PECULIARITIES OF FORMATION OF STRUCTURE IN THE TRANSITION ZONE OF THE Cu–Ta JOINT MADE BY EXPLOSION WELDING

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Structure of the transition zone in a joint of metals having no mutual solubility was studied. It was determined that surface of the explosion welded Cu–Ta joint is not smooth, but contains protrusions with a size of about 5–10 μm. The transition zone of the joint consists of chaotically distributed non-melted regions of copper and tantalum, as well as zones of local melting of copper containing tantalum nanoparticles 30–50 nm in size. Two processes, i.e. formation of protrusions at the interface and local melting zones, determine stirring of the materials having no mutual solubility.

Keywords: *explosion welding, limited solubility, formation of joint, transition zone, local melting, nanoparticles*

At a wide variety of materials and welding conditions, an important problem in explosion welding is stirring in the transition zone near the interface in the welded joints. It is structure of the transition zone that determines the possibility of adhesion of two materials. Stirring is a complex process for different pairs of the materials joined, especially for the metal–intermetallic pair, as well as for metals having no mutual solubility.

Difficulties in welding metals to intermetallics are caused by the fact that, firstly, the latter are high-temperature chemical compounds with strong interatomic bonds, and, secondly, they are characterised by increased brittleness. Nevertheless, explosion welding provided a joint between titanium and orthorhombic titanium aluminide [1–9].

The problem of stirring is not less important for metals having no mutual solubility. To find out how important the presence of mutual solubility of the initial materials is the pair of copper and tantalum, which have no mutual solubility and form immiscible suspensions in liquid state, was chosen for explosion welding. The principle of formation of such suspensions is investigated in this study.

Tantalum of grade TVCh and copper M1 were used as initial materials. Welding was performed at Open Joint Stock Company «Ural Chemical Engineering Factory» (Ekaterinburg, Russia) by using different schemes and different parameters. After that the joints were selected for further investigations. The parallel arrangement of plates was used. Thickness of the tantalum plate was 1 mm, and that of the copper plate was 4 mm, the gap between the plates being 1 mm. The copper plate was a flyer plate with respect to the tantalum one, which lay on a support plate of titanium and steel with thickness of 4.5 + 20 mm. The detonation velocity was 2680 m/s. The plates collided at an angle of 5.22° at a velocity of 234 m/s. The choice of the welding parameters corresponded to a lower limit of weldability. These welding conditions traditionally applied by «Khimmash» are most cost effective because of a smaller charge and, hence, lower expenses for explosives. Moreover, this diminishes the impact of the detonation wave on the surrounding facilities.

Metallographic analysis was performed by the optical microscopy (OM) method using optical microscope «Epiquant» equipped with computation system SIAMS. Microstructure of the welded joints was examined by the transmission electron microscopy (TEM) method using transmission electron microscopes «JEM 200CX» and «CM-30 Super Twin», by the scanning electron microscopy (SEM) method using scanning electron microscopes «Quanta 200 3D» and «Quanta 600» with a maximal resolution of about 2 nm, as well as by using ion gun «Faschione 1010 ION MILL».

It can be clearly seen in microstructures of transverse sections of the transition zone of the Cu–Ta joint that the interface is heterogeneous (Figure 1, *a*) and has thickness of about 5–10 μm (Figure 1, *b*, *c*). It turned out that the interface of the investigated joint is not wavy but corrugated, which can be readily seen in microstructure of a longitudinal section of the Cu–Ta joint interface (Figure 2). Instead of parallel bands characteristic of the corrugated interface, the microstructure consists of spots of three colours: white, black and gray. This is indicative of the fact that the transition zone comprises chaotically distributed regions of three types. These regions in the longitudinal section of the Cu–Ta interface (see Figure 2) have

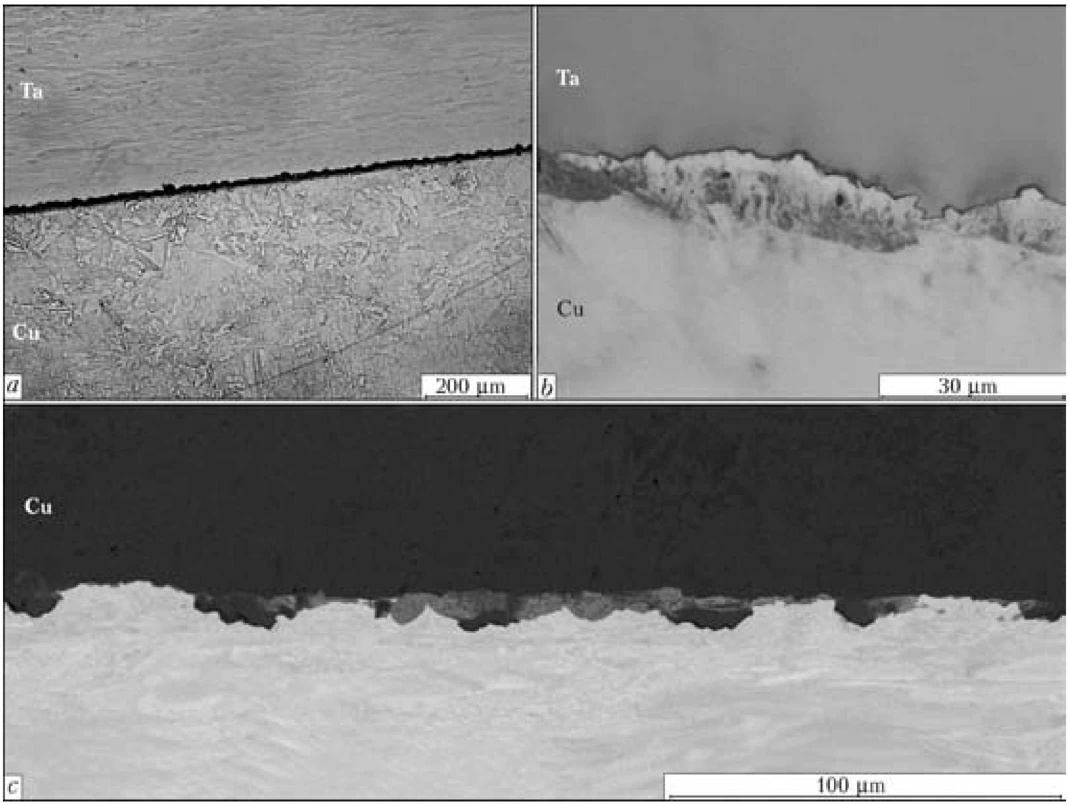

Figure 1. OM (*a*, *b*) and SEM (*c*) microstructures of transverse section of the Cu–Ta joint interface

approximately identical sizes (30–50 μm). Also, they can be seen in microstructure of the transverse section of the interface (see Figure 1, *c*).

In the Cu–Ta joint, the titanium to aluminide interface is not smooth. It comprises protrusions that can be readily seen in Figure 1 at different magnifications. Supposedly, it is these protrusions that determine the shape of the interface (see Figure 1, *a*) in the transverse section of the Cu–Ta joint interface and presence of regions of the three colours in the longitudinal section (see Figure 2). The protrusions (see Figure 1, *c*) are about 5–10 μm in size. Earlier [5] the protrusions were detected in the transition zone of the titanium–orthorhombic titanium aluminide joint. The depth of their penetration from one material into the other was tens of micrometres.

The data on chemical composition of the regions of three colours, which form the transition zone, were obtained by SEM on the basis of numerous measurements. It was determined that the white colour corresponds to the tantalum zone and black colour – to copper. Particular attention was given to a region of the gray colour, which was hereafter referred to as the gray zone. Figure 3 is extremely important for revealing its structure. This Figure shows at different magnifications the longitudinal section of the transition zone after complete etching out of copper. Tantalum particles having mostly nanometric sizes can be seen on the surface of tantalum (Figure 3, *b*).

In cases where copper was not etched out, it was detected at a magnification of 5000 that the concentration of both metals in the gray zone was approximately the same. However, as can be seen at a higher magnification (12,000), structure of the gray zone is heterogeneous and the concentration of both metals is different, i.e. some regions contain more copper, and other regions contain more tantalum. At the same time, the internal structure of the gray zone remains indiscernible until a higher magnification is used. Structuring of the gray zone begins at a magnification of 25,000 (Figure 4, *a*). The white zone of tantalum and the black zone of copper can be seen nearby. The microheterogeneous structure of the gray zone is even more pronounced in the microstructure revealed at a high magnification (Figure 4, *b*). This type of the structure evidences that the gray zone is a zone of stirring. The alternating microvolumes of the white

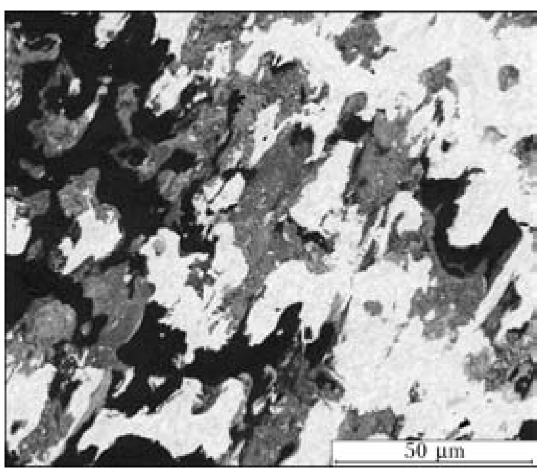

Figure 2. SEM microstructure of longitudinal section of the Cu–Ta joint interface (white spots – tantalum, black spots – copper, gray spots – mixture of tantalum and copper)

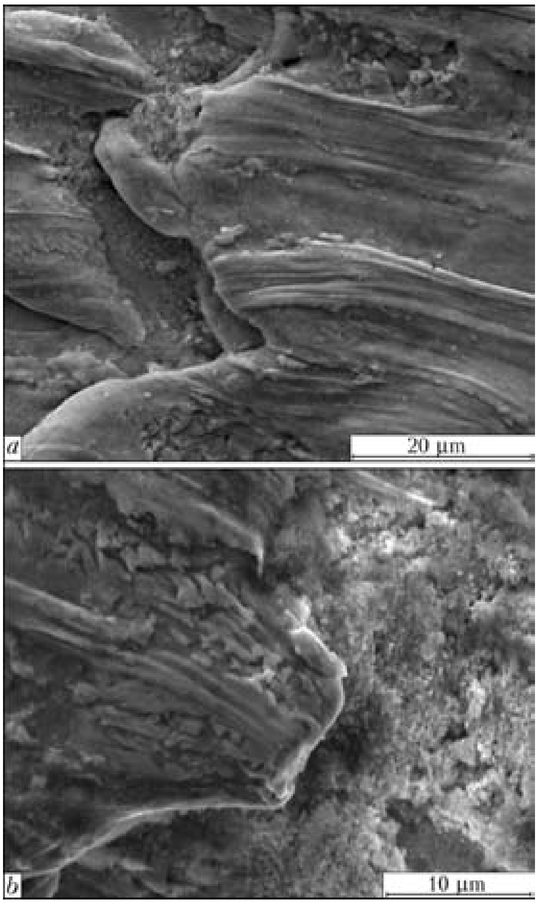

Figure 3. SEM microstructures of longitudinal section of the Cu–Tal joint transition zone (copper is fully etched out)

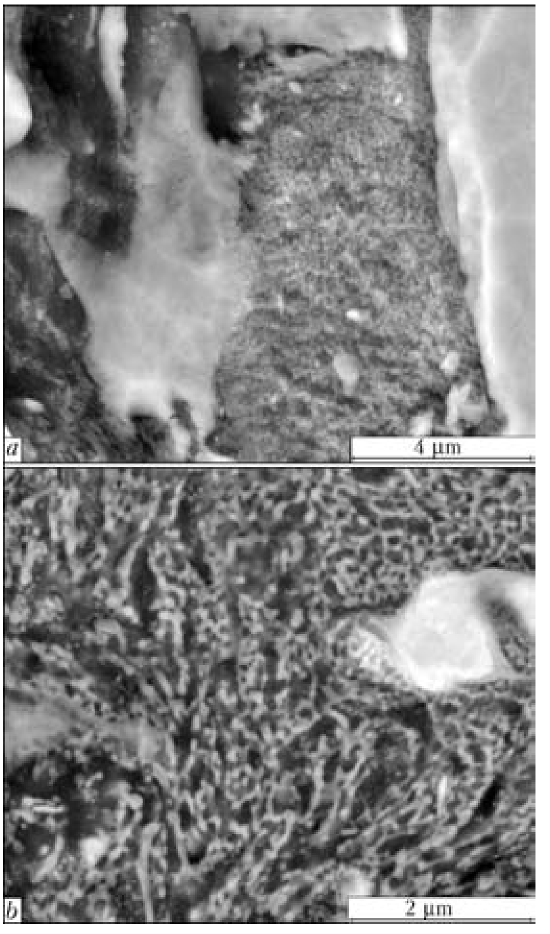

Figure 4. SEM microstructures of longitudinal section of the Cu–Ta joint at different magnifications

and black colours inside the gray zone are different. Only the black regions (copper) are elongated.

Examination of the Cu–Ta joints by the TEM method involves difficulties. Selection of reagents to prepare foils for such dissimilar materials is very complicated, as copper might be fully etched out under the effect of the reagents suitable for tantalum. In this connection, for this study the foils were made by using an ion gun.

Microstructure of the gray zone revealed by TEM gives an idea of the above microvolumes. Note that tantalum is of a dark shade in these images. Many dark particles with a shape close to the spherical one, the sizes of which are approximately 30–50 nm, can be seen in the light-field image (see Figure 5, *a*). Figure 5, *b* clearly shows a system of rings consisting of separate reflexes, which were decoded to belong to tantalum. Intensive spot-like reflexes are reflections of copper. It can be considered that nanoparticles of tantalum remain on its surface after etching out of copper (see Figure 3). Figure 6, *a*, *b* shows a light-field image of the gray zone at different magnifications. The dark particles can be seen here, like in Figure 5. Figure 6, *c*, *d* shows the dark-field images of the same joining zone as in Figure 6, *b*, in reflex <111> Cu and reflex <110> Ta, respectively. The particles of tantalum can be seen in both dark-field images. Electron-diffraction pattern reveals a substantial disorientation of both tantalum and copper particles, as well as the presence of individual amorphous interlayers having a characteristic microdiffraction, which consists of two diffusion rings.

Figure 7 also shows the particles of copper oxides. As indicated by the calculation of interplanar spacings, they sufficiently accurately coincide with the values obtained for Cu_2O. The possible cause of formation of copper oxides is as follows. When the plates collide, a shock-compressed gas saturated with fine copper particles is formed in the welding gap ahead of the contact point. It can be suggested on the basis of the data of study [10] that copper oxides are formed as a result of their burning. The surface layer of tantalum participating in formation of the joint has a higher thermal conductivity than copper, and is heated to a much lower temperature than copper. Possibly, this is a cause of the absence of tantalum oxides.

In addition to the gray zone, the transition zone comprises the above-mentioned zones of copper and tantalum (see Figure 2). As indicated by the analysis performed by the TEM method, all these zones do not experience melting and have a structure typical of intensive plastic deformation: both materials are characterised by the presence of a band structure and recrystallised regions. The size of grains in these regions is about 100–300 nm, which is several orders of mag-

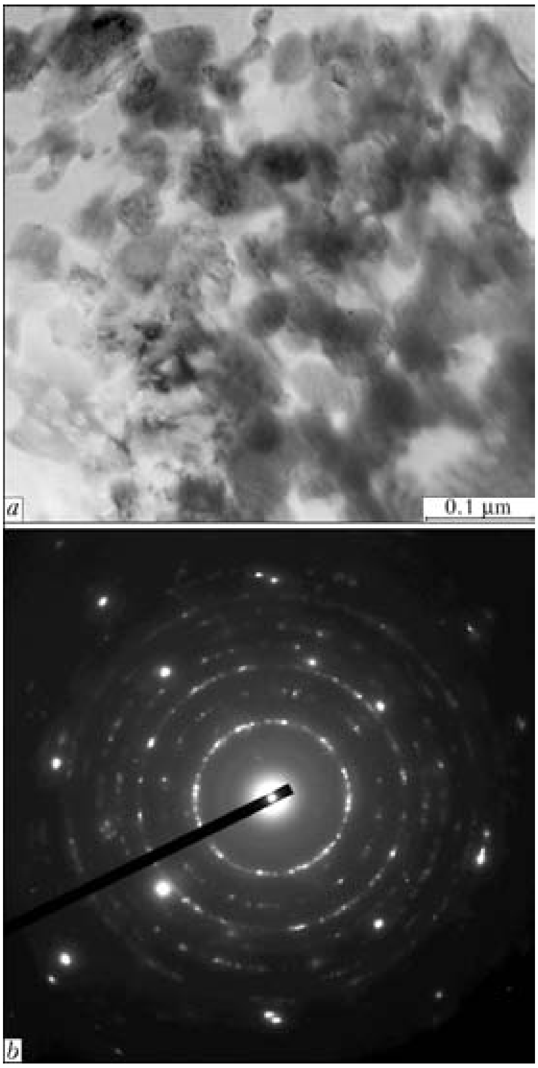

Figure 5. TEM microstructure of gray zone of the Cu–Ta joint: *a* — light-field image; *b* — microdiffraction pattern

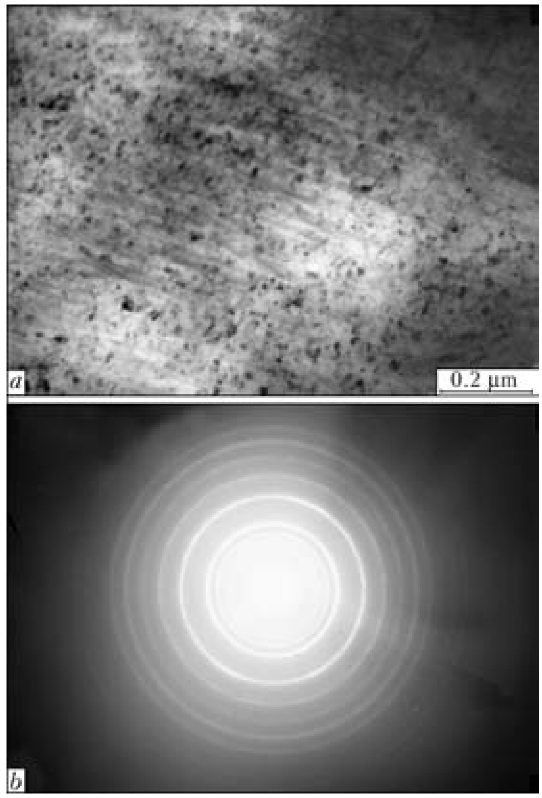

Figure 7. Light-field TEM image of copper oxides in the Cu–Ta joint (*a*) and microdiffraction pattern (*b*) [8]

nitude smaller than the initial size (approximately 100 μm). Also, a high density of dislocations and twins is fixed.

Different notions of the mechanisms of weldability are available [11]. According to one of them, the process of formation of a welded joint requires only atomic-pure and atomic-smooth surfaces. Moreover, because of a high pressure the process occurs in a solid state

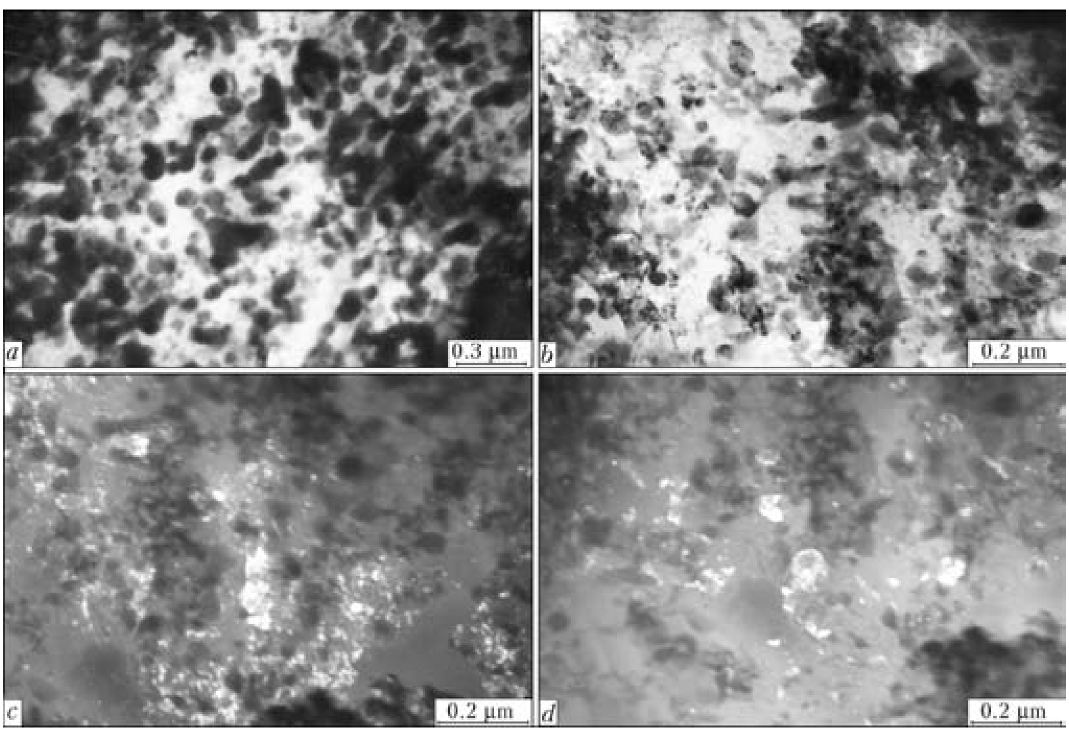

Figure 6. TEM microstructure of gray zone of the Cu–Tal joint (*a*–*d* — see in the text)

and excludes melting. According to other notions, good weldability is provided by formation of a continuous submicron layer, which solidifies extremely rapidly. Study [11] contains descriptions of numerous models of stirring, and some of them are discussed in study [5].

We think that stirring at the atomic level does not take place in materials having no mutual solubility. Otherwise they could form true solutions. This fact forms the basis of further development of the notion of colloidal systems in regions of local melting, which was confirmed by the structural examination results obtained in this study. Furthermore, the notion that the weldability requires only atomic-smooth surfaces does not agree with the presence of protrusions at the interface (see Figures 1 and 2). Formation of protrusions near the interface, compared to natural roughness of the initial samples, proves that they are formed particularly during the welding process. As a result, regions filled with any of the materials welded can be found on each side near the interface, which is indicative of their interpenetration. In fact, protrusions act as «nails» providing adhesion of the surfaces. It's true, especially if we allow for a high microhardness of tantalum (about 3000 MPa), which is 3 times as high as that of copper. The protrusions are formed as a result of a strong external effect of a number of factors: high plastic deformation (including pressure, shear components, turning moments of stresses, heterogeneity of deformation, etc.), friction of the surfaces, effect of the cumulative jet, etc.

The developed plastic deformation is characterised by the fact that it always occurs in bursts [12]. So, we suggest the following scenario of formation of gray zones as regions of local melting in locations where the external effect is strongest. Here there is no continuous molten layer, which was mentioned above. We give just the rounded values of temperature, as we know its different values, which differ because of composition and methods used to produce materials. At a normal pressure the boiling temperature of tantalum is T_b^{Ta} = 5700 K, its melting temperature is T_m^{Ta} = 3300 K, while the boiling temperature of copper is T_b^{Cu} = 2800 K, and its melting temperature is T_m^{Cu} = = 1400 K. At a high pressure the characteristic temperature values increase, but it can be assumed that their sequence persists: $T_b^{Ta} > T_m^{Ta} > T_b^{Cu} > T_m^{Cu}$.

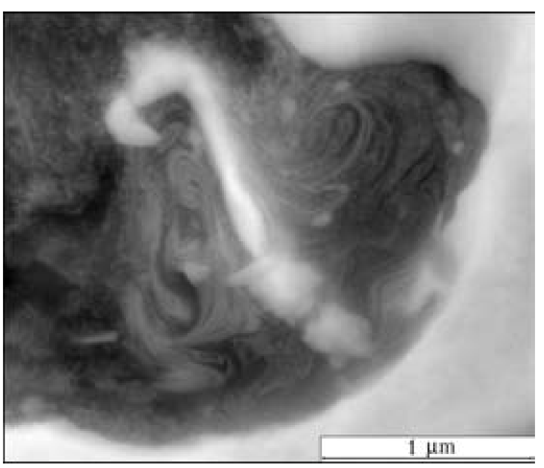

Figure 8. SEM image of vortices in the Cu–Ta joint

Assume that in explosion welding the temperature that is a bit higher than T_m^{Ta} is achieved in some individual regions, thus causing melting of tantalum [13]. Boiling of copper (so-called boiling with forced circulation) occurs at the said temperature, leading to formation of a vapour, which contains the scattered tantalum droplets having a spherical shape, this minimising their surface energy. The vapour may also contain a small amount of the copper droplets. In transition to a temperature below T_m^{Ta}, the tantalum droplets will immediately solidify, and will not change thereafter. Below T_b^{Cu}, neither vapour nor droplets of copper are formed, but only its uniform melt is produced. Because of a short duration of the welding process, these transitions occur during a time of about a microsecond. In transition through T_m^{Cu} the copper melt solidifies at a residual temperature.

Vortices similar to those taking place in many joints could be formed in the copper melt. The titanium–orthorhombic titanium aluminide joint, which we investigated earlier, contains local zones with a laminated structure in the form of concentric rings that copy in shape the contour of a cavity wherein melting occurred. The size of the vortex zones is about 50–100 μm. Optical microphotographs of the vortex zones are given in studies [5, 6, 8]. However, no vortices are seen in optical microphotographs of the Cu–Ta joint. The vortices are seen in the SEM microphotographs obtained at high magnification (Figure 8), but they do not look like the above vortices either in shape or in size (about 0.5 μm), and appear very rarely. The issue of their origin is unstudied as yet.

The key assumption used as a basis of our scenario is the possibility of melting of tantalum. This possibility is really implemented in many joints on refractory metals, such as Mo–Fe, Mo–Cu, Nb–Ti and Nb–Zr [14]. Note the following fact [14]: vortices are formed in welding of niobium to titanium alloys, which evidences melting of niobium.

What is important is that in the case investigated the gray zone is a dispersed system, the different stages of formation of which are characterised by changing of the dispersed medium and dispersed phase (this fact was mentioned above). In any case, the dispersed phase is a finely scattered material (tantalum) with linear sizes smaller than 100 nm, whereas the dispersed medium is a homogeneous material (copper) with the dispersed phase distributed in it. Dispersed systems with such characteristics are usually called colloidal [14]. They take an intermediate position between true solutions, on the one hand, and coarse-dispersed systems (emulsions and suspensions), on the other hand.

Therefore, the gray zone is a solidified colloidal system consisting of two immiscible phases.

Nevertheless, there is an alternative version of the scenario — scattering of tantalum in solid state into particles, which is a case of explosion welding of orthorhombic titanium aluminide to titanium. Figure 7 shows a fragmented layer consisting of the orthorhombic aluminide particles, and Figure 8 shows penetration of these particles into the zone of local melting of titanium. Comparison of the two joints clearly indicates a difference between the aluminide and tantalum particles. Thus, the aluminide particles, unlike the tantalum particles, have an irregular shape, rather than the spherical one. Moreover, they have micron and submicron sizes, this being an order of magnitude larger than size of the tantalum particles, which in fact are nanoparticles. It is the spherical shape of the tantalum particles and their small sizes that allow us to prefer the first version of the scenario.

Therefore, the choice of the scenario is reduced to the choice between two versions of formation of the tantalum particles, i.e. from the molten or solid phase.

To explain why scattering of tantalum in the solid state is difficult, it should be taken into account that brittleness of the materials leads to their refining under a strong external effect. Tantalum is a ductile material, capable of deforming to a high degree of deformation (approximately by 45 %, tension at room temperature). In this connection, mechanical refining of tantalum to nanoparticles can hardly be expected, considering that even a more brittle orthorhombic aluminide does not transform into a powder during explosion welding. In addition, we conducted a simulation experiment. Plates of tantalum and orthorhombic titanium aluminide (alloy VTI-1) were subjected to forging. Omitting details of the experiment, note that we used pneumatic hammer M-413 with impact energy of 5300 J, and placed both plates into one jacket of titanium foil. When we opened the jacket, we found out that the tantalum plate became flat and the aluminide plate crumbled.

The data obtained allow a conclusion that the transition zone of the joint consists of the copper and tantalum regions that experienced no melting, as well as zones of local melting of copper, which contain tantalum nanoparticles in the form of solidified droplets.

The local zones, where the external effect was strongest, require special consideration, as strength of the joints is determined particularly by these zones. Melting is one of the efficient processes leading to dissipation of the kinetic energy of the flyer plate. The local melting zones are «inserts» in the transition zone, having a different structure compared to the surrounding. It can be assumed that the resulting structure of the zones provides strengthening of the entire joint, rather than causing its brittleness. Microhardness was measured at different points of the transition zone, and it was established that microhardness of copper and tantalum increased insignificantly compared to the initial values. However, microhardness of the gray zone is over 4000 MPa, which is approximately 1000 MPa higher than that of tantalum. The observed effect is a result of dispersion hardening of copper due to the tantalum nanoparticles.

The process of formation of protrusions at the interface and local melting zones is determined by stirring of the materials having no mutual solubility. Mutual solubility is not required for interpenetration of one material into the other by way of formation of protrusions. Interpenetration of the materials in the local melting zones is provided by their scattering into droplets and intensive stirring owing to circulation of the melt, which prevents separation of the colloidal system into components during the time period required for solidification of the refractory liquid.

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