Peculiarities of Transition-Zone Formation at Al−Cu Explosion Welding

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The formation of transition zones during explosive welding of aluminium with copper in vacuum and in air is studied. The distribution of element concentrations is used to determine the coefficient of mutual mass transfer in the undetachable Al−Cu joint. It turns out that the mass transfer of copper into aluminium in the conditions of explosion welding occurs at greater depths compared to the depth of penetration of aluminium into copper. This is explained by the fact that the nature of the penetrating atoms does not have such a strong influence as in conditions of diffusion annealing, and the determining factor is the lattice period, which in aluminium is larger than in

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copper. Concentration curves are plotted for the Al−Cu interaction zone during explosion welding, in which there are no intermetallic phases corresponding to the equilibrium diagram of state. The coefficient of mutual mass transfer turns out to be several orders of magnitude higher than the heterodiffusion coefficient of the same elements under diffusion-annealing conditions. The explosion welding is performed using a coaxial scheme by throwing a copper tube on an aluminium rod.

Key words: aluminium, copper, explosion welding, mass transfer, diffusion, interaction zone.

Вивчено формування перехідної зони під час зварювання вибухом алюмінію з міддю у вакуумі та на повітрі. За розподілом концентрації елементів визначено коефіцієнт взаємного масоперенесення у нерознімному з'єднанні Al−Cu. Виявилося, що масоперенесення Купруму в алюміній за умов зварювання вибухом відбувається на більші глибини порівняно з глибиною проникнення Алюмінію у мідь. Це пояснюється тим, що природа проникальних атомів не має такого сильного впливу, як в умовах дифузійного відпалу, а визначальним є період ґратниці, який у алюмінію є більшим, аніж у міді. Концентраційні криві побудовано для зони взаємодії Al−Cu під час зварювання вибухом, в якій відсутні інтерметалеві фази, що відповідають рівноважній діяграмі стану. Коефіцієнт взаємного масоперенесення виявився на кілька порядків вищим, аніж коефіцієнт гетеродифузії тих же елементів в умовах дифузійного відпалу. Зварювання вибухом виконували за коаксіяльною схемою шляхом метання мідної трубки на алюмінійовий стрижень.

Ключові слова: алюміній, мідь, зварювання вибухом, масоперенесення, дифузія, зона взаємодії.

1. INTRODUCTION

Explosive welding [1−3] attracts the attention of researchers and technologists due to the presence of a number of advantages over other types of welding. The most important of them is the possibility to obtain reliable joints of a wide range of metals, which may differ in hardness, melting point, the possibility of the formation of solid solutions and intermetallides, linear expansion and mass-transfer coefficients. The use of the term 'diffusion' in explosion welding is not quite correct. The term 'mass transfer' is more accurate, since the process takes place under conditions far from equilibrium. Under such conditions, the phenomenon of accelerated mass transfer in the solid phase [4−6], which is realized in shock waves [7−10] and in other types of pulse processing, such as shock welding [11, 12], pulse laser processing [13, 14], martensitic transformations [15−17], elastic compression waves [18, 19], ultrasonic shock processing [20] and other ones [21], is manifested

during high-speed deformation.

An important circumstance is also the fact that explosion welding can combine obtaining a joint with simultaneous strengthening of the base metal and the joint zone.

Consequently, for the selection of welding, modes should be based on the concepts of the interaction of metals in the solid phase, because the presence of a liquid phase can lead to the formation of brittle intermetallides. This approach allows us to establish the role of the main factors affecting the course of solid-phase reactions, namely, the defects in the crystal structure and the mutual mass transfer of the components to be joined.

The process of joint formation during explosion welding of copper with aluminium, including coaxial joints, is greatly influenced by the heated shock-compressed gas filling the gap between the surfaces due to the low melting point of aluminium and the formation of a low-melting ($T_{melt} = 548°C$) $Al_2Cu + \kappa$ eutectic [22]. To eliminate the influence of shock-compressed air during explosion welding, vacuuming of the welding gap is used.

Therefore, the purpose of this work is to study the peculiarities of mutual penetration of copper and aluminium and the influence of mass transfer on the formation of an unbreakable Al–Cu coaxial joint during explosion welding, including vacuumization of the welding gap.

Let us consider the equilibrium diagram of the states of this pair [23]. As follows from the above diagram, the solubility with the formation of a solid solution of Cu in aluminium and aluminium in Cu is very different. At room temperature, Cu dissolves into Al in an amount of 0.2% wt. (0.1 at.%) and the maximum solubility is of 5.65% wt. (2.48 at.%) at eutectic temperature. At the same time, the face-centred cubic lattice (f.c.c.) parameter of aluminium decreases from 0.4049 nm to 0.4038–0.4041 nm.

At room temperature, Al is dissolved in copper in an amount of 7.1% wt. (3.2 at.%). At the same time, the f.c.c. lattice parameter of Cu increases from 0.36151 nm to 0.36633 nm.

A comparison of the above data indicates that the solubility of Al in Cu with the preservation of a solid solution is about 35 times higher as compared to the solubility of Cu in Al. Analysis of diffusion data showed that this affects the diffusion rate of aluminium and copper atoms [24, 25].

Comparison of diffusion coefficients of copper in aluminium and aluminium in copper in works [26, 27] showed that, when the reduced temperature is equal ($T_{anneal}/T_{melt.}$), where T_{anneal} is the diffusion annealing temperature, $T_{melt.}$ is the melting temperature of the metal, in which diffusion takes place, the diffusion coefficients are equal.

Diffusion (mass transfer) processes in metals and alloys under conditions of explosion processing are currently insufficiently studied.

This information is necessary for the development of explosion welding modes for new combinations of materials. In particular, we can cite the works [5, 6, 9, 11], in which the authors considered these issues. In Ref. [30], they studied the processes of diffusion interaction in the boundary layer of the Cu–S235 (analogue of steel 3) bimetal. It was found that the mutual penetration of copper and iron is directly dependent on the pressure in the contact zone. Thus, at pressure $P = 4$ GPa, the effect of copper penetration into S235 and back within the sensitivity of the method is not detected, and at $P = 21$ GPa, the depth of the interaction zone is of about 7 microns.

The estimated value of the coefficient of mutual diffusion (mass transfer) in the system Cu–S235 is of the order 10^{-2}–10^{-3} cm^2/s. This value is much higher than the values of the diffusion coefficients of copper in iron and iron in copper under steady-state conditions. Thus, at normal pressure, $D_{Fe \to Cu} = 1.32 \cdot 10^{-13}$ cm^2/s and $D_{Cu \to Fe} = 2.7 \cdot 10^{-13}$ cm^2/s [29]. Possible mechanisms of such excess at explosion welding of Cu–S235 compared to diffusion under steady state conditions are considered in Refs. [5, 31].

However, under the conditions of explosion welding, as noted above, this issue is practically not studied. Among other things, it concerns the Al–Cu bimetallic pair. This metal pair is of practical interest for the fabrication of bimetallic conductors (copper sheath on an aluminium base with low density).

To avoid confusion, the following terminology will be adopted in this paper. The processes of atom transfer due to concentration gradients under conditions close to equilibrium will be called as 'diffusion processes'. The term 'mass transfer' will be used to describe the processes occurring in nonstationary conditions and accompanied by plastic deformation. Calculations of the corresponding coefficients for these processes occurring at the atomic level will be performed by similar methods, taking into account the process time.

2. EXPERIMENTAL AND THEORETICAL DETAILS

2.1. Description of Materials

In this work, aluminium (1230) and copper (11000) were used as weldable materials. The choice of these materials is primarily due to their use as basic or cladding elements of composite materials for electrical applications.

Explosion welding was carried out according to the coaxial scheme by throwing a copper tube of 28 mm diameter with a wall thickness of 1 mm onto an aluminium rod of 24 mm diameter. The length of the welded blanks was 1000 mm. Explosive welding of the tubes was performed at an impact velocity of 320 m/s, with and without vacuumization of

TABLE 1. Chemical composition of aluminium 1230.

Al (not less), %	Impurities (no more), %						
	Ti	Mg	Cu	Mn	Zn	Si	Fe
99.3	0.15	0.05	0.05	0.025	0.1	0.3	0.3

TABLE 2. Chemical composition of copper 11000.

Cu (not less), %	Impurities (no more), %								
	Bi	Sb	As	Fe	Ni	Pb	Sn	S	O	Zn
99.90	0.001	0.002	0.002	0.005	0.002	0.005	0.002	0.004	0.05	0.004

the welding gap.

The chemical composition of aluminium and M1 copper is presented in Tables 1, 2.

2.2. Preparation of Specimens for the Study

After fabrication of the bimetallic rod by explosion welding, cylindrical specimens (up to 17 mm in length) were cut from it for microstructure study. These specimens were then cut in half in the longitudinal direction so that the cutting line passed through the centre of the specimen, taking into account the thickness of the cutting tool.

After cutting and polishing, the obtained specimens were washed in ethyl alcohol using an ultrasonic cleaning machine. The study was carried out on non-polished specimens.

2.3. Obtaining Concentration Curves

The main studies of the structure and chemical composition of the compound zone were carried out on the Auger microprobe JAMP-9500F of the 'JEOL' company (Japan).

The Auger-microprobe JAMP-9500F combines the properties of a high-precision Auger-analyser with energy resolution ($\Delta E/E = 0.05\%$), and a good scanning electron microscope with secondary electron resolution, which is not worse than 3 nm on a 24-millimeter working section.

Concentration curves of the transition zone of a copper–aluminium joint obtained by explosion welding in air and in vacuum were plotted as a result of the research.

The penetration depths were determined by the corresponding concentration curves, and the interface between the two materials was

taken as the origin of the co-ordinates.

2.4. Method of Layer-by-Layer Analysis

This method is used, when using radioactive isotopes [5], as well as when using other methods, by means of which the concentration of the diffusing element at a certain depth of the diffusion zone is determined. Thus, according to the measured values of the activity of labelled atoms or their concentration, the concentration curve of the element distribution in the diffusion zone is constructed, which is the basic curve for determining the values of mass-transfer coefficients.

The concentration distribution curves of diffusing elements in the sample serve as an experimental basis in this case. The essence of this method is described in Refs. [4, 31].

The calculation formula for determining the value of the mass-transfer (diffusion) coefficient has the following form:

$$\text{(1)}$$

or, on the other hand, when passing to the decimal logarithm),

$$D_{\text{M}} = -\frac{0.1086}{\tau \text{tg}\alpha}; \tag{2}$$

τ is processing time, $\text{tg}\alpha$ is angle of inclination of the tangent to the concentration curve.

2.5. Boltzmann−Matano Analysis

This method is used to calculate the coefficient of mutual diffusion, which depends on concentration [32]. The concentration curve in the co-ordinates $c = f(x)$ corresponds to a certain mass-transfer (diffusion) time. Hence, $\tau = \text{const}$. Then,

$$D_{\text{mut.}} = -\frac{1}{2\tau}\frac{dx}{dc}\int_0^c xdc. \tag{3}$$

The influence of concentration on the diffusion coefficient must be taken into account, when the metals to be welded have a whole set of intermetallic compounds. Such metals include Al−Cu pair, Fe−Ti, Al-stainless steel, and many others, which also find application in various fields of science and technology.

It should also be noted that, in explosion welding, the problem of experimental and theoretical assessment of the temperature in the joint zone is difficult.

This factor should be taken into account, when developing the welding mode, since it has a significant impact on the formation of phases (including the formation of intermetallides) and the structural state of the welded joint material, as well as on the diffusion characteristics of the components of this joint. The temperature factor is especially important in the case of the explosion welding of metals with very different melting points.

3. RESULTS AND DISCUSSION

3.1. Explosion Welding of Copper with Aluminium with Vacuuming of the Welding Gap

The distribution curves of copper in aluminium and aluminium in copper after explosion welding are shown in Fig. 1.

In the above graphs, we can see that the depth of aluminium penetration into copper (0.7 µm) is smaller than the size of the copper–aluminium interaction zone (approximately 1.8 µm).

It should be noted that, according to the equilibrium diagram, the solubility of the elements in the Cu–Al system is different. According to Ref. [23], at room temperature, copper dissolves in aluminium in the amount of 0.1 at.%. Under these conditions, the amount of aluminium in copper reaches 18 at.%, *i.e.*, much more.

In addition, the shape of the concentration curves (Fig. 1) and the microstructure photograph (Fig. 2) indicate that there are no intermetallics in the area of interaction between copper and aluminium, which are formed in this system in a wide range of concentrations under equi-
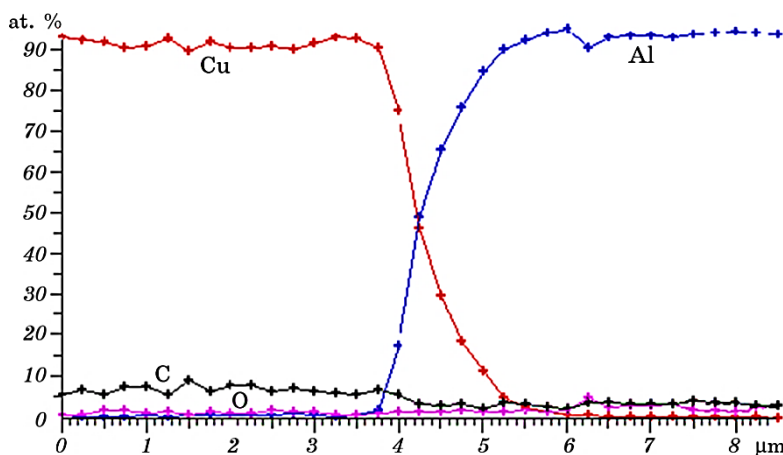

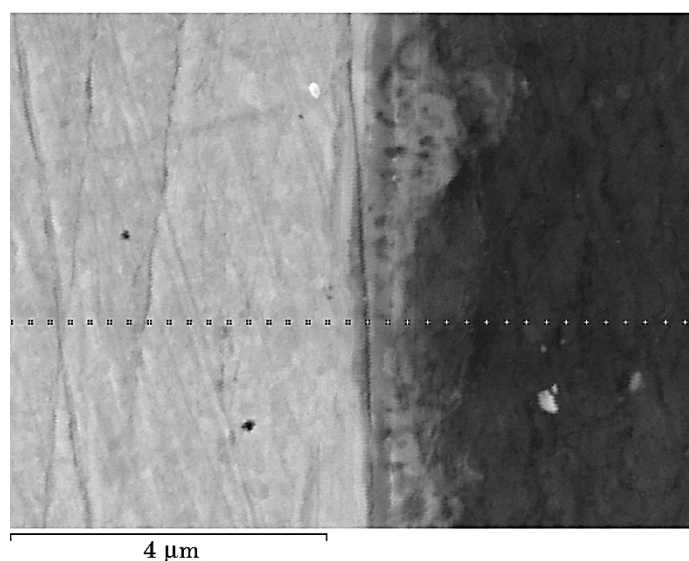**Fig. 1.** Concentration curves of copper in aluminium and aluminium in copper after vacuum explosion welding.

4 μm

Fig. 2. Microstructure of the contact zone after vacuum explosion welding of copper with aluminium (×150, back (secondary) electrons).

librium conditions. In this case, a solid solution of aluminium in copper is formed and *vice versa*. This can be explained by the short time of the pulse action and rapid cooling due to effective heat removal from the zone of elevated temperatures. In other words, the duration of the elevated temperature is shorter than the latent period of intermetallic formation. It should be noted that the copper distribution curves in aluminium were determined both in areas with and without intermetallics. In our publication, we present the curves in the areas without intermetallics, while the area next to it, which is not visible in the photo, has intermetallics.

In Figure 2, the dots show the places where the concentration of elements was determined.

From the graphs shown in Fig. 1, it is possible to determine the coefficients of mutual diffusion in the Cu–Al system by the Matano–Boltzmann method [32]. Mutual diffusion—mutual penetration of atoms or molecules at contact of two environs (solid, liquid, gaseous). Mutual diffusion coefficient D_{12} is related to partial self-diffusion coefficients D_1 and D_2 (n_1, n_2 are concentrations of corresponding elements):

$$D_{12} = \frac{D_2 n_1 + D_1 n_2}{n_1 + n_2}.$$

(4)

In this case, the value of the Cu–Al mutual mass-transfer coefficient

was equal to $D_{\text{mut.}} = 1.0 \cdot 10^{-2}$ cm^2/s. At the same time, during the stationary annealing of this pair (523 K) [29], $D_{\text{mut.}}$ is of $1.2 \cdot 10^{-12}$ cm^2/s. Their ratio is $8.3 \cdot 10^9$. Thus, an increase in the coefficient of mutual mass transfer by almost 10 orders of magnitude is observed during explosion welding of copper with aluminium that indicates the realisation of the phenomenon of anomalous mass transfer.

To determine the values of the partial mass-transfer coefficients of copper into aluminium and aluminium into copper, the corresponding branches shown in Fig. 1 were used. The mass-transfer coefficient was calculated according to the Gruzin method using the data from Fig. 1 [4].

As can be seen in Fig. 3, the graphs represent a straight line, which suggests that this transfer process is realised in the volume, which resembles the classical diffusion transport. As can be seen in Fig. 3, the atoms penetration of aluminium into copper in comparison with copper into aluminium occurs at greater depths, despite the fact that the solubility, according to the diagram of states, is greater for aluminium in copper [23]. The calculated mass-transfer coefficients are $7.4 \cdot 10^{-4}$ cm^2/s and $4.3 \cdot 10^{-3}$ cm^2/s, which is practically equal to or even higher than the coefficients of atomic diffusion in the liquid metal [33]. Under equilibrium conditions, the values of the heterodiffusion coefficients at the deposition temperature of 973 K are $1.6 \cdot 10^{-11}$ cm^2/s and $6.7 \cdot 10^{-9}$ cm^2/s at 903 K [29]. The excess of the mass-transfer coefficients of aluminium atoms into copper in comparison with the heterodiffusion coefficient is approximately 8 orders of magnitude, and of copper atoms into aluminium is 6 orders.
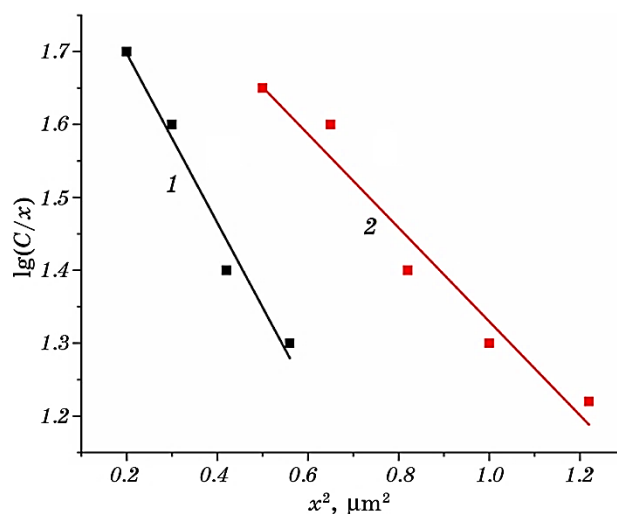

Fig. 3. $\lg(C/x) = f(x^2)$ dependence: transport Al to Cu (*1*) and Cu to Al (*2*) at explosion welding in vacuum.

The obtained result can be explained by the fact that the nature of the penetrating atoms does not have such a strong influence at the values of the mass-transfer and diffusion coefficients, as in conditions of diffusion annealing, and the determining factor is the lattice period, which is larger in aluminium. Under conditions of mass transfer, a major role is played by mobile dislocations, which can entrap the penetrating copper atoms. This leads to the fact that the deformation and mass-transfer processes are easier in less strong aluminium. As can be seen, the increased solubility of Al in Cu according to the equilibrium state diagram [23] has no effect on the mass-transfer coefficients during blasting.

3.2. Explosive Welding of Copper with Aluminium in Air

The welding parameters in air are identical to those used for welding in vacuum except for the temperature in the contact zone, which was higher in air due to heating by shock-compressed gas.

Concentration-distribution curves of Al in Cu and Cu in Al obtained in the study of the welded joint of the Al−Cu-metal system are presented in Fig. 4.

From the comparison of the presented curves, it follows that the sizes of the zones of penetration of the mentioned elements into each other are significantly different. Thus, the size of the Al → Cu zone is of about 0.7 μm, and for the Cu → Al zone, it is of 2.6 μm.

From comparison of the shape of concentration curves (Fig. 4) and microstructure (Fig. 5), it follows that there are no intermetallides in the zones of copper−aluminium interaction (as well as in the case of
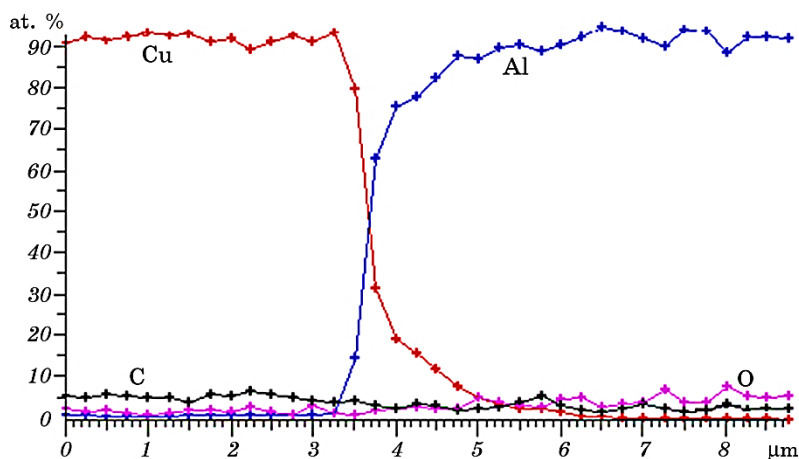

Fig. 4. Concentration distribution curves of copper in aluminium and aluminium in copper after explosion welding in air.
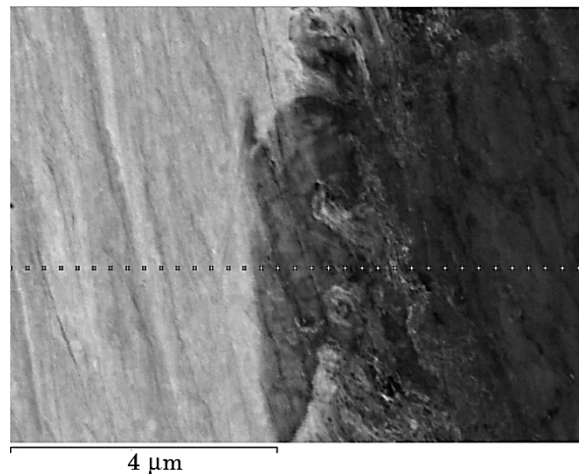
Fig. 5. Microstructure of Cu–Al contact zone after explosion welding in air (×150, back (secondary) electrons).

welding in vacuum).

Figure 5 shows the contact zone of the welded element Cu–Al.

The coefficient of mutual mass transfer in the Cu–Al system is $D_{mut.} = 1.0 \cdot 10^{-2}\ cm^2/s$ during air blast welding. Consequently, in the case of explosion welding in air of copper and aluminium, there is observed (as well as in the case of welding in vacuum) an increase in the coefficient of mutual mass transfer by more than 9 orders of magnitude.

For the determination of the values of the mass-transfer coefficients of copper to aluminium and aluminium to copper at welding by explosion in air, the corresponding concentration curves (to the right and left from the zero axis x [µm], Fig. 4) were used. Figure 6 presents concentration curves of distributions of copper in aluminium and aluminium in copper, which are rearranged in $lg(C/x) = f(x^2)$ co-ordinates.

For the explosion welding of Cu–Al in air, the volumetric character of the mass-transfer process is observed, as well as for the welding of this pair in vacuum.

Calculation of the mass-transfer coefficients in this case gives the values $(D_m)^{Cu \to Al} = 7.2 \cdot 10^{-3}\ cm^2/s$ and $(D_m)^{Al \to Cu} = 1.03 \cdot 10^{-3}\ cm^2/s$. These values of D_m slightly exceed the values of diffusion coefficients in Cu–Al vapour in the liquid state [33].

Comparing the mass-transfer coefficients at explosion welding in air with the case of stationary annealing at temperatures of about 900 K (see data in the section on explosion welding in vacuum), we obtain that the increase in the mass-transfer coefficient of copper to aluminium is about 6 orders of magnitude, and for aluminium to copper, it is about 8 orders of magnitude.
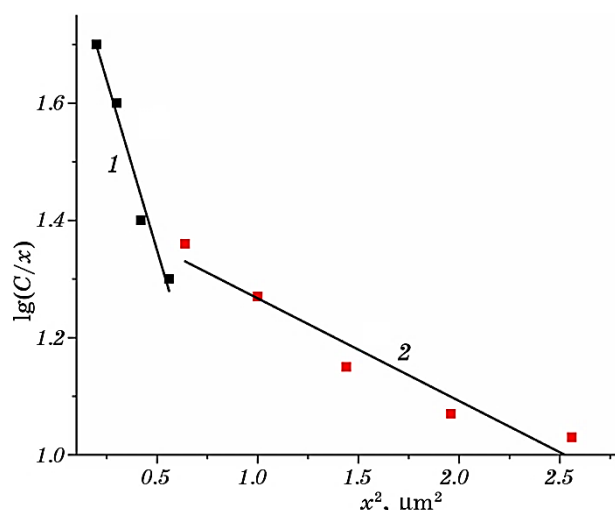
Fig. 6. $\lg(C/x) = f(x^2)$ dependence: transport Al to Cu (*1*) and Cu to Al (*2*) at explosion welding on air.

In the case of explosion welding in air, the depth of mass transfer of copper to aluminium is greater, which can be explained by the higher temperature compared to vacuum welding. As can be seen from Table 3, the mutual mass-transfer coefficient does not depend on the atmosphere used at welding. Since the heating temperature during explosion welding is higher on air, and $D_{mut.}$ are equalled for both atmospheres, it can be concluded that heating due to the presence of air can be neglected, and the main contribution to mass transfer is made by the movement of atoms under the action of the driving force.

As mentioned above, the experimental results on mass transfer in copper–aluminium explosion welding indicate its high speed and volumetric nature. Usually, atomic migration can be associated with various diffusion mechanisms: vacancy, dislocation, internodal, *etc*. Consideration of these mechanisms has shown that migration through vacancies or along dislocation tubes cannot provide the available penetra-

TABLE 3. Values of the diffusion and mass-transfer coefficients.

$D_{mut.}$, cm²/s (Cu–Al)		D_{getero}, cm²/s		D_M, cm²/s			
Explosion welding in vacuum and air	Diffusion annealing (523 K)	Cu → Al (903 K)	Al → Cu (973 K)	Explosion welding in vacuum (Cu → Al)	Explosion welding in vacuum (Al → Cu)	Explosion welding in air (Cu → Al)	Explosion welding in air (Al → Cu)
$1.0 \cdot 10^{-2}$	$1.2 \cdot 10^{-12}$	$6.7 \cdot 10^{-9}$	$1.6 \cdot 10^{-11}$	$4.3 \cdot 10^{-3}$	$7.4 \cdot 10^{-4}$	$7.2 \cdot 10^{-3}$	$1.03 \cdot 10^{-3}$

tion rates and depths. As for the internodal mechanism, despite the erroneous statement of the author of Ref. [34] about the high energies required for the creation and movement of its own (or substituted) internodal atom, this assumption of the internodal mechanism has the right to exist since the creation of this defect occurs at the expense of the energy of impulse deformation, and for its movement, the required energies are by 1−2 orders of magnitude lower than for the movement of the vacancy.

4. CONCLUSIONS

The main conclusions are as follow.

1. The coefficient of mutual mass transfer in an unbreakable Cu−Al joint obtained by explosion welding has been determined. The coefficients of mass transfer at vibe welding exceed their values at diffusion welding by $\cong 7-10$ orders of magnitude and by $\cong 11-12$ at isothermal falls.

2. Despite the higher solubility of aluminium in copper, according to the equilibrium diagram of state, it turned out that the mass transfer of copper to aluminium in the conditions of explosion welding occurs at greater depths compared to the depth of penetration of aluminium into copper. This is explained by the fact that the nature of the penetrating atoms does not have such a strong influence on the conditions of thermal annealing, and the determining factor is the lattice period, which is larger in aluminium than in copper. In addition, under conditions of mass transfer, mobile dislocations play an important role, which can entrain penetrating atoms. This leads to the fact that, in less durable aluminium, deformation processes occur much more easily, and this leads to the fact that deformation processes are much easier in less strong aluminium.

3. In the explosive welding of copper with aluminium, the mutual mass-transfer coefficient does not depend on the atmosphere. Since the heating temperature of the surface layer of the metals being joined is higher when air is used, this indicates that heating due to the presence of air can be neglected, and the main contribution to mass transfer is made by the movement of atoms under the action of the driving force. It should be taken into account that the temperature in the joining zone is a determining factor that causes the formation of brittle intermetallic phases and fusible eutectic.

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