Effect of annealing on the structure and phase composition of Al-Cu laminated metal-matrix composites produced by shear deformation under pressure

V N Danilenko, G F Korznikova, A P Zhilyaev, S N Sergeev, G R Khalikova, R Kh Khisamov, K S Nazarov, L U Kiekkuzhina, R R Mulyukov

Institute for Metals Superplasticity Problems, Russian Academy of Sciences, 39 Khalturin st., Ufa 450001, Russia

E-mail: vdan@anrb.ru

Abstract. This paper presents the data from the study on the effect of annealing of Al-Cu metal-matrix composites, produced by severe plastic deformation via shear under pressure. Three-layer compositions with different alternations of layers (Al-Cu-Al and Cu-Al-Cu) were subjected to deformation by high-pressure torsion, and then annealed at a temperature of 450 °C for 15 minutes. Using X-ray phase analysis and energy-dispersive analysis, we revealed the formation of the Al\(_2\)Cu, AlCu and Al\(_4\)Cu\(_9\) intermetallic compounds, surrounded by solid solutions of Al in Cu and Cu in Al. Their fraction and distribution in the volume depend on the way of stacking of the layer components. The produced microstructure is an “in situ” composite formed in the process of deformation and annealing.

1. Introduction
The need for new materials with a predetermined level of properties promotes the development of novel composite materials. The use of solid-phase processes to produce new materials is currently an intensively developing area of physical materials science. In this area, a great attention is drawn to the possibility of using deformation techniques to produce (synthesize) new materials. Among various deformation techniques for solid-phase fabrication of compounds from dissimilar materials, high-pressure torsion is of special interest is. This technique enables producing materials with a nanocrystalline structure and a large fraction of grain boundaries [1], and also stimulates phase transformations due to a high pressure and/or a high strain rate [2,3]. At present, there are several papers showing solid-phase transformations of dissimilar materials under high-pressure torsion, for example: Al-Nb [4], Al-Cu [5,6], Al-Mg [6]. The solid-phase transformations in the above-mentioned Al-based binary systems occurred in the process of deformation and annealing under various conditions.

We demonstrated earlier [7] that, after high-pressure torsion (HPT) processing at room temperature, Cu-Al-Cu and Al-Cu-Al non-porous three-layer composites were produced. Different alternations of layers resulted in different degrees of mixing of the initial components. The present study is focused on the analysis of the structure and phase formation processes during the annealing of composites with different layer alternations of Cu-Al-Cu and Al-Cu-Al, produced by HPT.
2. Experimental

In the experiment, rods of commercially pure Al (99.5%) and Cu (99.9%) were used. To produce a coarse-grained structure, Al and Cu were subjected to preliminary annealing at 400 and 900 °C, respectively. The disks with a diameter of 12.0 mm and a thickness of 0.5 mm were stacked with layer alternations of Al-Cu-Al and Cu-Al-Cu, and were subjected to deformation via high-pressure torsion under a pressure of up to 5.2 GPa by rotating a stack of disks for 10 revolutions with a rotation rate of 1 revolution per minute at room temperature. For deformation processing, anvils having a groove with a diameter of 12 mm and a total depth of 0.5 mm were used [1]. Then, halves of the produced Al-Cu-Al and Cu-Al-Cu disks were annealed at a temperature of 450 °C for 15 minutes. The temperature and duration of annealing were selected from preliminary X-ray data.

The samples were prepared using conventional metallographic techniques, followed by polishing with a colloidal silica suspension. Microstructure analysis was performed using a Tescan Mira 3LMH scanning electron microscope (SEM) with a back scattered electron (BSE) detector. The chemical composition of the Al-Cu-Al and Cu-Al-Cu samples was investigated using a Tescan VEGA 3 SBH SEM with an energy-dispersive X-ray spectroscopy (EDS) attachment.

The structure of the composites was studied in the cross section of the samples. XRD phase analysis from the surface of the studied samples was conducted on a DRON-4-07 diffractometer.

3. Results and discussion

The vertical cross sections of the Cu-Al-Cu and Al-Cu-Al composites in the as-deformed state are presented in figure 1. The first insight into the microstructure reveals that both composites are monolithic and do not contain any pores, as shown in the SEM images at low magnification in figure 1a and e. The detailed microstructure at higher magnification revealed a complex lamellar microstructure developed in each of the composites. In the central part, highlighted with square 3, both samples have three clear layers (figure 1 d and h) and look very similar to the initial layered structure, but the layers are much thinner than in the original samples before HPT. In the mid-radius area highlighted with square 2, some fine lengthened lamellas of Al in the Cu matrix and Cu lamellas in the Al matrix appear (figure 1 c and h). The latter is much more pronounced in Al-Cu-Al sample (figure 1 h). Close to the edge of both samples, highlighted with square 1, lamellas became much thinner and longer (figure 1 b and f), which is conditioned by an increase in strain from the center towards the periphery. XRD revealed the presence of α-Al and α-Cu and a negligibly small amount of the Al2Cu phase in the Al-Cu-Al sample, whereas in the Cu-Al-Cu sample only α-Cu peaks were detected.

The microstructure and the data from the EDS analysis of the annealed samples are shown in figure 2. BSE images of the annealed samples at low magnification are not significantly different from those not annealed. The use of a higher magnification made it possible to detect the emergence of individual layers of other phases at the boundaries of the initial components (areas 2, 3 and 4, 5, 6 in figure 2). For chemical analysis, we selected the most typical areas in the center, at mid-radius and at the edge of the samples. Their magnified images and the data from the EDS analysis are shown in figure 2 b, c, d and f, j, h. In the center of the Cu-Al-Cu sample, no mixing of the Al and Cu layers is observed (figure 2 b). The EDS analysis reveals the presence of solid solutions of α-Cu and α-Al, and also in the layers between the basic metals there is a compound close in composition to the Al4Cu9 phase. At the mid-radius (figure 2 c) and at the edge (figure 2 d) of the Cu-Al-Cu sample, the interaction of the layers was more intensive. The EDS analysis reveals that during the transition from the Cu layer to the Al layer, there formed successively the regions of the α-Cu solid solution, of the Al2Cu, AlCu and Al4Cu9 intermetallic compounds, and of the α-Al solid solution, which are different in contrast.

Since in the Al-Cu-Al sample HPT processing resulted in a more pronounced mixing and the formation of longer interphase Al/Cu boundaries than in the Cu-Al-Cu composite, after annealing phase transformations occurred almost in the whole volume of the sample (figure 2e). The EDS analysis revealed the presence of solid solutions of α-Al and α-Cu and the Al2Cu, AlCu and Al4Cu9 intermetallic compounds in all the studied areas during the transition from the Al layer to the Cu layer (figure 2 f-h).
The data from the EDS analysis of the Cu-Al-Cu and Al-Cu-Al samples are in good agreement with the data from the X-ray phase analysis (figure 3). Alongside with the lines from $\alpha$-Cu, diffraction peaks belonging to $\text{Al}_3\text{Cu}$, AlCu and $\text{Al}_4\text{Cu}_9$ also appear in the Cu-Al-Cu sample. In the Al-Cu-Al composite, the process of structural and phase transformations is more pronounced, which follows from a significant increase in the quantity and intensity of the diffraction peaks belonging to $\text{Al}_3\text{Cu}$, AlCu and $\text{Al}_4\text{Cu}_9$. As noted above, the energy-dispersive analysis shows that Cu and Al are present only in the form of a solid solution with different contents of the solute element. This is confirmed by a displacement of the diffraction peaks from Cu and Al, related to a change in the lattice parameter in solid solutions of $\alpha$-Cu and $\alpha$-Al.
Figure 2. BSE image of the Cu-Al-Cu (a-d) and Al-Cu-Al (e-h) samples at a small magnification after annealing (a, e), microstructure details in the center (1, 4), at mid-radius (2, 5), at the peripheral area (3, 6). Elemental distribution along the line depicted at the top of the picture in the form of a graph of relative intensities with relation to the scanning distance for Cu and Al, marked in green color and in red color, respectively, in the center (b, f), at mid-radius (c, j) and in the peripheral area (d, h).
Figure 3. XRD of the Cu-Al-Cu and Al-Cu-Al samples after annealing at 450 °C for 15 minutes.

As can be seen from figure 2, the Al2Cu, AlCu and Al4Cu9 intermetallic compounds are surrounded by solid solutions of Al in Cu and Cu in Al. If we take into account that the microhardness of intermetallic compounds is higher than that in the solid solution formed during annealing [4], we see a microstructure where the “hard” intermetallic phases are surrounded by a “soft” solid solution. In other words, during the annealing of the deformed three-layer composites, a structure is formed, which can be called an “in situ” composite.

4. Summary
Thus, the X-ray phase analysis and the study by scanning electron microscopy with energy-dispersive analysis showed that the three-layer stacking of the samples for deformation has an effect on the process of solid-phase mixing and phase formation during annealing. In the Cu-Al-Cu and Al-Cu-Al samples, during annealing there occurs the formation of the Al2Cu, AlCu and Al4Cu9 intermetallic compounds, which are surrounded by solid solutions of Al in Cu and Cu in Al. In the Al-Cu-Al sample, the phases that have emerged are distributed throughout the sample, and their proportion is greater. The produced structure can be called an “in situ” composite.

Acknowledgements
The present work was accomplished according to the state assignment of IMSP RAS and supported by the Russian Science Foundation (Grant No. 18-12-00440).

References
[1] Zhilyaev A P and Langdon T G 2008 Prog. Mat. Sci. 53 893
[2] Perez-Prado M T, Gimazov A A, Ruano O A, Kassner M E and Zhilyaev A P 2008 Scripta Mater 58 219
[3] Straumal B B, Mazilkin A A, Baretzky B, Schutz G, Rabkin E and Valiev R Z. 2012 Scripta Mater 53 63
[4] Danilenko V N, Galushev S N and Mulyukov R R 2009 Perspektivnye Materialy 7 94 (in Russian)
[5] Oh-ishii K, Edalati K, Kim H S, Hono K, Horita Z 2013 Acta Mater. 61 3482
[6] Kawasaki M, Han D K Han J K, Jang J and Langdon T G 2017 IOP Conf. Series: Mater. Sci. Eng. 194 012002
[7] Korznikova G, Mulyukov R, Zhilyaev et al., 2018 AIP Conf. Proc. (in press)