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The Distinctive Features of Diffusion Interaction of Copper and Molybdenum under Pressure Welding Through the Layers Modified by Ion-Beam Processing

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The process of diffusion welding of copper with molybdenum in the vacuum through the layers modified by ion-beam processing is investigated. The distinctive features of diffusion interaction of copper and molybdenum are determined using the radioactive tracer technique. The possibility of substantial influence of changing the critical parameters of the mode of ion-beam processing of surface layer of the welded material on the sizes of interaction zone and mechanical properties of the joint weld is shown. Expediency of previous modification of molybdenum surface with copper in the process of diffusion welding of copper with the molybdenum in the vacuum is established.

Досліджено процес дифузійного зварювання у вакуумі міді з молібденом через модифіковані йонним обробленням прошарки. За допомогою методи радіоактивних ізотопів визначено особливості дифузійної взаємодії міді з молібденом. Показано, що, змінюючи основні параметри режиму йонного оброблення поверхневого шару зварного матеріялу, можна істотно впливати на розміри зони взаємодії та механічні властивості зварного з'єднання. Встановлено доцільність при дифузійному зварюванні у вакуумі міді з молібденом попередньої модифікації поверхні молібдену міддю.

Исследован процесс диффузионной сварки в вакууме меди с молибденом через модифицированные ионной обработкой прослойки. С помощью метода радиоактивных изотопов определены особенности диффузионного взаимодействия меди с молибденом. Показано, что, изменяя основные параметры режима ионной обработки поверхностного слоя сварного материала, можно существенно влиять на размеры зоны взаимодействия и механические свойства сварного соединения. Установлена целесообразность при диффузионной сварке в вакууме меди с молибденом предварительной модификации поверхности молибдена медью.

Key words: copper, molybdenum, diffusive welding, ion-beam processing, radioactive tracer technique.

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1. INTRODUCTION

Thin interlayers (superdispersed powders of 1 micron thickness which are applied using different methods) of metals (copper, nickel, cobalt, *etc.*, as well as their mixtures) with a superdispersed structure are applied as interfacial layers in the process of diffusive welding of heterogeneous materials [1–3]. Moreover, the advantage of such interlayers is that their presence in a joint leads to substantial intensification of the process of formation of junctions and provides the mechanical and thermal activation of surfaces in the process of welding. Their influence also results in high diffusive activity in the contact zone [4–6], which is a consequence of extremely small sizes of particles and highly developed free surface. It is related to the fact that the diffusion coefficient is a structure-sensitive quantity and its effective value can substantially exceed the equilibrium value involving macroscopic defects and developed by the system of interface.

This technological method is practically the only variant in the case when it is necessary to connect metals, which are difficulty welded qualitatively in the solid state. It concerns the couple of molybdenum–copper, the combination of which is used to produce various wares in electrical engineering industry. The distinctive feature of such diffusive couple is absence of solubility of Mo in Cu [7, 8]. This circumstance prevents qualitative connection of these metals using diffusion-welding method.

2. EXPERIMENTAL METHODOLOGY

In this work, the results of investigations of the processes of atoms redistribution and diffusion rates in the process of diffusion welding of molybdenum with a copper through the layers modified by ion-beam processing are presented.

The layer of copper of 1 mkm thick is being deposited on the surface

TABLE 1. The modes of preparation of samples and their diffusion welding.

No. of sample	No. of mode	T_{welding}, °C	τ_{welding}, min	$\tau^{\text{Ar}}_{\text{treatment}}$, min	$U_{\text{acceleration}}$, V	P, kilogram-force/mm^2	Thickness of Cu layer, mkm
1	I	950	20	0	0	1.5	1
2	II	950	20	10	1200	1.5	1
3	III	950	20	20	1200	1.5	1
4	IV	950	20	20	2000	1.5	1

of molybdenum using ion-plasma method. Then, all samples of Mo with the layer of copper, except the basic ones, are being subjected to the treatment by ions of argon to create the atomic interaction zone of Mo with Cu for the purpose of increasing of adhesion. After that, the samples are being subjected to diffusion welding. The modes of preparation of samples and their diffusion welding are presented in Table 1.

3. RESULTS AND DISCUSSIONS

The number 1 (I) in this case corresponds to the virgin sample. The other modes differ from the basic one and one from another by the duration of argon treatment ($\tau^{\text{Ar}}_{\text{treatment}}$) and accelerating voltage ($U_{\text{acceleration}}$) of such treatment.

The diffusion processes in the interface in the process of welding of the samples are being studied using the radioactive tracer technique. The radioactive isotope of nickel ^{63}Ni, which is being inflicted onto the Mo by the electrolytic method, is used in this work. The thickness of ^{63}Ni layer is about $\cong 0.3$ micron and the activity—$5\cdot10^3$ imp./min. After welding process, the samples are cut and disposed on the X-ray film. Holding on the film comprises about 100 hours. The film is being scanned photometrically and the concentration distribution of radioactive isotope of nickel into the both Mo and Cu is obtained. The estimation of values of the diffusion coefficient is conducted using the methodology described in Ref. [9].

The concentration distribution of the radioactive isotope of nickel into the both Mo and Cu after the diffusion welding according to the four modes is presented in the Fig. 1.

The analysis of the presented curves demonstrates that in the process of welding the ^{63}Ni penetrates into considerable depths (X zones of tens and hundreds of microns) into the both Mo and Cu.

The clearly defined curves asymmetry appearing in noticeably greater penetration depth of ^{63}Ni into the Cu in comparison with Mo should be noticed. This result can be related to the circumstance that,

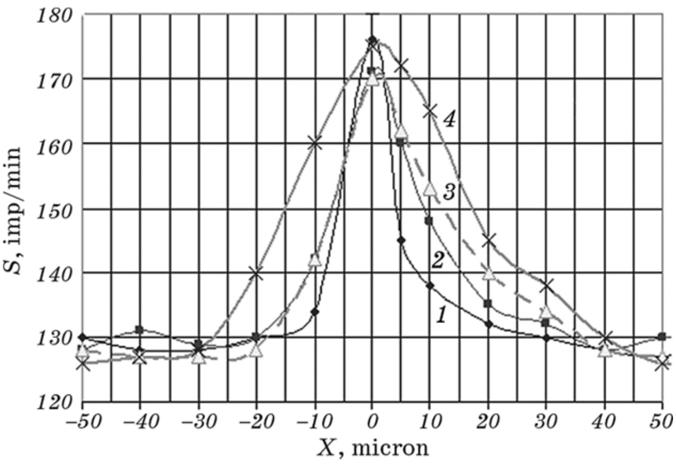

Fig. 1. Concentration distribution of ^{63}Ni in Mo and Cu after welding on the base mode I and on the II, III, IV modes (see Table 1).

in accordance with the phase diagram of Cu–Ni [10, 11], these metals within the all range of concentrations have unlimited solubility.

At the same time, the solubility of Ni in Mo and Mo in Ni is substantially limited in accordance with the phase diagram [12–14]. In addition, it is necessary to take into account the considerable difference in the melting temperatures of Mo and Cu, 2610°C and 1083°C respectively [15]. Thus, the temperature in the process of diffusion welding (950°C) is considerably higher for Cu as compared with Mo. Thereby, the reduced temperature ($T_{\text{annealing}}/T_{\text{melting}}$) for Cu in this case is 0.88 and that for Mo is 0.37.

The experimental data obtained as a result of concentration–response curves processing are presented in Table 2 and Figs. 2–5.

As seen from Table 2, the depth of ^{63}Ni distribution in Cu is in all cases bigger in comparison with Mo. At the same time, the ratio of these depths is different (see Fig. 2).

TABLE 2. The results of concentration–response curves processing.

No. of sample	X zones, mkm		$\tau^{\text{Ar}}_{\text{treatment}}$, min		τ_{welding}, min		$U_{\text{acceleration}}$, V		$X_{\text{Cu}}/X_{\text{Mo}}$
	Cu	Mo	Cu	Mo	Cu	Mo	Cu	Mo	
1	11	6	–	0	20	20	0	0	1.8
2	26	19	–	10	20	20	1200	1200	1.4
3	45	33	–	20	20	20	1200	1200	1.4
4	60	44	–	20	20	20	2000	2000	1.4

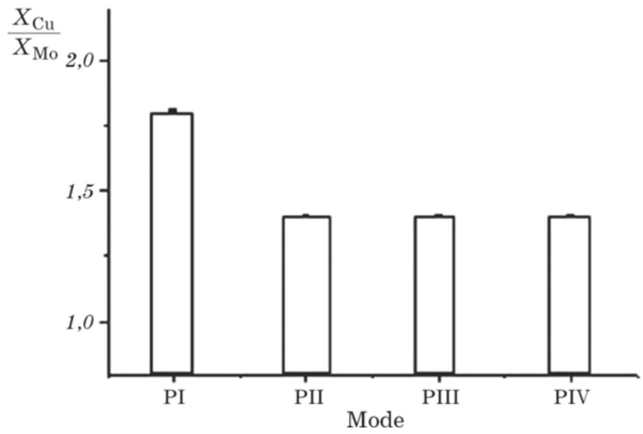

Fig. 2. The change of ratio penetration depth (*X*) of ^{63}Ni into the both Cu and Mo depending on the mode of treatment.

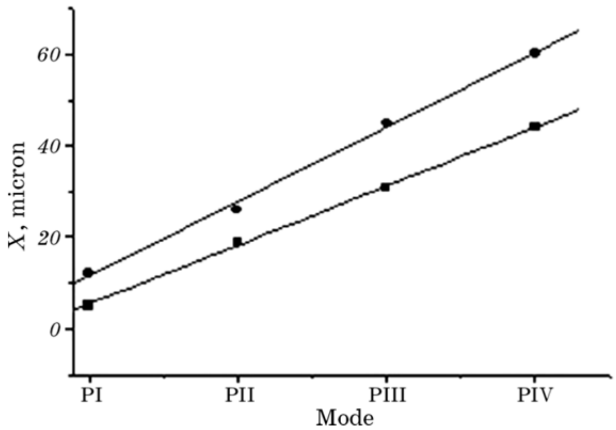

Fig. 3. The change of penetration depth of ^{63}Ni into the both Cu (●) and Mo (■) depending on the mode of treatment.

Such character of the presented dependence is related to the fact that the maximal ratio of X_{Cu}/X_{Mo} concerns to the mode I (basic). The distinctive feature of this mode is the absence of treatment of Mo by means of applying Cu layer on it by Ar ions. It is obvious that the treatment of the indicated surface by using Ar results in the mixing of atoms of the layers that are on Mo. They are copper, ^{63}Ni, and Mo. As a result, the part of ^{63}Ni atoms penetrates into the Mo and forms a pseudo diffusion layer after this treatment. In the process of the subsequent diffusion welding, ^{63}Ni atoms diffuse into the Mo from the certain depth and a total diffusion layer becomes thicker. When the mode I is applied, these processes do not occur.

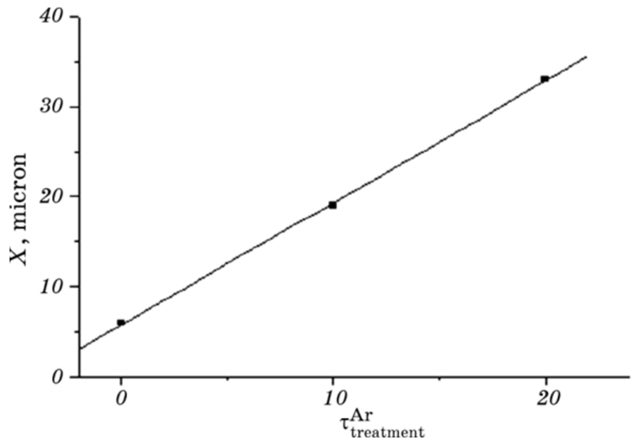

Fig. 4. The dependence of penetration depth of ^{63}Ni into the Mo on the time of treatment by Ar ions.

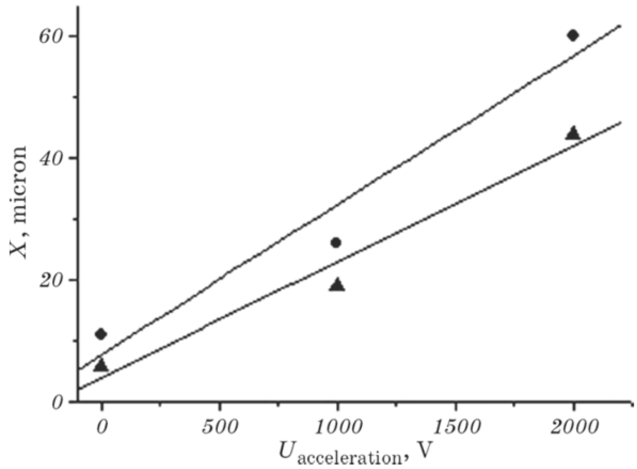

Fig. 5. The dependence of penetration depth of ^{63}Ni into the both Cu (●) and Mo (▲) depending on the value of accelerating potential during treatment by ions of argon.

The dependence of penetration depth of ^{63}Ni into the both Cu and Mo depending on the mode of treatment is presented in Fig. 3.

The dependence of penetration depth of ^{63}Ni into the Mo on duration of argon ions treatment is presented in Fig. 4.

As Figure 4 shows, the specified dependence has the linear character. The increase of treatment time of Mo with applied copper layer by Ar ions results in the substantial increase of penetration depth of ^{63}Ni into it, namely, from 6 to 33 mkm. Based on this fact, one can come to the conclusion that, changing the time of treatment of Mo by Ar ions, it is possible to influence on the sizes of interaction zone of copper with molybdenum substantially and, at the same time, to control the adhesion value of Cu to Mo. The similar conclusion can be made using the data presented in Fig. 5.

As Figure 5 demonstrates, the penetration depths of ^{63}Ni into copper and molybdenum depend substantially on the values of accelerating potential during molybdenum treatment by ions of argon. These dependences have the linear character. Besides, it should be noted that the values of penetration zone (X) of ^{63}Ni in copper are considerably higher as compared with molybdenum.

The important parameter determining mechanical parameters of the welds is the diffusion coefficient. This parameter characterizes features and regularities of interaction of materials in many technological processes. Namely, it is very important in chemicothermal treatment, welding and other processes. Owing to such importance, the investigation of diffusive features of welding copper with molybdenum with the use of radioactive isotope of ^{63}Ni is presented in this work. The results of this research are presented in Table 3.

Analysing the data presented in Table 3, one can see that the mobility of ^{63}Ni in copper is almost 3.5 times as high as compared with molybdenum. As mentioned above, it can be related to the melting temperature of molybdenum and copper, which almost three times differs and, at the same time, the solubility of nickel in copper is considerably higher.

The dependence of logarithms of diffusion coefficients depending on the modes of treatment and welding is presented in Fig. 6. Therefore, the mobility of nickel in copper is in all cases higher than that in molybdenum. Thus, it is clear that the increase of value of diffusion coefficients diminishes in the process of change of the mode of treatment.

The mechanical tests of the obtained welds show that, depending on the mode of welding, the durability of the weld also changes (see Fig. 7).

TABLE 3. The distinctive features of diffusive processes in the process of welding of copper with molybdenum.

No. of sample	$D_{eff.}$, sm^2/s		$D_{eff.}^{Cu}/D_{eff.}^{Mo}$	lg($D_{eff.}$)	
	Cu	Mo		Cu	Mo
1	$5.1 \cdot 10^{-10}$	$1.5 \cdot 10^{-10}$	3.4	–9.3	–9.8
2	$3.22 \cdot 10^{-9}$	$1.0 \cdot 10^{-9}$	3.22	–8.6	–9.0
3	$8.44 \cdot 10^{-9}$	$2.26 \cdot 10^{-9}$	3.7	–8.1	–8.7
4	$1.5 \cdot 10^{-8}$	$4.3 \cdot 10^{-9}$	3.5	–7.8	–8.4

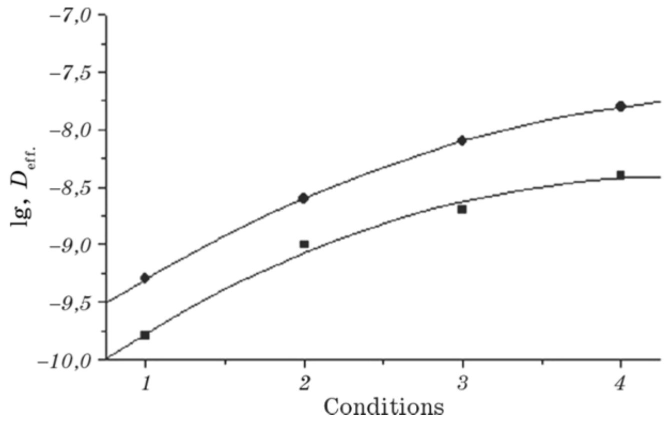

Fig. 6. The dependence of logarithms of diffusion coefficients depending on the modes of treatment: Cu (●), Mo (■).

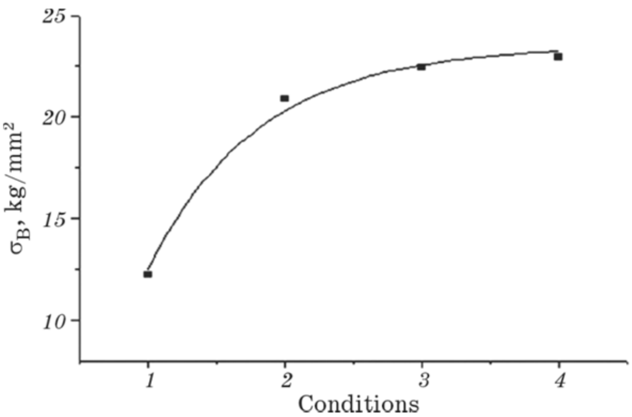

Fig. 7. The dependence of durability of the weld of copper with molybdenum on the mode of welding.

As it is presented in Fig. 7, the mechanical characteristics of welds on pulling after using the mode 4 achieve the durability of the basic metal—copper. Such result proves the possibility of the use of the considered method of surface activating in the process of welding of heterogeneous metals, which physicochemical and mechanical properties differ substantially.

4. CONCLUSIONS

1. It is determined that the increase of time of treatment and accelerating potential under treatment of Mo with copper layer applied on it by

ions of argon allows to extend substantially the sizes of interaction zone of copper and molybdenum. It is shown that ion-beam treatment of copper covering on molybdenum allows extending the zone of diffusive interaction in the process of diffusive welding of copper with molybdenum by 4–5 times in comparison with direct welding.
2. It is shown that ion-beam treatment of copper covering on molybdenum allows doubling the durability of the weld of copper with molybdenum in comparison with welding without treatment.

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