

## ANALYSIS

### OF LOW-CARBON STEEL/TANTALUM INTERFACE AFTER EXPLOSIVE WELDING

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The article discusses the morphology, microstructure and chemical composition of the boundary layer in explosive welding of low-carbon steel and tantalum plates. Pools of melt composed of both metals and having a heterogeneous chemical composition appear on the boundary layer. This phenomenon is the result of incomplete mixing of both: molten metals due to rapid cooling and solidification. Within the pools of alloy, a large number of non-metallic oxide inclusions, products of deoxidation of molten alloys, can be found. The non-metal inclusions are round (0.1 to 2  $\mu\text{m}$  in diameter) and vary in both: size and chemical composition. The main component of the non-metal inclusions is  $\text{Ta}_2\text{O}_5$ , which also contains iron, manganese, and aluminium oxides. The number of non-metal inclusions is unusually high for low-carbon steel and comparable to the number of non-metal inclusions in welds. Oxygen which causes the large number of deoxidised products does not originate in the low-carbon steel, as this steel is relatively clean. The most probable source of oxygen is the oxide scale from the surface of the low-carbon steel, which was poorly cleaned prior to welding.

**Key words:** *explosive welding, deoxidation, boundary layer, low-carbon steel, tantalum*

**Analiza međupovršine niskougličnog čelika/tantal nakon eksplozivnog zavarivanja.** Članak raspravlja o morfologiji, mikrostrukturi i kemijskom sastavu pograničnog sloja pri eksplozivnom zavarivanju traka niskougličnog čelika i tantala. Područje zone taljenja sastavljeno je od oba metala i na graničnom sloju pojavljuje se heterogeni kemijski sastav. Ova pojava je posljedica nepotpunog miješanja oba rastaljena metala, zbog naglog hlađenja i očvršćivanja. Unutar područja spajanja legure može se utvrditi veliki broj nemetalnih oksidnih uključaka, produkata dezoksidacije rastaljenih legura. Nemetalni uključci su okrugli (0.1 do 2  $\mu\text{m}$ ) i variraju u veličini i kemijskom sastavu. Glavna komponenta nemetalnih uključaka je  $\text{Ta}_2\text{O}_5$ , koji također sadrži željezne, manganove i aluminijske okside. Broj nemetalnih uključaka je neobičajeno velik za niskouglični čelik i usporediv je s brojem nemetalnih uključaka u zavarima. Kisik koji dovodi do nastanka velikog broja produkata dezoksidacije ne potiče iz niskougličnog čelika jer je čelik relativno čist. Najvjerojatniji izvor kisika je površinski oksidni sloj niskougličnog čelika koji je slabo očišćen prije procesa zavarivanja.

**Ključne riječi:** *eksplozivno zavarivanje, dezoksidacija, granični sloj, niskouglični čelik, tantal*

## INTRODUCTION

The work deals with deoxidation [1, 2], a high rate chemical reaction, as it occurs in isolated pools of molten alloy during explosive welding of large low-carbon steel and tantalum plates. During the same procedure of welding of large plates of low-carbon steel with either titanium or aluminium, this phenomenon was not observed [3-4].

B. Kosec, L. Kosec, V. Gontarev, P. Škraba, Faculty of Natural Sciences and Engineering, University of Ljubljana, Ljubljana, Slovenia, S. Petrović, Exploengineering, Ljubljana, Slovenia, G. Kosec, Institute of Metals and Technology, Ljubljana, Slovenia, M. Gojić, Faculty of Metallurgy, University of Zagreb, Sisak, Croatia

Large plates of this type are important to the manufacture of chemical reactors. This type of material is resistant to corrosion (tantalum), while the thicker steel base is the carrier of important mechanical properties. Compound materials of this type of composition are both structurally efficient and economical.

Due to the properties and size of the basic components (steel, tantalum), explosive welding is, at present, the only efficient way of joining the two metals into the assembled material [5]. Large dynamic forces and pressures on the interface are the particularity of this type of welding, where vast heat sources are generated locally, increasing temperatures above the melting point of both metals. This is the basis

for the formation of alloys. Large quantities of heat conductive metal cause rapid and large temperature changes and effect the development of microstructures. Insulated pools of molten alloys form a significant part of the interface and contribute to the strength of the joint. After solidification, pools of alloyed molten steel and tantalum become part of the vortice shaped interface of both metals. This type of interface is less distinctive in other explosively welded pairs, such as titanium - aluminium, or titanium - low-carbon steel.

## EXPERIMENTAL WORK

Manufacturing conditions of the investigated bimetallic materials are not known. However, they are known for assembled plates of low-carbon steel - titanium, where the estimated pressures on the contact surface are about 50 - 100 kbars, and the duration of dynamic contact is about  $10^{-4}$  to  $10^{-3}$  seconds [6].

Samples were analysed using the optical microscope and the scanning electron microscope (SEM) equipped with the energy dispersive spectrometer (EDS). Corrections of the quantitative measurements of chemical composition were performed using ZAF software.

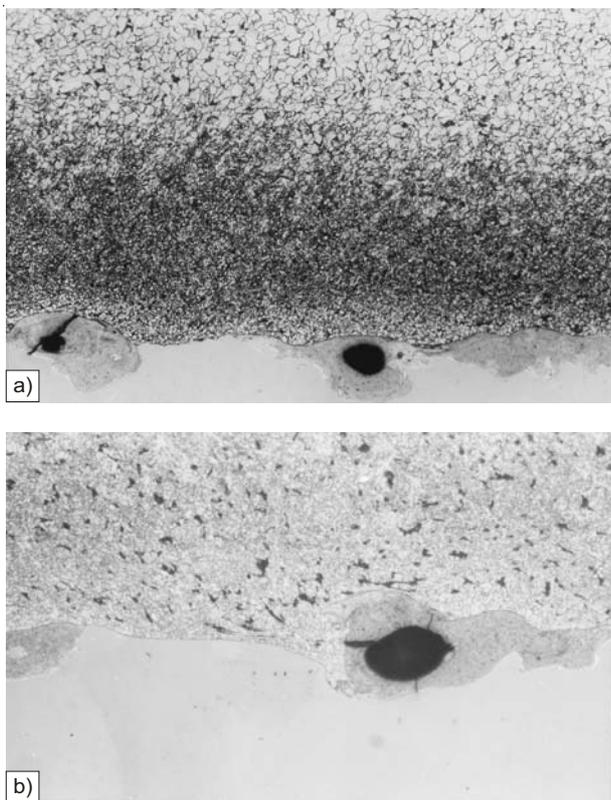


Figure 1. Explosively welded metal interface, top: low-carbon steel, bottom: tantalum. The pools of molten alloy (black). Magnification 100x (a). Magnification 200x (b)  
Slika 1. Međupovršina metala zavarenog eksplozijom, gore: niskouglični čelik, dolje: tantal. Područje rastaljene legure (crno), Povećanje 100x (a), Povećanje 200x (b)

The contact surface between the steel and tantalum consists of periodically repeating elements of wave motion with relatively small amplitudes and large wavelengths. The interface consists of islands of heterogeneous alloy (formed from the low-carbon steel and tantalum) and intermediate regions of both metals in direct contact [7].

Relatively large spherical pores were found in the remelted islands, many of which had one or more cracks. On the steel side of the weld, temperature effects on the solid state transformation could be observed from the interface to a distance of 0.3 mm. It was not possible to estimate such influences (Figure 1.) on the tantalum side.

The islands represent the alloys formed from locally melted steel and tantalum. Within the islands formed on the tantalum side, unreacted particles of tantalum were also found (Figure 2.). The alloy islands were chemically het-

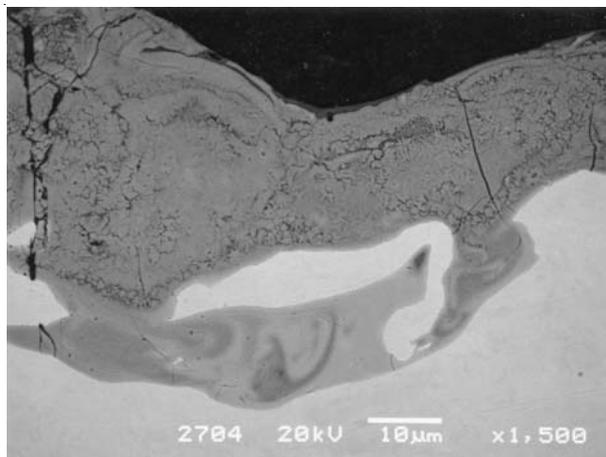


Figure 2. Nucleation of non-metal inclusions at the melt-tantalum interface; SEM. Magnification 1500x

Slika 2. Nukleacija nemetalnih uključaka na međupovršini metal/tantal; SEM. Povećanje 1500x

erogeneous. The patterns showing the currents produced by different chemical compositions of the melted metal were well visible on the back scattered electron (BSE) images (Figure 3.).

On the microscopic level, dynamic forces of explosive welding caused intensive mixing and eddying of metal within the pools. Despite both the intensive mixing of the liquid metal and the possibility of diffusion occurring between molten regions of varying chemical composition, the alloy remained chemically heterogeneous because of rapid solidification. On the basis of dendrite arm spaces (DAS), a cooling rate of  $10^4$  K/s was estimated. The stability times of the pools were so short that homogenisation could not occur despite of the complete miscibility of the components in the liquid state.

Large numbers of non-metallic inclusions, resulting from the deoxidation of the melt, were the particularity of the explosively welded steel - tantalum alloys. Typically, the majority of these non-metallic inclusions nucleated on

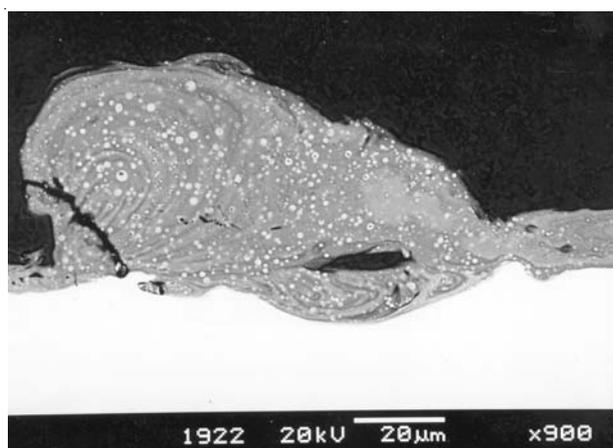


Figure 3. Pool of molten alloy. The pattern in the chemically inhomogeneous alloy shows the movement of the melt. White regions: tantalum, dark regions: low-carbon steel; SEM. Magnification 900x

Slika 3. Područje rastaljene legure. Oblik kemijske nehomogenosti legure pokazuje kretanje taline. Bijela područja: tantal, tamna područja: niskougljični čelik; SEM. Povećanje 900x

the melt-tantalum interface, while none could be found on the steel-melt interface (Figure 2.). These inclusions have characteristic semicircular (hemispheric) shapes, which points to relieved heterogeneous nucleation on the solid-liquid interface. However, the deoxidised inclusions within the melt were spheroidal.

The shape, distribution and number of non-metallic inclusions were very similar to those formed during deoxidation of melts through classical welding, such as electroslag welding. However, in the latter case, the size of the inclusions was substantially larger. However, many

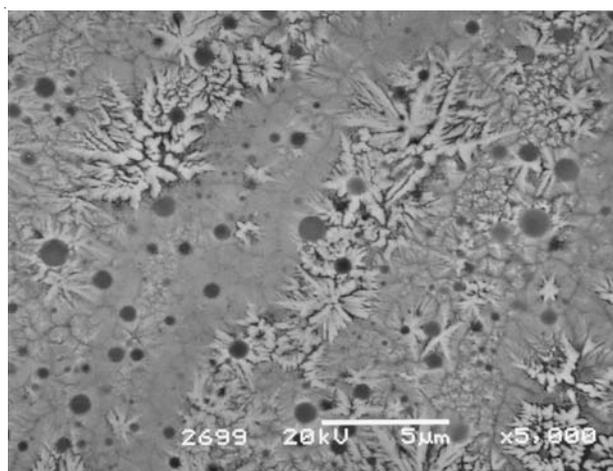


Figure 4. The microstructure of the molten alloy at the steel-tantalum interface. Circular particles are deoxidised non-metal inclusions; SEM. Magnification 5000x

Slika 4. Mikrostruktura rastaljene legure na međupovršini čelik/tantal. Kružne čestice su dezoksidirani nemetalni uključci; SEM. Povećanje 5000x

inclusions were found to be heterogeneous nuclei for alloy solidification. In this case, the solidification conditions of the melt dictated the formation of dendrites and, to some degree, the formation of cells. In many dendrites, the deoxidation inclusions behaved as heterogeneous nuclei. The measured diameters of deoxidation inclusions ranged from 0.1 to 2  $\mu\text{m}$ . The chemical compositions of the alloy and non-metallic inclusions were determined on characteristic sites using EDS (Figures 4., 5.).

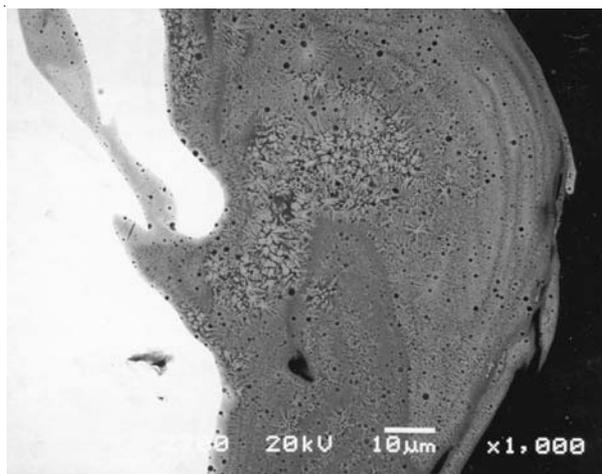


Figure 5. Region of alloy containing cells and dendrites. Circular particles are deoxidised products; SEM. Magnification 1000x

Slika 5. Područje legure koje sadrži ćelije i dendrite. Kružne čestice su produkti dezoksidacije; SEM. Povećanje 1000x

Non-metallic inclusions, which were the products of deoxidation, had complex chemical compositions. The relative differences in chemical composition of individual inclusions were considerable [8]. The measured concentration intervals of single elements in non-metallic inclusions, with the exception of oxygen, were as follows [mass contents in %]: Ta 46 - 74, Al 4 - 7, Mn 1.4 - 1.8, and Fe 4 - 19.

It was also possible to determine the differences in chemical composition of deoxidation inclusions by means of back scattered electron (BSE) images. Smaller inclusions were darker, while the large ones were lighter. This showed that the larger inclusions had a higher concentration of tantalum than the smaller ones. Quantitative and semi-quantitative chemical analyses verified these results (Figures 6., 7.).

In the region of cellular microstructure, the chemical composition of alloys, which solidified in the form of dendrites, were determined. In the region of dendrites (sizes range from 1 to 5  $\mu\text{m}$ ), the measured concentration range of tantalum was 45 - 52 mass content in % (21 - 25 atomic mass fraction in %), while the remainder was iron. In the region of the cellular microstructure, the measured concentration range of tantalum was 34 - 37 mass contents in %. According to the Fe-Ta equilibrium phase diagram [9], the chemical compositions of both regions were between

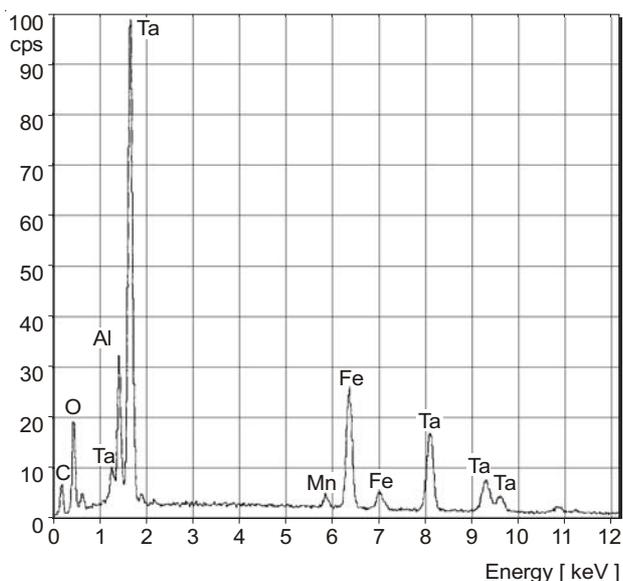


Figure 6. **Qualitative chemical composition of the non-metal inclusion (products of deoxidation) within the molten alloy (darker region); EDS**

Slika 6. **Kvalitativni kemijski sastav nemetalnog uključka (produkti dezoksidacije) unutar rastaljene legure (tamnije područje); EDS**

the intermetallic composition of  $\text{Fe}_2\text{Ta}$  (closer to the dendrite composition) and the eutectic on the iron end of the phase diagram. According to the phase diagram, melting points of corresponding alloys, at a pressure of  $p = 1$  bar, range from 1700 to 1500 °C.

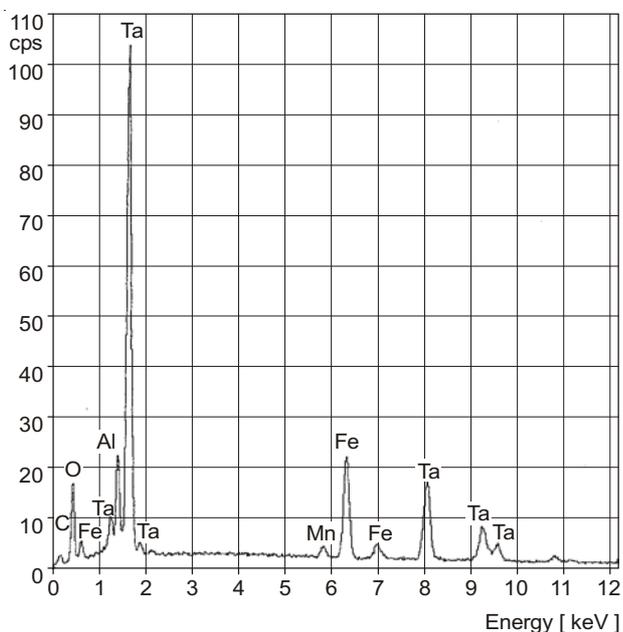


Figure 7. **Qualitative chemical composition of the non-metal inclusion (products of deoxidation) within the molten alloy (lighter region); EDS**

Slika 7. **Kvalitativni kemijski sastav nemetalnog uključka (produkti dezoksidacije) unutar rastaljene legure (svjetlije područje); EDS**

The microstructure of the alloy formed by melting was very fine and not in equilibrium. This, along with the mentioned geometric resolving power of the electron beam, prevented unequivocal in situ analysis of the microstructure of the constituents. Therefore, the most accurately measured values are the defined averages of a number of microstructural constituents. Microhardness within the islands of the remelted alloy was approximately 680 to 880 HV, in tantalum (close to the interface) 170 to 180 HV, and in steel about 180 HV.

Among the elements present in the molten alloy, i.e. in the interface vortices, tantalum is the strongest deoxidiser. As an example, minimal oxygen potential of  $\text{Ta}_2\text{O}_5$  formation is only  $p_{\text{O}_2} = 1.2 \times 10^{-35}$  bars at 1600 °C. At the same temperature, the required oxygen potential for  $\text{Al}_2\text{O}_3$  formation is  $p_{\text{O}_2} = 2 \times 10^{-20}$  bars, while for  $\text{FeO}$ ,  $p_{\text{O}_2} = 2.1 \times 10^{-8}$  bars. The deoxidation products are complex oxides of Ta, Fe, Al and Mn. As can be seen from the data, tantalum is by far the strongest deoxidiser. Due to the high concentration of tantalum in the alloy, its deoxidising effect is increased, which also holds for iron.

The difference in chemical composition of non-metallic inclusions can be explained either by chemical inhomogeneity of the melt or through the evolution of its chemical composition during the growth of non-metallic inclusions. The high deoxidising power of tantalum also has a significant influence on the kinetics of deoxidation, where the chemical reaction is dominant. Alloys, which were formed in vortices on the interface during the explosive welding of steel to titanium and steel to aluminium, were also investigated. In both cases, the deoxidising effect demonstrated by oxide inclusions was not observed in the SEM.

The maximum concentration of tantalum in non-metallic inclusions was approximately 50 % higher than the highest local concentrations of tantalum in the heterogeneous alloy.

## DISCUSSION

A large number of non-metal inclusions resulted from the deoxidation of molten alloy. While this number was also high for non-metal inclusions in low-carbon steel, the metallurgical analysis showed no inclusions in tantalum. Due to this discrepancy, the possibility that the non-metal inclusions within the molten islands of the alloy did not result from deoxidation of the molten steel, or rather from the oxygen present in both metals, must be considered. This discrepancy is further reinforced by the fact that comparable islands were not observed during the welding of low-carbon steel to either titanium or aluminium.

It is therefore necessary to look for an external oxygen source large enough to result in deoxidation and deoxidation products on the observed scale. Islands of molten alloy could become oxygen-rich from oxides located on the

surface of either low-carbon steel or tantalum. The steel surface was not cleaned of oxide scale prior to welding. However, no oxides were found on sample slices made through the boundary between steel and tantalum. It seems most probable that the oxides were either very thin or locally distributed and were therefore either expunged or absorbed into the molten islands of alloy. Direct evidence of the existence of oxides at the boundary between steel and tantalum was not discovered. Indirectly, however, their presence can be deduced from the accumulation of oxides, oxide layers and similar groups of oxide inclusions at the boundary surface between explosively welded low-carbon steel and molybdenum.

The temperature within the molten islands exceeds the melting point of tantalum (approximately 3000 °C) and the melting point of iron (approximately 1500 °C), as well as that of steel oxides (up to 1600 °C). Both the activity of the components in the melt and the equilibrium constants are temperature dependent. During explosive welding, the temperature drops very rapidly from around 3000 °C down to the solidification temperature of the alloy, between 1700 - 1800 °C. Solidification occurs in approximately 0.1 seconds. The duration of this process appears to be long enough to allow intensive deoxidation to occur, but not sufficient for the chemical homogenisation of the alloy, which results from the mixing of two very different metals.

## CONCLUSIONS

The alloy formed in the shape of islands on the metal interface contributes to the bond strength of explosively welded plates of low-carbon steel and tantalum. The alloy is formed by the mixing of locally melted steel and tantalum.

The chemical composition of the alloy was within a concentration range, however, it did not correspond to any typical phase or microstructural constituent of the Fe-Ta equilibrium phase diagram.

The solidification of the alloy islands occurred through the formation of dendrites (mainly smaller than 5 µm) and cells.

Within the melt, existent for a very short time ( $10^{-2}$  to  $10^{-1}$  seconds), intensive deoxidation took place, resulting in the formation of non-metallic inclusions of a complex chemical composition.

The basic metallic constituent of the inclusions was tantalum with a relatively high concentration of iron and aluminium and a low concentration of manganese. Aluminium, which originated from either one or both components, was also found in the dendrites of remelted alloys. In the region of cell solidification, its concentration was below the sensitivity of the measuring instrument (approximately 0.1 mass contents in %).

The inclusion diameters were about 0.1 to 2 µm. The tantalum concentrations increased with inclusion size. Differences in chemical composition of the inclusions could be the result of either an inhomogeneous melt or changes connected with their growth.

Many non-metallic inclusions nucleated on the melt – tantalum interface while none were found on the melt-steel interface. Non-metallic inclusions also behaved as nuclei for melt crystallisation in the dendrite region.

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