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Changes in the Structure and Properties of the Cu-Pb Composites Interface Jointed in the Solid Phase

Boris V. Borts, Aleksander A. Parkhomenko, Igor A. Vorobiev, Aleksander A. Lopata, Vitaliy A. Aleksandrov, Michael P. Domnich

National Science Center "Kharkov Institute of Physics and Technology", Kharkov, Ukraine
Email: borts@kipt.kharkov.ua

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Abstract

Research of structure and properties of Cu-Pb-Cu composite interface, obtained by technology of pack rolling of the pair of mutually insoluble metals, was performed using the methods of metallography, micro- and nano-hardness, mechanical tests, energy-dispersion elementary analyses. The work was aimed at the analyses of possible mechanical mechanisms of mass-transfer, determining the hardness of metal joint in conditions of absence of inter-diffusion. It was shown that different intensity of mass transfer of copper and lead takes place through the composite interface, which corresponds to the results obtained on the other system of dissimilar materials—copper-niobium. Qualitative explanation of these patterns was offered on the basis of more intensive plastic flow of fusible compound of the composite in conditions of roll-bond joining.

Keywords

Solid State Joint, Cu-Pb Composite, Deformation, Mass Transfer, Interface

1. Introduction

Formation of physical contact and velocity of formation of atomic bonds in the process of pressure welding are mostly depending on the velocity of formation of so called *active centers* on the joining surfaces [1]. They are formed in the areas of dislocations output on the surface of metals interface, where the atoms can tear from part of their closest neighbors, create steps on the surface and pass into the new position. Such pass is accompanied with excitation of the atom itself and of the group of the nearest neighbors—the *active center* is created.

The role of dislocation mechanisms in the creation of active centers and during the pressure welding was determined more than 50 years ago [2] [3]. These papers demonstrated that firm joint was formed in those areas where active plastic deformation took place in the subsurface layers, accompanied with motion of a large number of dislocations. At this, it was determined that "old" dislocations which were in the material before deformation, do not significantly influence on the process of pressure welding.

Dislocation mechanisms have large influence during the joining of dissimilar materials significantly different on hot strength, solubility and diffusion characteristics.

The system Cu-Pb is the typical example of such composite. Its peculiarity consists not only in the significant difference of materials melting temperatures but also in the fact that mutual solubility of components is almost absent (according to the phase diagram). In this case it can be expected that structural changes (recovery, polygonization, etc.) in this composite will take place inside of each layer without influence of diffusion fluxes from outside.

The authors of works have already managed to research deformation mechanisms, mass-transfer, bond strength of dissimilar materials, joined by solid phase rolling in vacuum under high temperature [4]-[11]. Input into mechanisms of deformation of the new carriers of rotation modes—nano-dipoles of partial disclinations and nanodipoles of torsion was analyzed [5].

The possibility of structural material solid-phase junctions' quality improvement by means of vacuum high-temperature roll bonding was shown in [6] [7] [12] [13]. The present work is intended on the analyses of changes of structure and properties of the interface of composites copper-lead, formed during the rolling under room temperature in order to determine the possible mechanisms of mass-transfer.

2. Materials and Methods of Experiment

Samples for research were prepared in the following way. Copper lead with length 120 mm, diameter 16 mm and wall thickness 1 mm was unshackled from one side. Melted lead was poured into the tube on 30% - 40%. After cooling, the obtained sample was rolled under room temperature with speed 0.15 m/sec after obtaining the flat plate with thickness 3.8 mm. Internal lead plate was obtained with thickness 2.2 mm and limited with copper sidewall with thickness 0.8 mm. In the process of rolling the copper tube and lead were subject to mutual plastic deformation. In the process of rolling of the assembled pack and its plastic deformation each of the materials changes its shape in its own way. More plastic lead obtained deformation 1.52. Copper shell obtained deformation 0.21 in the result of rolling. In order to obtain the test samples the obtained materials were treated on electric-spark machine with further etching and polishing.

Metallographic research was performed on optical microscope Olympus GX-51 and raster electronic microscope ZEISS-EVO-50 equipped with energy dispersion analyzer INCA-450. Research of changes of micro-hardness and nano-hardness through the interface of Pb-Cu was performed on micro hardness meter LECO LM-700 and nano hardness meter NANO G200 produced in the USA according to the regulations of international standard ISO 14577.

Mechanical tests were performed on flat samples with total thickness 3 mm (further "thin samples") and 4 mm (further "thick" samples) and length of the gauge length 20 mm under room temperature on the universal facility INSTRON 5581. Extension rate was 1 mm/min. Tests were performed on 3-layers samples—lead in the middle plated with copper on sides.

3. Results of the Experiment

As it can be seen from **Figure 1**, the stress-strain curves of the samples are of classical character—have the developed stage of deformation strengthening and significant uniform elongation. Using of optical record in the process of deformation allowed determining the characteristic peculiarities of composite behavior under load. It was determined that layering of materials in the result of different deformability takes place only under loads corresponding to the ultimate tensile strength. They constitute 112 - 128 MPa depending on the thickness of lead layer. This indicates on the high shear strength of the interface, which exceeds 5 - 6 times the ultimate stress of the lead under these conditions of deformation (22 - 25 MPa). As it is seen from the inserts on the figure, destruction of samples in initiated with appearance of localized under 45˚ microscopic slip-band which reflects the operation of maximal shear stress.

The structure of composite metals interface surface was investigated on meso-level by means of metallographic research. **Figure 2** demonstrates the surface of the section made in the place of roll-bond joining the copper and lead. Points of measurement of micro-hardness in the volume of materials and through the interface are marked with prints.

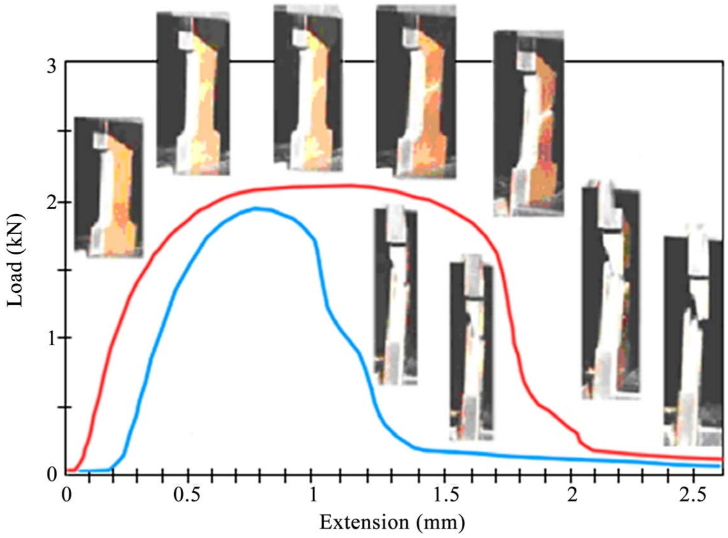

Figure 1. Stress-strain curve of the composite copper-lead under room temperature. The inserts demonstrate deformation stages and destruction of composite samples.

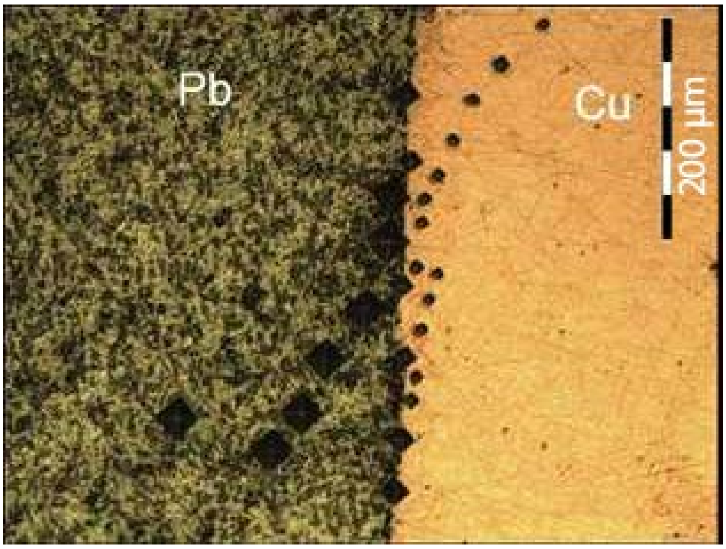

Figure 2. Interface of solid-phase joining of Pb-Cu. Prints of measurement of micro-hardness through the interface can be observed.

Figure 3 demonstrates the corresponding change of micro-hardness along the interface of the two metals. It can be observed that micro-hardness of copper is changing on the distance from the interface up to 10 μm (on the "thick" samples), while almost no change of micro-hardness of lead is observed on the mesic-scale structural level (tens of μm).

Treatment of the results of energy dispersion elementary analyses demonstrated the presence of different intensity of mass-transfer through the interface of the composite as it is shown on **Figure 4** and **Figure 5** with higher resolution. Depth of lead penetration into the copper (about 4 μm) exceeds the depth of copper penetration into the lead (about 2.5 μm).

This corresponds to the results determined earlier on the system niobium-copper (obtained using the same technology, but under high temperatures of the rolling) [4], where significantly deep penetration of copper into niobium than niobium into the copper was observed. It can be explained by the fact of intense plastic flow rate of the fusible component (and hence the mass transfer).

Research of *nano-hardness* was performed using the method of Oliver-Farah [14]. Due to this method, not only the meanings of nano-hardness of near-boundary area of the composite were obtained but also the meanings of Young modulus (E). It is important as E is connected with the strength of inter-atomic bonds and we receive the possibility to estimate the influence of alloying on its change in the near-boundary area.

From the data presented on **Figure 6**, different behavior of hardness and Young modulus in the lead and copper can be observed. If almost no changes of these values can be observed on several μm from the interface in the lead, but abrupt synchronous decrease of the modulus and nano-hardness on the distance 15 μm from the interface can be observed in copper.

Based on these results, it can be noted that softening of boundary area of the copper is connected with the decrease of degree of rigidity of atomic bonding of copper in presence of lead.

4. Discussion

The fact of penetration of the more fusible metal into the more refractory metal (based on the essential role of dislocation mechanism) in the process of rolling can be explained in the following way. The whole process of pressure welding can be divided into two stages. On the first, rather short stage, at the beginning of the process (when the area of real physical contact is rather small), abrupt increase of stress takes place which are applied to the joint materials, and plastic yielding starts in both of them. Yielding is accompanied with abrupt increase of a number of active centers, needed for joining of the surfaces of dissimilar materials.

At the same time, when keeping the values of applied loads (applied by the power of the rolling machine) unchanged, with the increase of the area of joining surface, the loading value on unit area will be decreasing, which will result in decrease of deformation intensity first of all of the more hard and more refractory material. At this the more fusible material will continue deformation, which in conditions of determining role of dislocation mechanism, will result in more intense mass-transfer into the depth of more refractory material. Due to this, taking into account the role of active centers, it is logically to assume that in particular the plastic deformation and connected with it so called *mechanical diffusion* [15] constitutes the mechanism which performs the mass-transfer in this experiment. Description of this mechanism is presented on **Figure 7**.

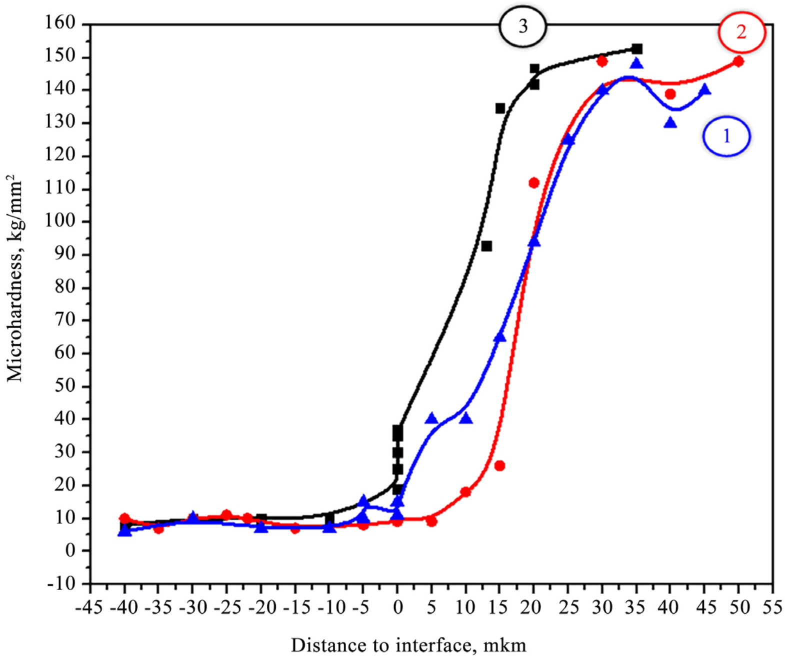

Figure 3. Micro-hardness through the interface in the solid phase Pb-Cu (1 and 2—"thin" samples, 3—"thick" sample).

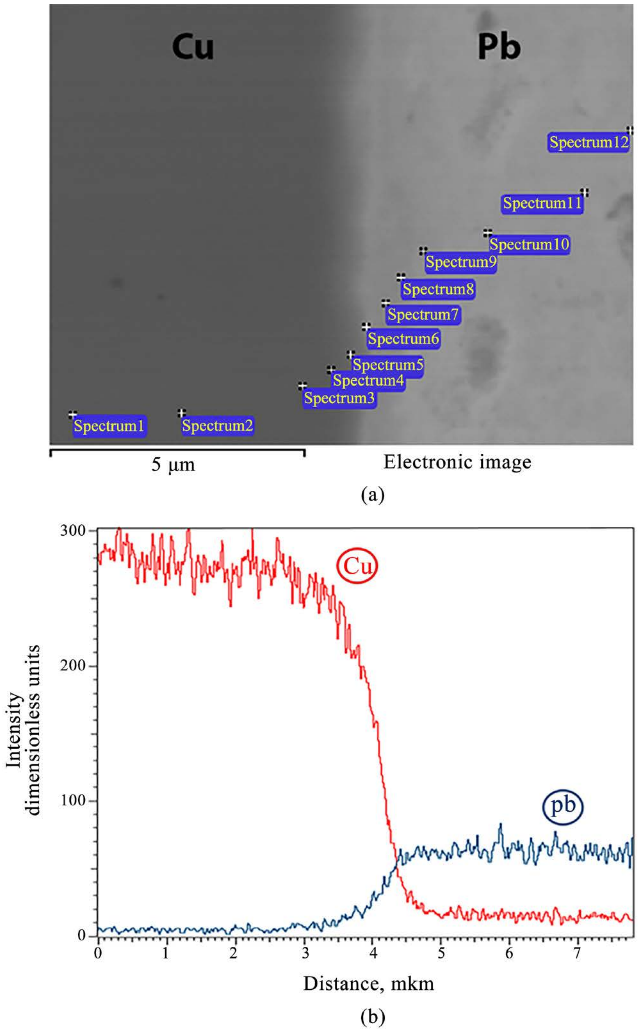

Figure 4. Electronic image (a) of the interface copper (to the left), lead (to the right) with points in which the micro-element analyses is performed (upper image) and results of analysis (b) of copper distribution (Cu-red curve) and lead (Pb-blue curve) through the interface (lower image).

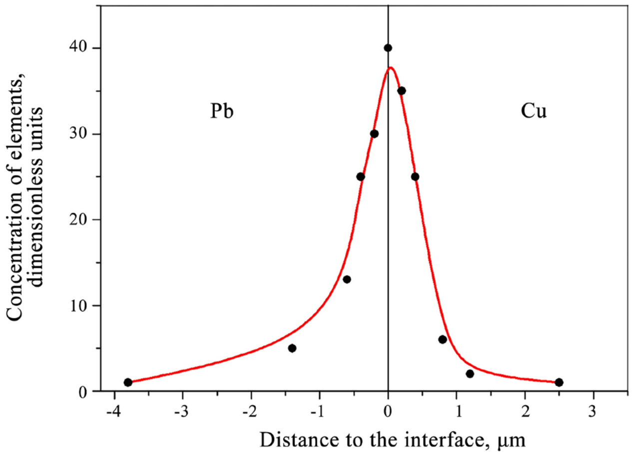

Figure 5. Dependence of lead concentration in copper (left curve) and copper in the lead (right curve) on the distance to the interface.

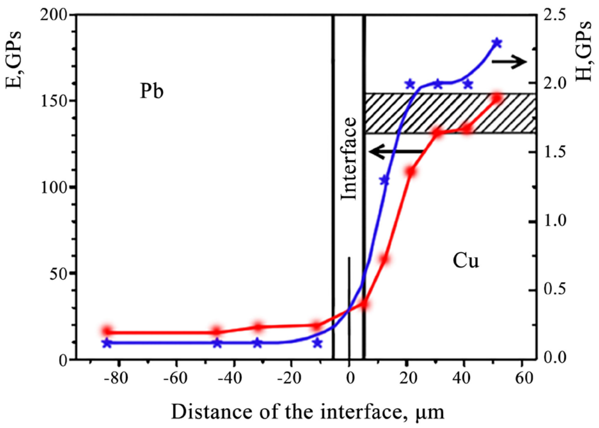

Figure 6. Change of nano-hardness (H) and modulus of elasticity (E) relatively to the interface copper-lead. Dashed lines mark the level of nano-hardness of the massive copper samples.

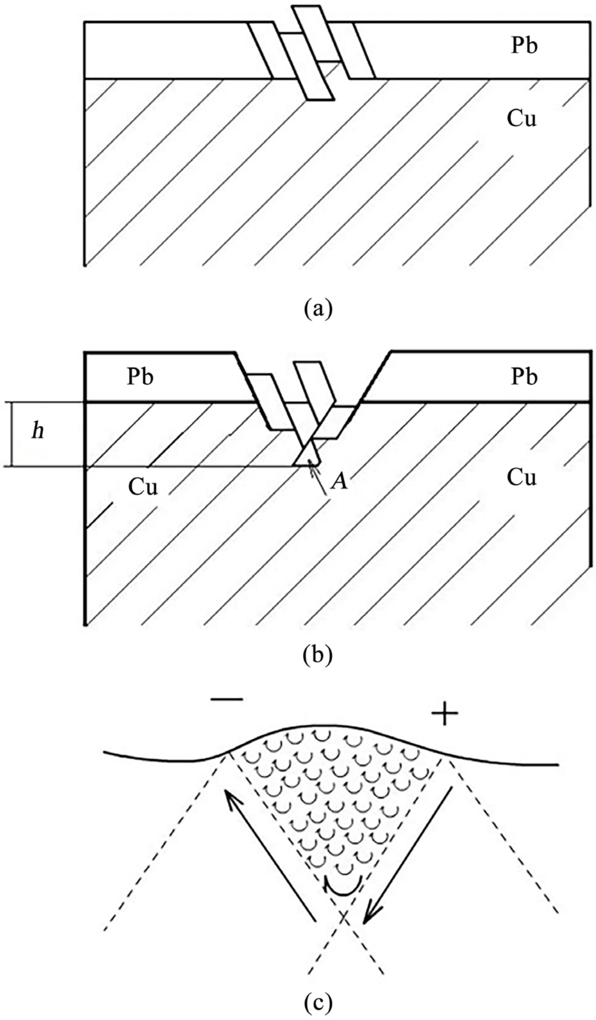

Figure 7. (a) (b) Stage-by-stage increase of the depth of penetration of lead into the copper caused by mechanical diffusion, due to development of plastic deformation localization [15]; (c) Development of rotation modes of plastic deformation on the surface of materials interface [16].

Non-homogenous, localized on all structural level nature of plastic deformation of the material, on meso- and micro-level, is also observed in creation on the surface of the deformable material of the slip-traces and slip-bands. Surface structure in the place of exposure of these traces constitutes the set of steps with height from hundreds of Angstroem units to micron depending on conditions of deformation. The steps are divided with lines of localized slipping of dislocations moving either in parallel or in conjugate planes. Each this step means the yield on the surface of the interface of a large number of dislocations ($\mathbf{n \geq 100 \ldots 1000}$). Already under level of deformation of about 20%, the height of the steps on the surface reached the micron values [15]. In particular this circumstance results in the abrupt concentration of stress ($\boldsymbol{\sigma} = \mathbf{bn}$), due to which local level of stress is reached, necessary for generation of dislocations and formation of shears in the more hard material. At the same time ejection of step into the volume of another material indicates on the flow of the process of mechanical mixing, not demanding consideration of diffusion. At this, the lead penetrates into the copper by means of shears in the direction of maximal shear stresses (**Figure 7(c)**) [17].

As it is shown in paper [15], the depth of atoms penetration (L) stipulated by mechanical diffusion is determined by a simple formula:

$$L = 2\left(b \cdot |\varepsilon|\right)^{1/2}$$

where $|\varepsilon|$—value of amplitude of the measured deformation (for lead—1.52, for copper—0.25), constant value b, according to the paper [4] is equal to about 0.3. Estimation on this formula provides for the penetration depth of the lead into the copper, the value for L, coinciding on the order of value with experimentally measured value, presented on **Figure 5**.

Determined in the last decade input of rotation modes into mechanical mixing and mass-transfer of materials [16] [17] should also be noted. According to it, under such levels of deformation, obtained by the lead, the processes of strong localizations of plastic deformation should be developing in it, which are certainly passing with participation of main carriers of rotation modes—known as disclinations. These carriers of disclinations are equal to development of micro vortexes in viscous mediums, significantly facilitating the process of mixing the materials layers on micro and further on meso-levels of deformation (**Figure 7(c)**). Mechanisms of mass-transfer with participation of nano-disclinations [17] which provide additional input into effect level have been proposed at present time.

If in paper [17], deformation value was equal to many hundreds of percents, but in paper [18], it was only several tens of percents, where by means of high-resolution electron microscopy, two new types of deformation nano-defects of rotation type were discovered, formed on the interface of dissimilar materials, joint by high-temperature rolling in vacuum. Similar to defects presented on **Figure 7**, they can also bring input into mass-transfer through the interface of the metals joined by high-temperature rolling.

5. Conclusions

1) Analysis of mechanical behavior of composites copper-lead-copper in conditions of active plastic deformation was performed. It was determined that layering of materials in the result of different deformability takes place only under load corresponding to the ultimate tensile strength. Depending on the thickness of the lead layer they constitute 112 - 128 MPa, which indicates on high shear stress of the interface, which is 5 - 6 times exceeding the ultimate stress of the lead under these conditions of deformation (22 - 25 MPa).
2) Depths of mutual mass-transfer of the components of composite lead-copper in conditions of low-temperature rolling were determined. It was shown that different intensity of mass-transfer through the composite interface takes place. Penetration depth of the lead into copper (about 4 … 5 μm) exceeds the depth of copper penetration into lead (about 2.5 μm).
3) These results correspond to the data on micro- and nano-hardness and also values of modulus of elasticity, which demonstrated invariability of these characteristics in the boundary area of the lead, and their significant decrease (in comparison with material volume) in the boundary area of the copper. At this, first, it was experimentally shown that penetration of the lead into the copper results in abrupt decrease of forces of atomic bonding of copper.
4) Compliance of the obtained patterns to the results obtained on another system of dissimilar materials—copper-niobium [4] was shown. Qualitative explanation of this pattern was offered based on more intensive plastic flow of fusible component of composite in conditions of joining by rolling
5) Taking into account the conditions of experiment (low temperatures, relatively high speeds of deformation and short times of metals interactions while rolling), the obtained results give a basis to assume that main mechanism of mass-transfer in this system can be the so called *mechanical diffusion* connected with processes of localization, multi-pole character and development of rotation modes of plastic deformation.
6) Taking into account the results obtained in the system niobium-copper, it can be concluded that likewise when joining the similar metals, neither abrupt difference in the sizes of atoms, nor absence of mutual solubility or difference in the types of crystal lattice can constitute the obstacle for formation of firm joints of dissimilar materials in the solid phase rolling.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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