EFFECT OF SINTERING CONDITIONS ON STRUCTURAL PHASES AND FORMABILITY OF Al-Cu POWDER MIXTURE

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Abstract

This experiment deals with preparation of Al plus 25 weight % Cu powder mixture. The powders were mixed, compressed using a double-step cold isostatic pressing (CIP) procedure and subsequently sintered in vacuum either at 500 °C or at 550 °C for 60 min. Analyses of chemical compositions of the pre-sintered samples performed using scanning electron microscopy showed a distinctive difference in structural phases compositions in the two samples featuring a presence of aluminum oxides (Al_2O_3) and brittle intermetallic phases ($CuAl_2$) in the two samples, respectively. The presence of both was also confirmed by microhardness measurements. The effects of the individual structural phases on formability of the two pre-sintered samples were evaluated with severe plastic deformation via high pressure torsion (HPT). While the sample featuring alumina exhibited sufficient formability at room temperature, the other sample featuring intermetallics was formable only at an elevated temperature.

Keywords: Powders, intermetallics, alumina, cold isostatic pressing, high pressure torsion

1. INTRODUCTION

For its excellent conductivity, copper is mostly used especially for electrotechnical applications, although its cost is relatively high. There have already been efforts to replace copper wires with aluminum ones, especially for high-voltage applications. However, the properties of Al are not sufficient to enable full replacement of Cu. Possible solutions are improvement of mechanical properties via structural refinement [1] and extreme plastic deformation[2, 3], or development of Cu-Al composites [4, 5] The combination of the two materials is economically favourable, since additions of aluminum decrease costs of the final products and their weights, which can also result in substantial cost savings [6-8]. The Al-Cu composite system features high thermal and electrical conductivities and low density (depending on the ratios of the individual elements). Cu/Al composites could also in future substitute Cu in automotive industry and electrotechnics. Nevertheless, the system is known for its tendency to form brittle intermetallics. These were found to form even on the interfaces between individual layers in clad composites [9]. Among the possibilities to reduce formation of intermetallic phases are especially optimization of fabrication conditions (sintering conditions for powders) and homogenization of structure, advantageously via methods of intensive and severe plastic deformation. For composites consisting of particles, complete mixing and small final structural units enhancing properties of the final materials can be supported by a combination of intensive plastic deformation and powder metallurgy [10-12].

The aim of this study was to find out the influence of various sintering conditions on the structure and content of phases in Al-25Cu powder mixture. These were evaluated using scanning electron microscopy analyses and microhardness measurements. The sintering temperatures were selected according to the Al-Cu binary diagram (temperatures of 500 °C and 550 °C, the first below and the second just above the eutectic temperature for this system) [13]. Subsequently, the pre-sintered samples were subjected to high pressure torsion (HPT). HPT is a representative of severe plastic deformation methods, which imposes the most

intensive shear strain into materials [12, 14] and thus should affect consolidation of the pre-sintered samples the most favourably. Processing by plastic deformation was performed to evaluate formability of samples with different structures.

2. EXPERIMENT

Powder fractions of particles smaller than 45 μm were selected and sieved for both the elements. The chemical compositions of the initial powders, determined by EDX analysis were as follows: Al - 96.1 wt.% Al and 3.9 wt.% O, Cu - 94.1 wt.% Cu and 5.9 wt.% O. The powders were mixed in the ratios of 75 wt.% Al and 25 wt.% Cu using a TURBULA powder mixing/stirring device. The mixture was subsequently pre-compacted using a double-step CIP procedure at 200 + 300 MPa into the form of little rods with diameters slightly larger than 20 mm, which were subsequently subjected to vacuum sintering. Sintering was performed at Mabave Company, Horka nad Moravou, CZ either at 500 °C or at 550 °C for 60 minutes. Heating of both the samples was performed at the rate of 10 °C/min, cooling was performed in the furnace down to the temperature of 100 °C and further on air. After sintering, the small rods were sliced using water jet cutter to prepare samples for analyses and deformation processing. The samples were subjected to 0.5 HPT revolutions at 0.5 RPM and 5 GPa; 0.5 revolutions were selected in order to find out the influences of various amounts of relatively low imposed strain on structural changes. The sample pre-sintered at 500 °C was processed at room temperature, while the sample pre-sintered at 550 °C was processed at 150°C due to the presupposition of higher brittleness of the sample [9]. Analyses were then performed on vertical cross-sectional cuts led through the axes of the samples. Samples for microscopic analyses were ground on SiC papers and polished by alumina particles and a soft cloth. The analyses were performed by a Tescan Lyra 3 FIB/SEM microscope and a Vickers microhardness testing device with the load of 100 g and dwell time of 15 s.

3. RESULTS AND DISCUSSION

3.1. Structural observations and phases

Figures 1a and **1b** clearly show the macro-scale difference between the two sintered structures. **Figure 1a** depicts the structure of Al-25Cu sample after compression and sintering at 500 °C. The dominant dark regions consisted of aluminum particles, while the brighter areas with dendritic shapes were copper particles. A subsequent SEM-EDX analysis depicts the composition of the sample more clearly. The **Figure 1** also shows a significant amount of pores. ImageJ analysis performed on five random scans of the sintered structure showed the average porosity of 1.8%. However, the content of pores together with the darkest inter-particle areas, which were most probably oxides, was significantly higher - 25.7% in average. **Figure 1b** then shows a detailed image of microstructure of the second sample sintered at 550 °C. This structure exhibited a better degree of sintering with no substantial presence of oxides on the boundaries of the particles. On the other hand, there was another substance filling most of the pores between the particles, which contributed to a significant decrease in porosity. ImageJ analysis showed the average porosity for this sample to be as low as 0.5%. Decreasing porosity with increasing content of Cu in a composite structure was already reported e.g. by Wolla et al. [15]. The substance generated during sintering was most probably an intermetallic phase or low-melting eutectics. In order to find out chemical compositions of the compounds within the two samples, point analyses of chemical compositions in several locations within the samples were performed. The analysed locations within both the samples are depicted in **Figures 2a** and **2b**, the results are summarized in **Table 1**.

For the sample sintered at 500 °C, the grey regions consisted of aluminum, the bright regions consisted of copper, and the darkest regions consisted of aluminum oxides. On the other hand, the grey regions within the sample sintered at 550 °C consisted almost exclusively of aluminum - the contents of Cu for locations 2 and 5 were negligible and could have been caused by influences of below located particles. Chemical compositions of the lighter locations then corresponded to the composition of $CuAl_2$ intermetallic phase. This phase

consumed the majority of the original Cu content. According to the Al-Cu binary phase diagram, a pre-sintered sample containing 75 wt.% Al and 25 wt.% Cu should contain Al and $CuAl_2$ intermetallic phase [16]. This difference in structures of the two samples is considered to be caused by the different temperatures. While sintering at 500 °C for 60 minutes was not sufficient for intermetallics to develop and a significant portion of oxides remained in the structure, the temperature of 550 °C was already enough for generation of $CuAl_2$. The favourable influence of this is on decreasing the overall porosity of the sintered sample by development of the intermetallic phase. However, a content of brittle intermetallics can lower plasticity and formability of the material, although it is supposed to increase its hardness. Severe plastic deformation via HPT was already shown to be successful for Al-based materials containing Al_2O_3 [14]. Nevertheless, the influence of HPT on a structure with a significant content of intermetallics was still to be investigated.

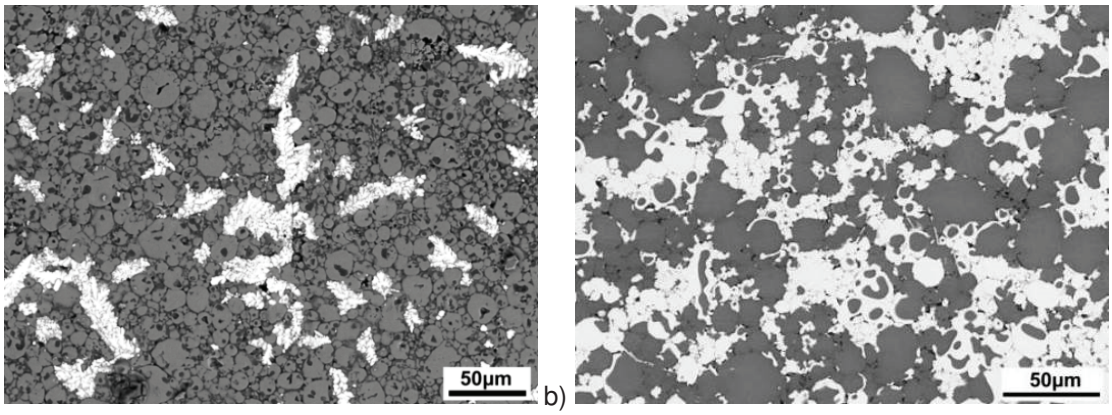


Figure 1 Scanning electron microscope images of samples sintered at (a) 500 °C; (b) 550 °C (BSE mode)

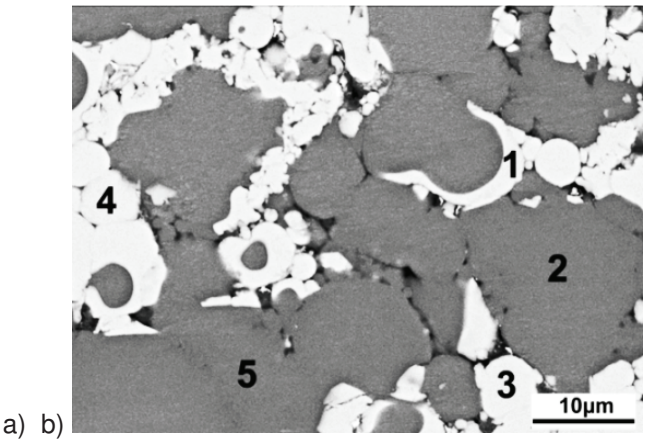


Figure 2 Locations of chemical composition analyses in samples sintered at (a) 500 °C; (b) 550 °C

Table 1 Phases in the Al-25Cu samples (weight %)

sample		**500 °C**	**500 °C**	**500 °C**	**500 °C**	**550 °C**	**550 °C**	**550 °C**	**550 °C**	**550 °C**
location		**1**	**2**	**3**	**4**	**1**	**2**	**3**	**4**	**5**
content	*Al*	0.8	90.8	52.6	45.8	96.8	45.3	45.1	99.2	70.4
	Cu	99.2	0.1	43.8	54.2	3.2	54.7	54.9	0.8	0.2
	O_2		9.1	3.6						29.4
phase		Cu	Al + Al_2O_3	$CuAl_2$	Al + Al_2O_3	$CuAl_2$	Al	$CuAl_2$	$CuAl_2$	Al

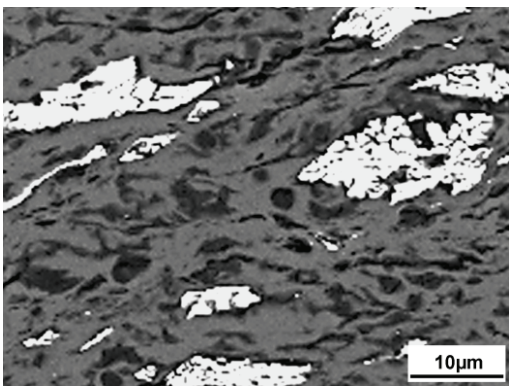
3.2. Structural observations and imposed strain after HPT

The primary aim of HPT processing was to investigate formability and to observe structure changes and its inhomogeneity caused by different amounts of the imposed strain in different locations of the processed samples. The imposed strain in the evaluated locations was calculated using Eq. (1),

$$\varphi = \frac{2\pi r N}{t} \quad (1)$$

where r is radius of the sample deformed using HPT (in the measured location) (mm), N is number of revolutions and t is thickness of the sample (mm). The strain imposed into the sample in the peripheral areas was approximately 6 (calculated radius 9.5 mm), while in the mid-radius location it was approximately 3 (average value for calculations for radii 4.5 and 5 mm).

The first sample pre-sintered at 500 °C already exhibited grain structure in the most deformed areas (**Figure 3a**), contrary to the second sample pre-sintered at 550 °C, within the structure of which grains were not clearly observed. However, the influence of the imposed strain on defragmentation of clusters of the intermetallic phase could be observed in the middle and peripheral areas (**Figure 3b**) of the sample. Especially the first sample then exhibited localized shear bands between its central (least deformed) and peripheral (most deformed) areas. Such a structure development is typical for HPT processing and was conformant to simulations performed e.g. by Yoon et al. [17]. Although the amount of the imposed strain was not so significant to cause full consolidation of the structures, two important results concluded from the deformation processing. Firstly, the conditions of high pressure caused both the structures to be formable, although the sample pre-sintered at 550 °C containing intermetallics had to be deformed at elevated temperature (the intermetallic phases are known to significantly decrease formability of Al/Cu composites [13]). Secondly, the porosity was lower than for the pre-sintered structures. The average content of pores decreased, according to ImageJ analyses, to approximately 0.3% for both the samples. Considering the original values of 1.8% and 0.5% for the 500 °C and 550 °C samples, respectively, the decrease in porosity was more substantial for the first sample, which suggests its better formability, even though the processing temperature for this sample was lower.

a) 

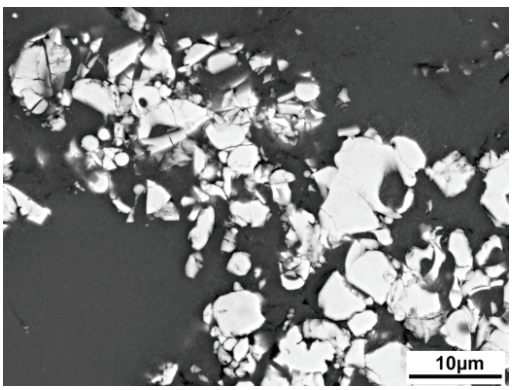
b) 

Figure 3 Structures of HPT-processed samples; (a) pre-sintered at 500 °C; (b) pre-sintered at 550 °C

3.3. Microhardness

Microhardnesses of the pre-sintered and HPT-processed samples were measured in 20 individual points (starting 0.5 mm from its edge) along the horizontal axes of the samples on transverse cross-sectional cuts.

The average microhardness for the sample pre-sintered at 500 °C was 25.2 HV and it was comparable to previously published results for Al samples pre-sintered at comparable conditions [14]. However, it was lower than for a cast Al-Cu sample processed with 0.5 HPT revolutions, as reported by Mohamed et al. [18]. Microhardness of the 500 °C sample increased to the average value of 70.6 $HV_{0.1}$ after 0.5 HPT revolutions.

The main reason for this was a transformation of the imperfectly sintered structure into a more homogenous one and development of grains induced by HPT. The average microhardness of the second pre-sintered sample featuring a significant amount of intermetallics was 91.4 $HV_{0.1}$ with quite a high standard deviation of 17.8, which suggested substantial differences in hardnesses of the regions of Al and $CuAl_2$. Comparing the two average microhardnesses, formation of intermetallics evidently caused a significant increase in this value. The microhardness then increased to the average value of 98.8 $HV_{0.1}$ after HPT and the standard deviation decreased to approx. 6, which was comparable to the deviation of the deformed sample pre-sintered at 500 °C. The distribution of strain throughout both the samples is depicted in **Figure 4**. The results did not show any clear tendency of increasing/decreasing HV values throughout the cross-sections and were mostly corresponding to the local microstructures in the observed locations. This was caused by the low imposed strain insufficient for a complete consolidation and transformation of the structure.

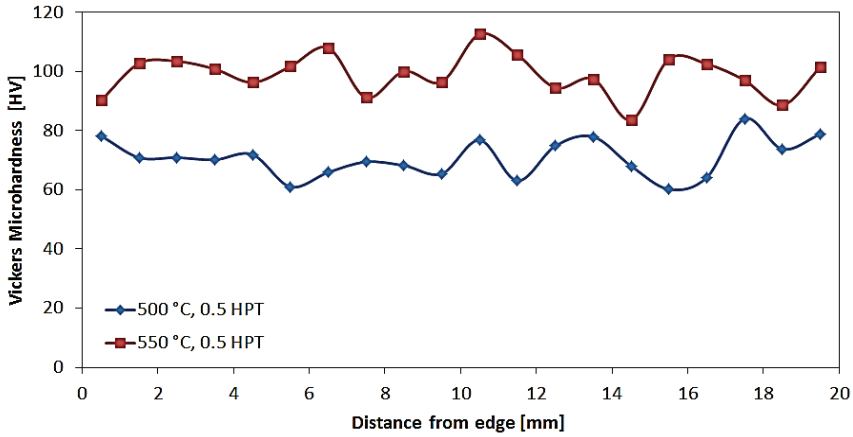


Figure 4 Distribution of microhardness throughout the HPT processed samples

4. CONCLUSIONS

Powder samples of the Al-25Cu composition were prepared by mixing of particles of both the elements, compacting by a double-step CIP and subsequent vacuum sintering for 60 minutes at 500 °C or at 550 °C. Chemical composition and phase observations showed the presence of Al, Cu and aluminum oxides in the first sample, while the second one exhibited a significant formation of the $CuAl_2$ intermetallic phase. This difference was caused by the sintering temperature, since 500 °C was below the eutectic temperature in the Al-Cu binary diagram, while 550 °C was slightly above it. The second pre-sintered sample also exhibited higher microhardness of 91.4 $HV_{0.1}$, comparing to 25.2 HV of the 500 °C sample. Processing of both the samples by 0.5 HPT revolutions showed sufficient formability of both the structures at high-pressures, although the sample featuring intermetallics had to be deformed at an elevated temperature (150 °C). The decrease in porosity was more significant for the 500 °C sample, which suggests its better formability when compared to the sample featuring intermetallics. The favourable influence of HPT was proven also by the increase in average microhardness values for both the samples. The increase was more substantial for the sample with oxides, which imparts its better formability when compared to the sample with intermetallics. The subsequent step of this research should be focused on measurement of electro-conductivity and a deeper analysis of mechanical properties.

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