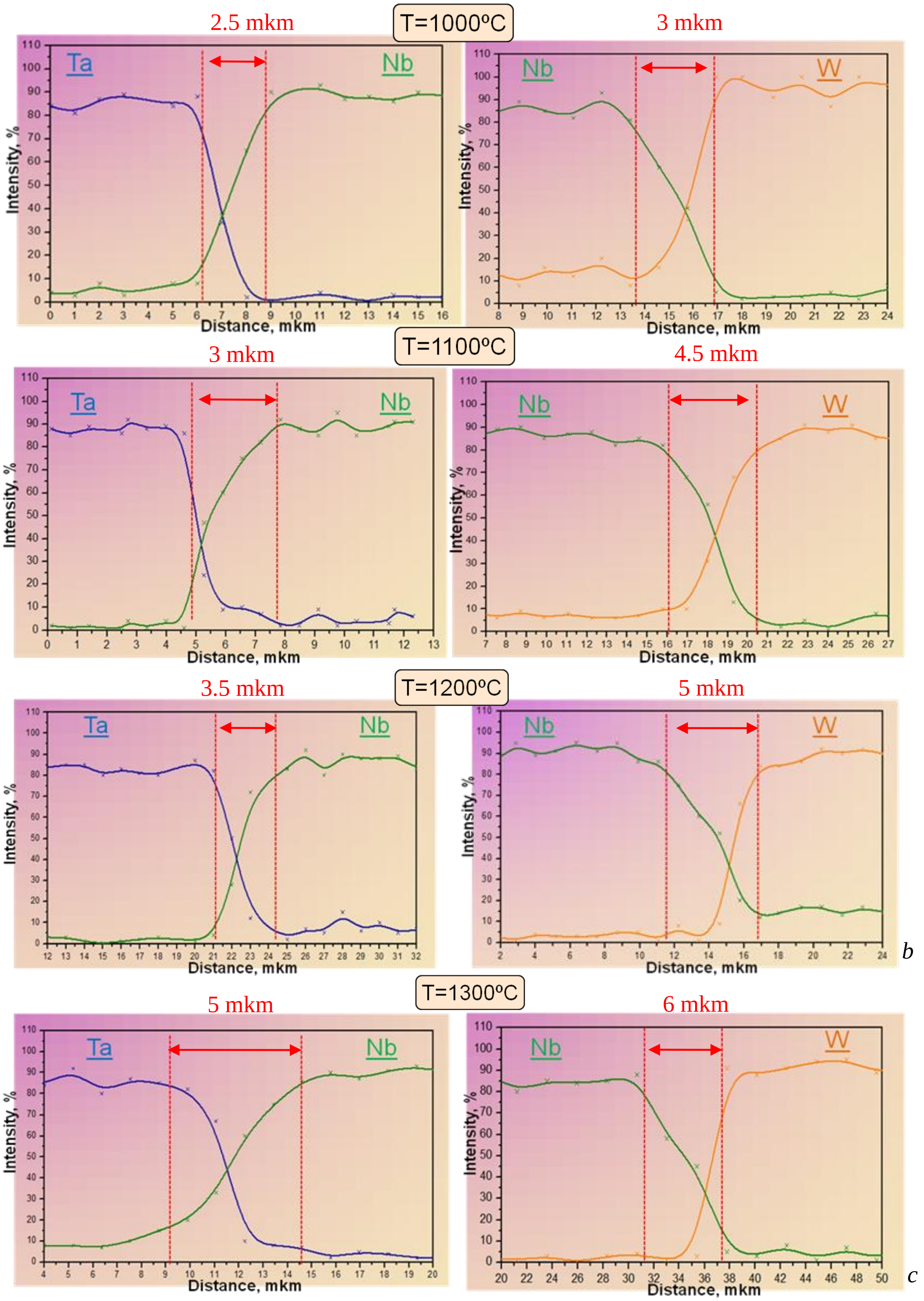


Fig. 5. X-ray electron probe analysis of Ta-W-Ta (using Nb interlayers) specimens originally after roll bonding (a); X-ray electron probe analysis of Ta-W-Ta (using Nb interlayers) specimens after thermal annealing at the range of 900…1200°C (after 100°C) for 1 hour (b); X-ray electron probe analysis of Ta-W-Ta (using Nb interlayers) specimens after thermal annealing at 1300°C for 1 hour (c)

Table 1

The diffusion zones width of Ta-W-Ta with Nb interlayers depending on the annealing temperature

	After Roll Bonding	Annealing Temperature				
		900˚C	1000˚C	1100˚C	1200˚C	1300˚C
Ta-Nb	1.5 mkm	2 mkm	2.5 mkm	3 mkm	3.5 mkm	5 mkm
Nb-W	3 mkm	3 mkm	3 mkm	4.5 mkm	5 mkm	6 mkm

The obtained mechanical tests results comparison on the tantalum-niobium solid-phase junction specimens after the different heat treatment reveals the difference in mechanical properties due to the deformation texture appearance. The texture presence in the sheet material leads to uneven deformation and the appearance of internal stresses during roll bonding [21]. Table 2 shows the grain sizes of tantalum and niobium in the tantalum-niobium layered material sheets after vacuum thermal annealing.

To carry out such studies the tantalum-niobium solid-phase junction specimens of 0.25 mm thick were prepared. The thickness of tantalum was 0.2 mm, and that of niobium was 0.05 mm. The sheets underwent the thermal annealing in a vacuum electric-resistance furnace at the temperature ranges of 1000…1300°C (after 50°C) for one hour. Uniaxial tensile tests were performed both on specimens whose tensile axis coincided with the rolling direction (longitudinal) and specimens whose tensile axis was perpendicular to the rolling direction (transverse).

Table 2

Tantalum and niobium grain sizes in the tantalum-niobium sheet laminates after vacuum thermal annealing

Annealing Temperature, °C	Grain Mean Diameter, mkm	
	Tantalum	Niobium
1000	19	15
1050	21	15
1100	24	16
1150	29	17
1200	30	25
1250	34	27
1300	38	60

As follows from Fig. 6 in the heat treatment temperature range of 1000…1100°C it is observed the material softening and the relative elongation rising. The optimal combination of strength and ductility (minimal tensile strength and ductility values and maximal relative elongation ones) are achieved at the annealing temperature of 1200°C. A further heat treatment temperatures increasing leads to a strength characteristics growth and ductility decline. A similar behavior of the mechanical characteristics dependencies on annealing temperatures also appears when testing the monolithic specimens of tantalum and niobium themselves [21].

The results comparison obtained on longitudinal and transverse specimens of the tantalum-niobium solid-phase junctions reveals a difference in mechanical properties due to the deformation texture appearance.

The mechanical characteristics of longitudinal and transverse specimens heat-treated at the temperatures of 1000…1100°C are somewhat different from each other. As the thermal annealing temperature rises to 1200°C, this difference in properties gradually decreases and at the temperatures of 1200…1300°C does not exceed 3…5% that is within the natural experimental data scattering.

Fig. 7 shows the microstructure of tantalum-niobium layered material sheets after various heat treatments. After thermal annealing at the temperatures of 1000…1050°C, the first recrystallized grains appear in both niobium and tantalum layers (see Fig. 7,a). Heat treatment at the temperature of 1200°C leads to a complete deformed structure rearrangement (see Fig. 7,b).

The recrystallization process of tantalum and niobium sheets in the tantalum-niobium-tungsten layered material composition occurs completely during heat treatment in vacuum at the temperature of 1200°C for one hour. Such heat treatment ensures the uniform fine-grained structure formation, internal residual stresses removal, high plasticity and quasi-isotropy of the tantalum-niobium solid-phase sheet junctions properties. A further heat treatment temperatures rising leads to the grains enlargement of both tantalum and niobium – the process of collective recrystallization takes place (see Fig. 7,c).

The above said can be summarized as follows. The multilayer solid-phase joint strength based on such refractory materials as tungsten and tantalum with niobium interlayers primarily depends on the strength characteristics of the tantalum and niobium clad layers, the mechanical properties of which first of all are determined by the heat treatment nature.

CONCLUSIONS

1. Using the niobium interlayers in the tantalum-tungsten-tantalum solid-state junction composition obtained at the DUO-175 mill it is possible to achieve the high-quality joining of these refractory materials layers by roll bonding due to the high compatibility of this interlayer with both tantalum and tungsten at such roll bonding parameters which can’t provide the high quality direct joining of these materials layers.

2. Niobium interlayers possess the sufficient plasticity for deformation without destruction under the conditions of the stress-strain state that is realized in a composition of refractory metals such as tantalum-tungsten-tantalum during their mutual high temperature vacuum roll bonding.

3. A positive thermal annealing effect of specimens on the diffusion zone width is shown, leading to its increase with the Nb predominant penetration both into the tantalum and tungsten target components.

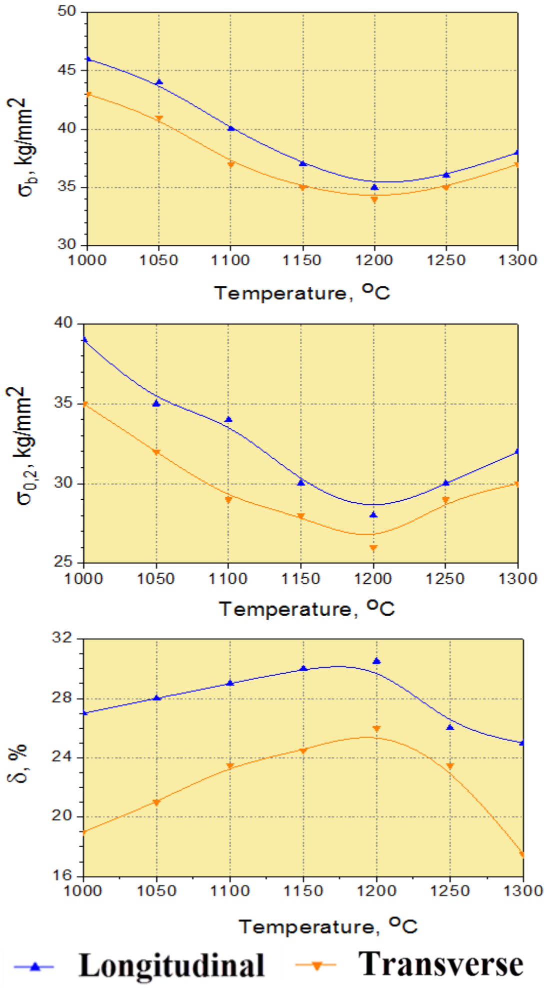


Fig. 6. Ta-Nb mechanical properties dependency on the annealing temperature

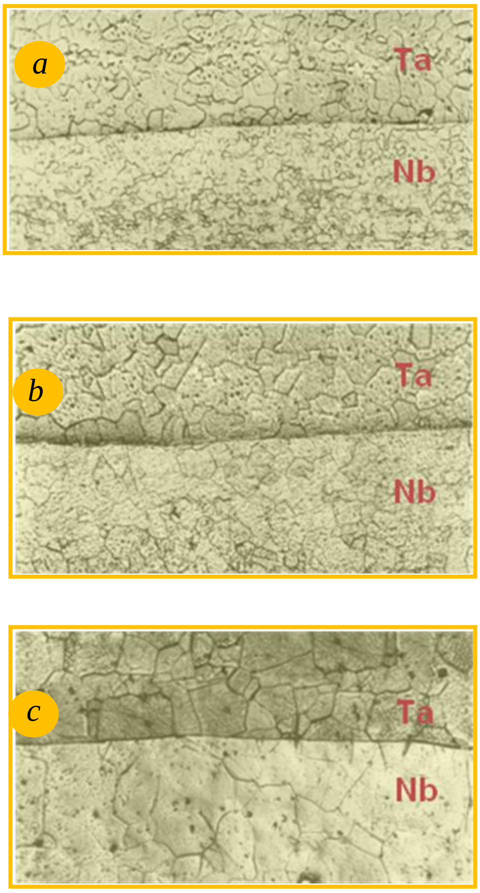


Fig. 7. The Ta-Nb solid-phase joint microstructure after thermal annealing for 1 hour at 1050°C (a), 1200°C (b), 1300°C (c). Magnification ×200

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ВПЛИВ ТЕМПЕРАТУРНОГО ВІДПАЛЮВАННЯ НА МІКРОСТРУКТУРУ ТА МЕХАНІЧНІ ВЛАСТИВОСТІ ТВЕРДОФАЗНОГО З'ЄДНАННЯ Ta-W-Ta ДЛЯ НЕЙТРОНОУТВОРЮЮЧИХ МІШЕНЕЙ ДОСЛІДНИЦЬКОЇ ЯДЕРНОЇ УСТАНОВКИ «ДЖЕРЕЛО НЕЙТРОНІВ»

О.О. Лопата, М.П. Домніч, Б.В. Борц, А.Ю. Зелінський, О.О. Пархоменко, В.І. Ситін, В.І. Ткаченко, І.О. Воробйов, О.Д. Підлісний, І.В. Паточкін, Л.І. Глущенко

Розглянуто варіант виготовлення нейтроноутворюючої мішені тантал-вольфрам-тантал з використанням прошарку ніобію методом гарячої вакуумної прокатки стосовно дослідницької ядерної установки «Джерело нейтронів». Представлені результати металографії та механічних випробувань зразків твердофазних з'єднань Ta-W-Ta, отриманих гарячою прокаткою у вакуумі як безпосередньо після отримання, так і термооброблених у діапазоні температур 900…1300°С після прокатки. Вивчено вплив температурного відпалювання на характер взаємодії плакованих шарів між собою та з матеріалом вольфрамового сердечника у твердофазних з’єднаннях Ta-W-Ta з прошарками Nb, які отримані методом гарячої прокатки у вакуумі.

ВЛИЯНИЕ ТЕМПЕРАТУРНОГО ОТЖИГА НА МИКРОСТРУКТУРУ И МЕХАНИЧЕСКИЕ СВОЙСТВА ТВЕРДОФАЗНОГО СОЕДИНЕНИЯ Ta-W-Ta ДЛЯ НЕЙТРОНООБРАЗУЮЩИХ МИШЕНЕЙ ИССЛЕДОВАТЕЛЬСКОЙ ЯДЕРНОЙ УСТАНОВКИ «ИСТОЧНИК НЕЙТРОНОВ»

А.А. Лопата, М.П. Домнич, Б.В. Борц, А.Ю. Зелинский, А.А. Пархоменко, В.И. Сытин, В.И. Ткаченко, И.А. Воробьёв, А.Д. Подлесный, И.В. Паточкин, Л.И. Глущенко

Рассмотрен вариант изготовления нейтронообразующей мишени тантал-вольфрам-тантал с использованием прослойки ниобия методом горячей вакуумной прокатки применительно для исследовательской ядерной установки «Источник нейтронов». Представлены результаты металлографии и механических испытаний образцов твердофазных соединений Ta-W-Ta, полученных горячей прокаткой в вакууме как непосредственно после получения, так и термообработанных в диапазоне температур 900…1300°С после прокатки. Изучено влияние температурного отжига на характер взаимодействия плакированных слоев между собой и с материалом вольфрамового сердечника в твердофазных соединениях Ta-W-Ta с прослойками Nb, полученных методом горячей прокатки в вакууме.