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THE ROLE OF THE MATRIX IN SiC REINFORCED COMPOSITES

This article presents a comparison of the properties of composites based on aluminum or aluminum alloy (Al4Cu) reinforced with silicon carbide SiCp. The main objective was to analyze the possibility of producing an Al + Cu alloy matrix by basic powder metallurgy methods and its influence on the final properties of the composite. The composites were produced by pressing and sintering, basic powder metallurgy techniques, in order to reduce the manufacturing costs. Sintering was carried out in nitrogen due to the favorable effect of this atmosphere on the sintering of aluminum-based materials. Silicon carbide SiC was used as the reinforcing phase. The study clearly showed that the use of a matrix made of a mixture of Al and Cu powders results in an almost twofold increase in hardness (from 32 to about 60 HB) and a more than twofold increase in flexural strength (from about 200 to more than 450 MPa). Observations of the microstructure confirmed the diffusion of copper into the aluminum and the facets of the Al2Cu phase.

Keywords: aluminum matrix, SiC particles, powder metallurgy, properties, microstructure

INTRODUCTION

Composite materials based on light metals are a response to the growing requirements for strong lightweight structures and the need to reduce the weight of elements while maintaining proper strength, especially in the automotive and aviation industry. The lightweight metal matrix consists mainly of aluminum, magnesium or titanium and their alloys [1]. Among those mentioned, a wide group of construction materials are aluminum-based composites, which have gained popularity owing to their several unique properties including low density, high relative strength, good dimensional stability, machinability, high electrical and thermal conductivity, corrosion resistance and low price [2-8]. One of the main advantages of metal matrix composites in relation to metals without the addition of a strengthening phase is increased stiffness and strength [9].

When designing composite materials, their specific weight is taken into account, and due to economic aspects – the cost associated with the production of the composite. The final properties of composites are influenced by a number of factors, which include the type of the matrix and strengthening phase, the volume ratio of the individual components of the composite, the geometry and the distribution of the strengthening particles, their orientation and size. The addition of silicon carbide particles to the aluminum matrix has a significant influence on the mechanical properties of aluminum. Studies on the correlation between the weight fraction of the SiC phase and the mechanical properties presented by authors [10-13] show that the strength, hardness and corrosion resistance of Al-SiC composites grow with the increase in the SiC particle content. The introduction of a hard SiC strengthening phase to the aluminum matrix allows significant improvement to be achieved in abrasion resistance, which rises with the increment in the content of strengthening phase [14-16]. Nonetheless, Mahdi et al. [17] observed that an increase in the weight fraction of SiC particles in aluminum matrix composites contributes to a higher corrosion rate.

The microstructure and final properties of metal matrix composites are strongly affected by the method of the manufacturing process and the reaction products formed during compaction. Mi et al. [18] indicated the formation of the Al_4C_3 phase as a result of the reaction between molten Al and SiC in Al-SiC composites fabricated by concurrent wire-powder feeding laser deposition. The Al_4C_3 phase located at the interface between SiC and Al contributes to the brittleness of the composite as it represents areas of potential crack initiation and propagation. Singh et al. [19] showed the impact of the process parameters (stirring speed and duration) on the wear, tensile strength, and hardness in Al-SiC composites fabricated using stir casting. The rotational speed influences the refinement of the composites and the homogeneous distribution of the reinforcing phase.

The mechanical and tribological properties of aluminum-based composites can also be increased by adding a copper to the aluminum matrix [20]. Mahamood et al. indicate the weight loss of the material subjected to tribological tests was lower for alloys containing copper compared to composites based on pure aluminum. Mei et al. [21] fabricated AlCu-SiC composites with enhanced mechanical performances using accumulative roll-bonding. The authors indicated that the introduction of copper to the aluminum matrix allowed the maximum tensile strength to be achieved of AlCu-SiC composites, which was twice that of the AlCu matrix and ten times that of the pure Al matrix. Composites based on an aluminum-copper alloy can also be subjected to precipitation hardening; as a result, an increase in the mechanical properties can be obtained. However, composite materials with the addition of SiC particles require different temperature parameters of heat treatment than the unreinforced AlCu matrix [22]. Bedir [23] reported that as a result of heat treatment of a composite based on an Al alloy with an addition of 5 wt.% Cu, the hardness increased from 70 to 107 HB. Commercial age-hardenable aluminum alloys applied as the matrix material in aluminum alloy matrix composites (AMC) are 7075 (AleZneMg) and 2024 (AleCueMg) [24, 25] series and AMC based on aluminum alloy 6061 (AleMgeSi) [26].

The purpose of the present study was to produce composites of aluminum and silicon carbide powders, as well as aluminum, copper and silicon carbide powders using basic powder metallurgy technologies. The produced composites were subjected to density, hardness and flexural strength tests in addition to microstructure observations. This will make it possible to compare these materials in terms of the influence of the matrix on the final properties and microstructure while maintaining similar contents of the reinforcing phase.

MATERIALS AND METHODS

Investigations were carried out on composite materials based on aluminum and its Al-Cu alloy with the addition of a strengthening phase in the form of SiC particles. The matrices of the composites were made of elemental powders. The main component of the matrix was aluminum powder grade 1070, sprayed with argon, produced by the Benda-Lutz company. The particle size of the aluminum powder was on average below 63 μm (the powder fraction consisted of the following fractions: above 63 μm – 5%, in the range of 32-63 μm – 45-70%, the rest – below 32 μm). Electrolytic copper powder grade ECu1, produced by Euromet, with a particle size below 40 μm, was used to produce composites based on an aluminum-copper alloy. Silicon carbide particles (SiC-α) obtained as a result of synthesis, produced by Alfa Aesar, were used as the strengthening phase. Silicon carbide with a particle size ranging from 40 to 60 μm was employed.

Figure 1 shows the particle morphology of the powders of the starting material used to produce the composites.

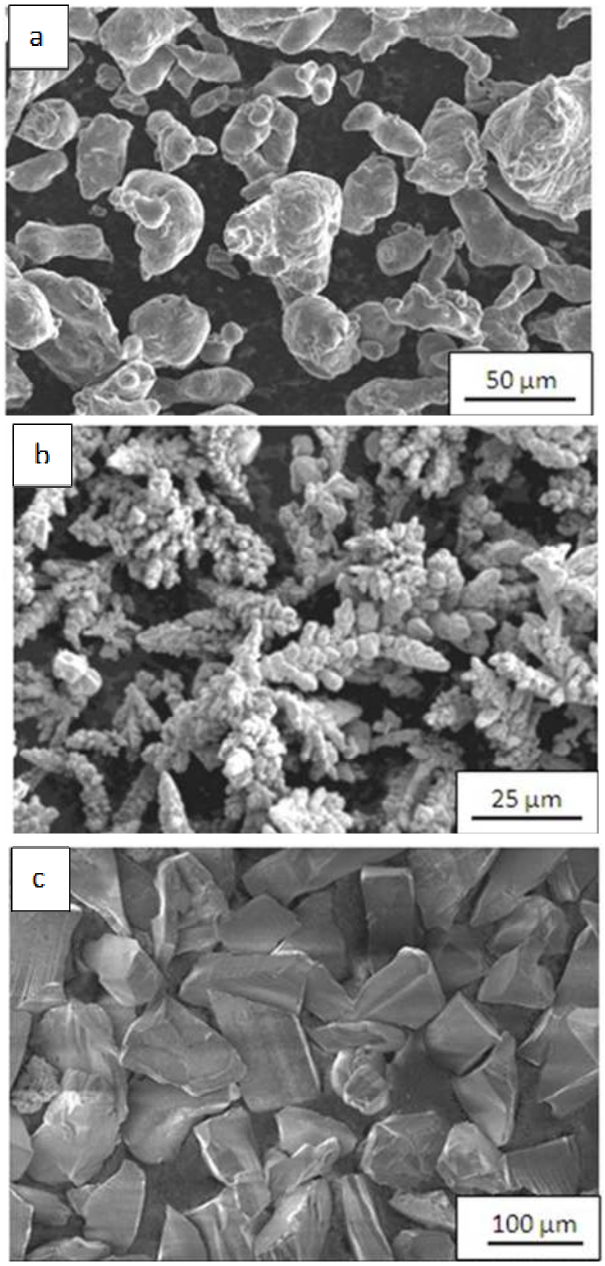


Fig. 1. SEM morphologies of used powders: a) Al, b) Cu, c) SiC

Aluminum powder particles sprayed with argon have a globular shape and have a relatively smooth surface. The copper powder obtained in the electrolysis process has a dendritic shape of particles with a large number of branches and a highly developed surface, which is characteristic for this method of production. The individual grains of the material of the SiC strengthening phase are generally irregular in shape, which means that their linear dimensions are not the same in different directions.

The starting material in the form of aluminum, copper and SiC particles was weighed in appropriate weight proportions allowing the production of a matrix material with two different chemical compositions, one of which was pure aluminum powder, and the other was an aluminum alloy with the addition of 4% by weight of copper. Strengthening in the form of ceramic silicon carbide particles was introduced to the two base materials prepared in this way, constituting the matrix of the composites. In the first stage of producing composites, the elementary powders were mixed in appropriate proportions in a Turbula T2F turbulent mixer. The mixing time for the powder mixtures was one hour.

TABLE 1. **Chemical composition of powder mixtures**

Al matrix	**Al4Cu matrix**
Al + 5 wt. % SiC	Al + 4 wt. % Cu + 5 wt. % SiC
Al + 10 wt. % SiC	Al + 4 wt. % Cu + 10 wt. % SiC

On the basis of previous tests, the team selected the pressing pressure of 300 MPa and sintering temperature of 600°C. These are the best production parameters from the point of view of economy and the obtained properties, especially since double pressing and double sintering were used.

Subsequently, the hardness of the as-sintered samples was tested (Brinell method, tungsten carbide ball, Ø2.5 mm, 613 N), the flexural strength was measured and microstructural examinations were conducted by means of scanning electron microscopy (SEM, Hitachi Su70, Japan). The phase identification of the composites was carried out using a Tur 62 X-ray diffraction apparatus (Germany) with a Cu target (Kα, $\lambda = 1.5406$ Å).

RESULTS

The first stage of the research was to produce sintered composite materials by double pressing and sintering. The first sintering, in addition to its densification function, was aimed at recrystallization, which made repressing possible. The results of the relative densities of the composites at each stage of their manufacture are shown in Figures 2 and 3, and a summary of the final densities for both matrix types is presented in Figure 4.

The results of the density measurements at the various stages of composite manufacturing presented in Figures 2 and 3 show that the stage responsible for the increase in density is pressing and trimming. Therefore, the selection of the optimum pressing pressure is extremely important. Interestingly, the addition of copper does not increase the density in the powder volume when poured into the die, after pressing, but rather lowers it slightly, resulting in a lower final density of the composites (Fig. 4). Copper powder is characterized by its dendritic shape and is relatively sparsely distributed in the powder volume when poured into the die. This can affect the formation of powder breakers, which consequently reduce the effect of the pressing pressure as they have to breakdown during compaction.

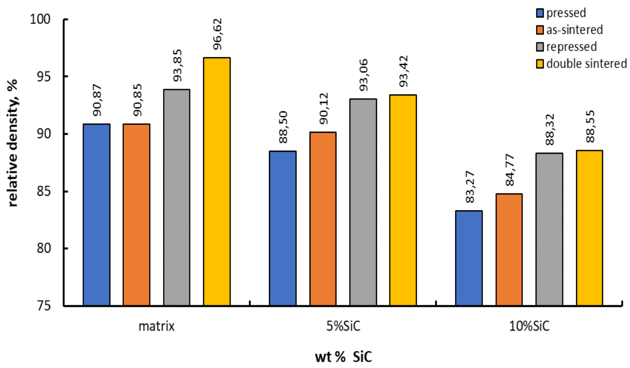


Fig. 2. Influence of different manufacturing steps on density of aluminum matrix composites

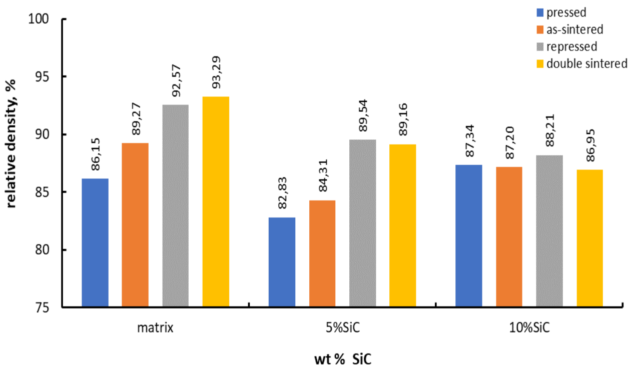


Fig. 3. Influence of different manufacturing steps on density of Al4Cu matrix composites

Due to their strong affinity for oxygen, the aluminum particles are coated with a thin layer of alumina in the form of a continuous film, which acts as a barrier to the proper bonding of the individual components, and can thus directly affect the final properties of the material. On the other hand, the presence of finely dispersed Al_2O_3 particles can have a positive effect on the material properties by acting as an additional strengthening phase. During pressing, the oxide coating present on the surface of the aluminum powder particles can fracture, allowing the adhesion force between the individual aluminum particles to increase and strong intermetallic bonds to form. One of the theories regarding the activation of the sintering process is that this layer is involved in the sintering process. Its cracking, which can result from a number of factors, is still being analyzed. The nitrogen atmosphere used for sintering plays a critical role. The alumina layer covering the aluminum powder particles is usually amorphous and thermodynamically unstable; therefore, it can easily be transformed into a crystalline form during the sintering process. This is mainly due to the fact that as a consequence of oxidation, the critical thickness value of the oxide film in the amorphous form is exceeded, which in turn causes a loss of stability and an increase in temperature above about 400°C, initiating its structural changes. As a result of the transformation from the amorphous structure of Al_2O_3 to the crystalline form of γ-Al_2O_3, owing to the difference in densities, the integrity of the continuous oxide film is compromised, and thus the active metal surfaces are exposed, allowing direct contact between the aluminum and the sintering atmosphere. The disruption of the oxide film during heating also occurs due to the presence of the inherent stresses generated by the large difference in the values of the thermal expansion coefficients between pure aluminum and the γ-Al_2O_3 oxide. When Al-SiC composites are sintered in a nitrogen atmosphere, aluminum oxide can be reduced by nitrogen and form aluminum nitrides according to reaction (1):

$$2Al_2O_3 + 2N_2 \rightarrow 4AlN + 3O_2 \qquad (1)$$

The formation of aluminum nitride is a highly exothermic reaction that can occur when nitrogen is left in direct contact with a freshly exposed aluminum surface. As already mentioned, in order to initiate the reaction between aluminum and nitrogen, the strong triple bonds present in the nitrogen molecule must be broken. This requires a large source of energy, which in turn comes from the energy released by the repeated exothermic allotropic transformations of aluminum oxide.

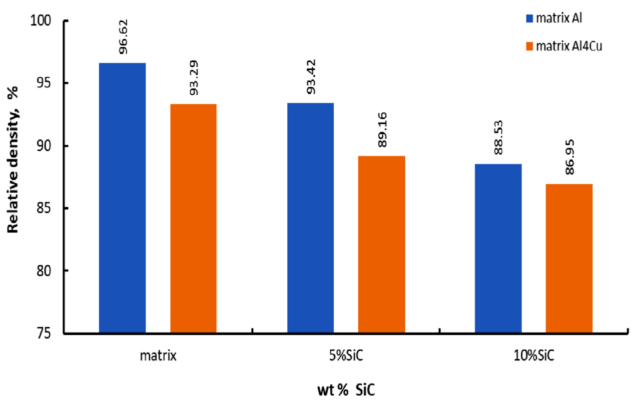


Fig. 4. Summary of final densities of composites

In the case of aluminum matrix composites, the hardness grows with an increasing SiC content (Fig. 5), but only slightly – by only 3 units. This is partly owing to the lowering of their density as the proportion of SiC increases. In the case of the Al_4Cu matrix composites, the hardness also increases less than expected considering the SiC content. However, changing the matrix to an Al_4Cu alloy resulted in a significant rise in hardness with respect to the aluminum matrix composites. The obtained hardness measurements indicate microstructural changes in the matrix; strengthening resulting from separation of the Al_2Cu phase.

A three-point bending test was performed to determine the plastic properties and quality of the bond at the reinforcing phase (SiC) matrix interface. The results are summarized in Figure 6.

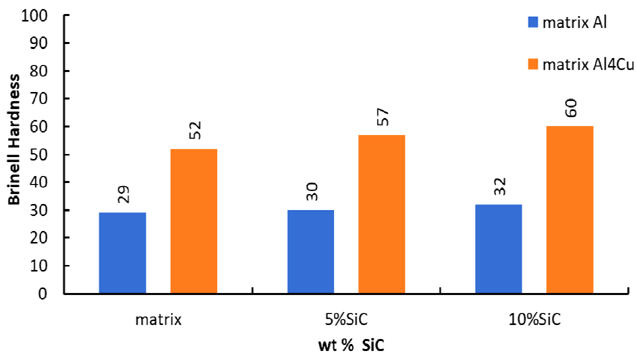


Fig. 5. Brinell hardness of composites

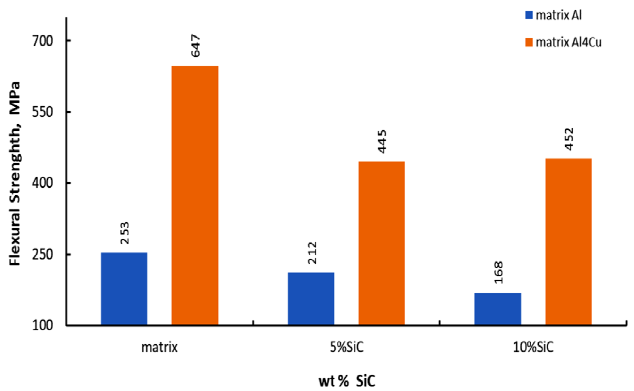


Fig. 6. Flexural strength of composites

The strength of the metal-ceramic interface in the tested composites was determined by the three-point bending test. As expected, the addition of SiC reduces the flexural strength as its proportion increases. The main difference is due to the type of matrix; replacing aluminum with the Al_4Cu alloy results in a significant increase in flexural strength. The increase in strength is significant; it also means that the bond between such a matrix and the reinforcing phase is improved. Mei et al. [21] demonstrated that it is possible to increase UTS to values of more than 600 MPa in composites by changing the matrix to an AlCu alloy, where strengthening was achieved without additional heat treatment. Ogel and Gurbuz [11] found that when producing such composites by hot pressing, results slightly exceeding 300 MPa are obtained. This indicates the main problem arising from sintering activation by using a nitrogen atmosphere for sintering.

Figures 7 and 8 present the microstructures of the sintered aluminum and Al_4Cu alloy.

The microstructure of the matrix material consists mostly of regular aluminum grains and a few pores which are located at the boundaries (Fig. 7). In addition, it can be stated that the precipitates are located mainly on the grain boundaries in the sintered Al4Cu alloy (Fig. 8). The microstructures of the composites are summarized in Figures 9 and 10.

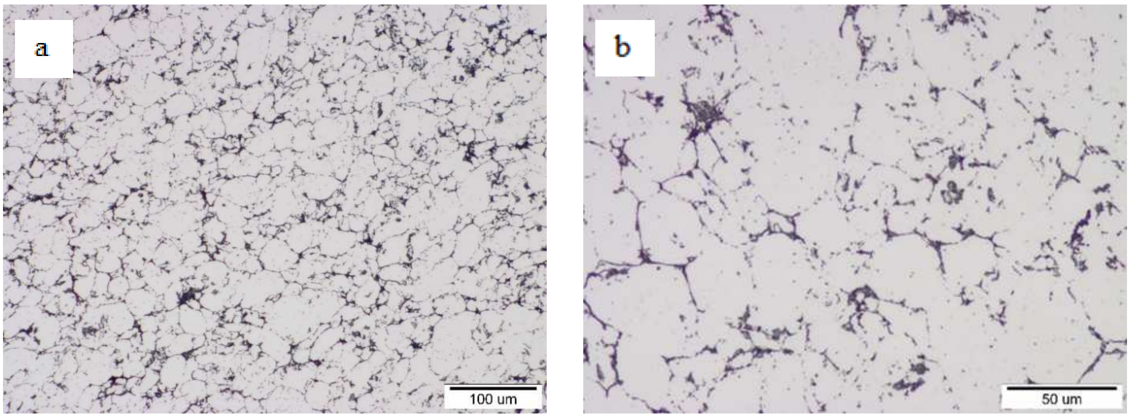


Fig. 7. Microstructure of as-sintered Al

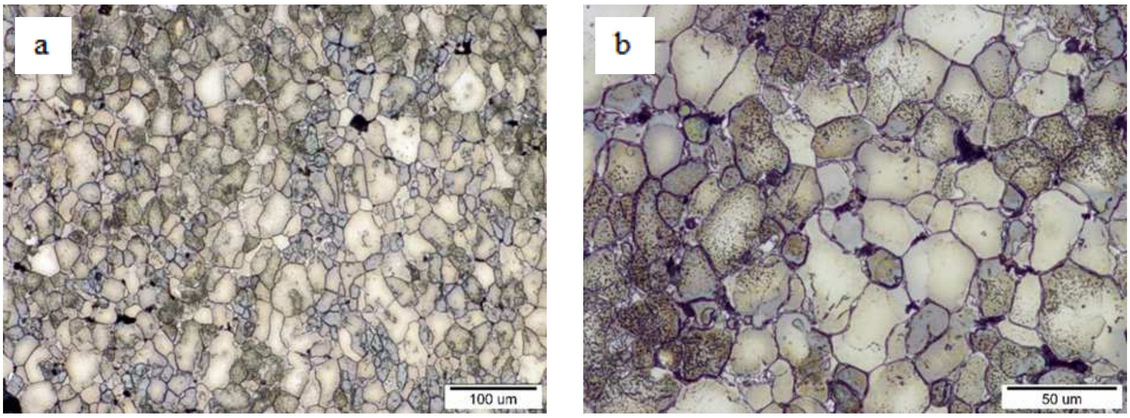


Fig. 8. Microstructure of as-sintered Al_4Cu

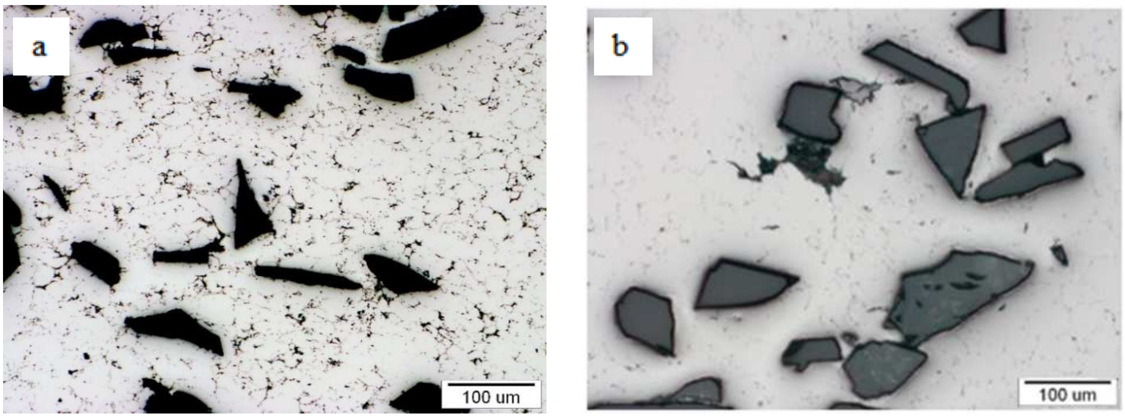


Fig. 9. Microstructure of Al + 10 wt.% SiC composite

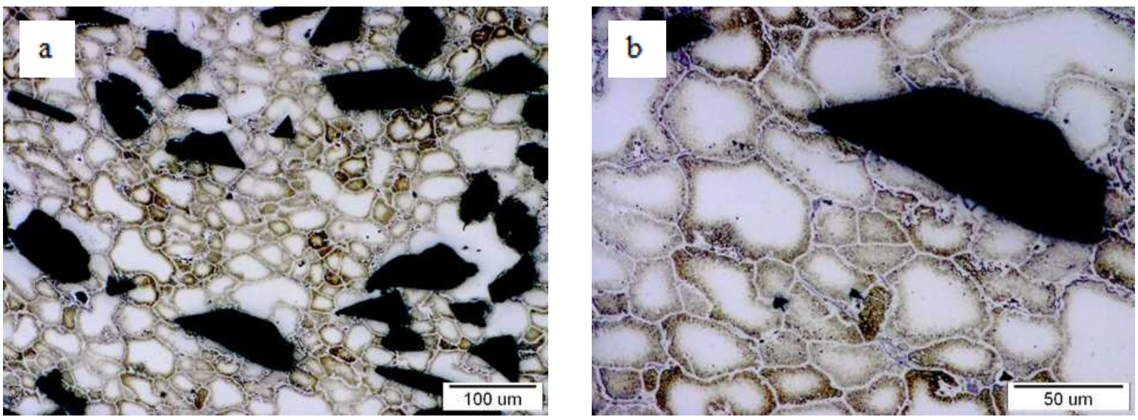
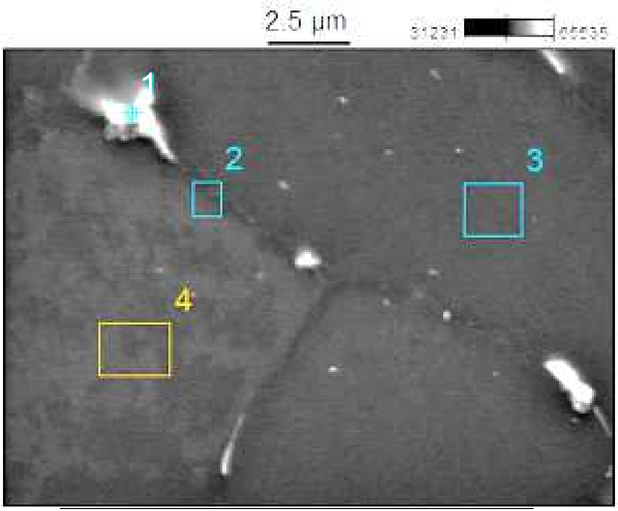


Fig. 10. Microstructure of Al_4Cu + 10 wt.% SiC composite

One of the main factors determining the homogeneous distribution of the reinforcing phase in the composite matrix is the ratio of the matrix particle size to the reinforcement particle size. The small difference between the particle size of the matrix and the reinforcement enables more uniform distribution of the reinforcing phase in the matrix. The use of reinforcing phase particles similar in size to the aluminum powder (63 μm) prevents the formation of agglomerates already at the storage stage of the SiC powder. Such agglomerates are difficult to break up using a mixing process without external forces (e.g. with balls). The microstructure of the Al_4Cu matrix (Fig. 8) consists of regular aluminum grains with copper dissolved in them and precipitates formed during cooling after the sintering process with stoichiometry corresponding to the Al_2Cu phase. According to the Al-Cu phase equilibrium diagram, increasing the sintering temperature above the eutectic transformation point (548ºC) favors the formation of a liquid phase in the composite, which fills the pores and penetrates the boundaries of the aluminum grains, consequently enabling better consolidation of the powders. In order to confirm the presence of the Al_2Cu phase, EDS studies were performed. The results are shown in the figure below.



Point.	Al, wt.%	Cu, wt.%	O, wt.%
1	68.29	30.83	0.88
2	96.28	3.60	0.56
3	95.05	4.47	0.48
4	94.96	4.56	0.48

Fig. 11. Point chemical composition analysis of Al_4Cu matrix sintered in nitrogen atmosphere; SEM

The presence of an Al_2Cu phase was also demonstrated by Ogel [11] and Mei [21], although the two publications used different fabrication methods. Paradoxically, classical powder metallurgy with sintering in nitrogen allows a relatively uniform distribution of these precipitates to be achieved in the easiest manner.

The results of the spot EDS analysis performed for the Al_4Cu matrix shown in Figure 11, revealed a high copper content in the precipitates located at the grain boundaries of the aluminum. The composition of the precipitates corresponds to that of the Al_2Cu phase (point 1), which is confirmed by the results of X-ray phase analysis [22]. The presence of precipitates with the given stoichiometric composition and copper dissolved in aluminum (points 2, 3, 4) confirms the diffusion of copper into aluminum occurring during the sintering process.

CONCLUSIONS

The application of the composite manufacturing technology consisting in double pressing and double sintering made it possible to obtain an increase in the density of the composites to a level exceeding 90% of the theoretical density in relation to composites obtained by single pressing and sintering, regardless of the employed atmosphere. Increasing the proportion of the hard reinforcing phase fraction results in a slight decrease in density. The microstructure of the composites is composed of aluminum grains in parallel, a reinforcing SiC phase located at the grain boundaries and small Al_2O_3 oxide precipitates, and additionally in the Al_4Cu matrix composites, also Al_2Cu phase precipitates. Few pores are observed locally against the aluminum matrix. The addition of copper to the aluminum matrix allows higher flexural strength values to be obtained, regardless of the utilized sintering atmosphere. The highest flexural strength values are obtained with a pure Al + 4 wt.% Cu matrix without the addition of an SiC strengthening phase, sintered in a nitrogen atmosphere (847 MPa). In summary, it can be concluded that the addition of elemental Cu powder to the aluminum matrix is very beneficial and that powder metallurgy technology makes it possible to produce an alloyed matrix.

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