

K. JACH*, K. PIETRZAK **, A. WAJLER*, A. SIDOROWICZ ***, U. BRYKAŁA*

APPLICATION OF CERAMIC PREFORMS TO THE MANUFACTURING OF CERAMIC – METAL COMPOSITES

ZASTOSOWANIE PREFORM CERAMICZNYCH DO WYTWARZANIA KOMPOZYTÓW CERAMIKA – METAL

This paper presents new solution to manufacturing Cu-Al₂O₃ composites with a dispersed reinforcement phase. It consists in substituting commonly used commercial alumina powder with crushed ceramic foam. Very low strength of thin-walled Al₂O₃ porous foams results in both their easy destruction and isolation of fragments <200 μm. The addition of the ceramic phase is 3-10 vol.%. The sintering process is performed for 30 min in a hot-press at 1050°C and 30 MPa. The aim of the presented paper is to study the effect of the volume content of the ceramic phase on the microstructure and selected physical and thermal properties of copper – alumina composites.

Keywords: ceramic preform, composite, copper, sintering

Artykuł przedstawia nowatorskie rozwiązanie wytwarzania kompozytów Cu-Al₂O₃ z rozproszoną fazą wzmacniającą. Polega ono na wykorzystaniu zamiast powszechnie stosowanego handlowego proszku tlenku glinu, rozkruszonej pianki ceramicznej. Wysoka kruchość cienkościennych, porowatych pianek Al₂O₃ pozwoliła na łatwe ich rozdrobnienie i wyodrębnienie frakcji o fragmentach wielkości < 200 μm. Dodatek fazy ceramicznej wynosił 3-10%obj. Proces spiekania prowadzono w temperaturze 1050°C w czasie 30 min i pod ciśnieniem 30 MPa. Celem pracy było określenie wpływu ilości dodatku fazy ceramicznej na mikrostrukturę oraz wybrane właściwości otrzymanych kompozytów.

1. Introduction

Alumina/copper composites are used when high thermal conductivity, high absorption and dissipation of heat, high resistance to thermal fatigue and good frictional wear resistance are required. Their properties depend on a number of factors, including the content, shape and distribution of the ceramic phase, the method of its preparation as well as the conditions under which they are fabricated. All these variables have strong influence on the above mentioned characteristics and, in consequence, on the future applications of the final material.

In this paper the preparation method of copper – alumina composites will be presented. Dispersion-strengthened Cu-Al₂O₃ composites are extensively used as materials for applications which require high-strength and good electrical conductivity[1]. Ceramic/metal composites can be produced by a number of methods, i.e. direct oxidation of most appropriate metal [2], metal infiltration (pressure or pressureless) of porous alumina foams [3-5], powder metallurgy [6-8], mechanical alloying [9-11], explosive compaction [12], in situ processing [13,14], spark plasma sintering [15], etc.

The most basic technique for producing copper-alumina composites includes mixing two powders in suitable proportions and sintering them. However, it has one considerable

drawback. Aluminium oxide added in powder form is agglomerated and does not sinter at the temperature at which the composite is prepared, which is determined by the melting point of copper. This article provides an efficient solution to this problem, which consists in using a fragment of the sintered ceramics derived from foams as the dispersed phase instead of alumina powder.

Studying the effect of using different amounts of ceramic phase additives on the microstructure and selected physical and thermal properties of copper – alumina composites is among the aims of the present investigation.

2. Experimental

The preparation procedure of ceramic foams was described in more detail in the previous work [16]. In short, ceramic foams were fabricated following the replication method. The ceramic powder, solvent, dispersant and binder were mixed in a Pulverisette planetary mill. The ceramic slurry thus prepared was deposited on polymeric sponge. After sintering, we obtained a ceramic material with the structure that was an ideal copy of the structure of the polymeric substrate (Fig. 1). In this work, the Al₂O₃ content in the slurry was 75.0 wt.%,

* INSTITUTE OF ELECTRONIC MATERIALS TECHNOLOGY, 133 WÓLCZYŃSKA STR., 01-919 WARSAW, POLAND

** INSTITUTE OF FUNDAMENTAL TECHNOLOGICAL RESEARCH, POLISH ACADEMY OF SCIENCE, 5B PAWIŃSKI STR., 02-106 WARSAW, POLAND

*** WARSAW UNIVERSITY OF TECHNOLOGY, FACULTY OF MATERIALS SCIENCE, 141 WOŁOSKA STR., 02-507 WARSAW, POLAND

whereas the addition of the acrylic-styrene dispersion was 5 wt.% of the mass of the ceramic powder.

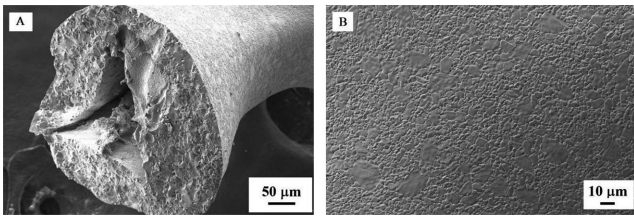


Fig. 1. The microstructure of sintered ceramic foam (SEM)

Very low strength of the structure allowed its easy destruction and isolation of a fraction with the size of < 200 μm. Next, the fabrication of composites began with the use of ceramics derived from the ceramic preform. The morphology and size distribution of the ceramic phase is shown in Figure 2. Ceramics obtained after a sieve analysis have different particle size fractions (2-400 μm), with the mean size of 46 μm.

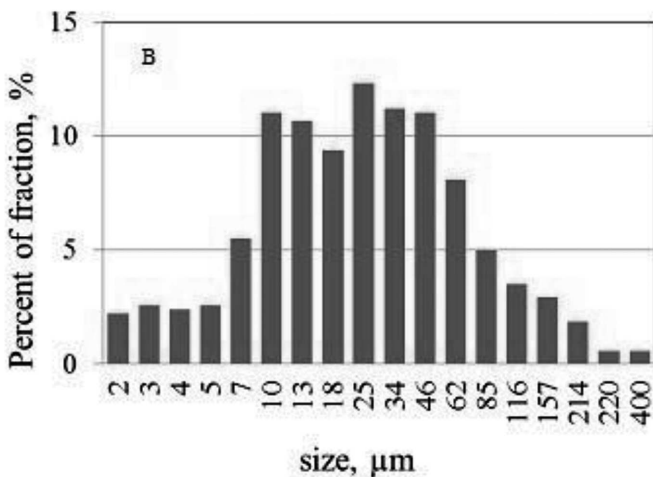
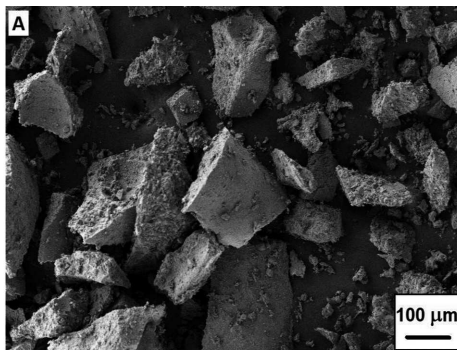


Fig. 2. Sintered and shattered Al₂O₃ preform; (A) SEM image, (B) distributions of the size

The first stage consisted in preparing a mixture of metal (copper powder <100 μm, 99.9% purity) and ceramics. The addition of the ceramic phase was 3.0 vol.%, 5.0 vol.% and 10.0 vol.%. In order to obtain a mixture of the highest possible homogeneity, mixing was performed in a planetary mill (Pulverisette 6, Fritsch) with tungsten carbide balls (Ø 5 mm). High-energy mechanical milling experiments were performed

at the room temperature under air atmosphere, at the rotation speed of 200 rpm and at the time of mixing 6h. The weight ratio of balls to powder (BPR) was 3:1. The sintering process of composites ran for 30 min in a hot-press (ASTRO, Thermal Technology, USA) at 1050°C and 30 MPa in an argon atmosphere.

3. Methods

The morphology and microstructure of the composite materials were examined using a scanning electron microscope (SEM AURIGA CrossBeam Workstation Zeiss). The characterization of the size distribution of the ceramic fraction was performed employing a CLEMEX television image analysis system. The relative density and open porosity were measured according to the Archimedes' principle. The hardness (HV5) was tested by Durascan 10/Emcotest with a Vickers diamond indenter using load 9.81 N with the loading time of 10 s. Thermal diffusivity was measured within the temperature range 50-600°C using the Laser Flash Analyser LFA457/Netzsch. Tribological tests were performed by using a DUCOM tribotester, in a "ball – flat surface" friction mode. The samples were pressed against a stainless steel ball with 6.5 mm in diameter with force ca. 10 N. After the test, the surface of the groove was analysed using scanning electron microscopy. The friction force thus generated was measured with a piezoelectric displacement sensor 24 times per second. It was induced and recorded during 30.0 min long friction processes.

4. Results and discussion

The microstructure of composite materials is shown in Fig. 3. As can be seen, the resulting composites are characterized by a homogeneous distribution of the reinforcing phase (Fig. 3A-C); however, the difference in size is significant. As a result of the conducted analysis, a strong binding was reported between the copper matrix of the composite and its ceramic phase (Fig. 3F).

TABLE 1
The properties of Cu-Al₂O₃ composite materials

Addition of alumina (vol.%)	Relative density (%)	Hardness HV ₅ (MPa)	Thermal Conductivity at 50°C (W/m·K)
3	98.8±0.2	48.2±1.2	329.9±2.7
5	99.3±0.5	61.8±1.5	299.0±3.3
10	98.4±0.7	87.2±1.5	262.1±5.2

Selected properties (physical and thermal) of Cu-Al₂O₃ composites produced by hot pressing (HP) are presented in Table I. It is worth noting here that by mixing the two phases of composites, very high densities, close to the theoretical ones, were obtained. The composites contained both many fragments of the size of about 100 μm (Fig. 3D), as well as fragments smaller than 10 μm (Fig. 3E). The reinforcement ceramic phase came in a wide range of sizes. Despite this, a trend of increased hardness of materials was clearly observed when increasing the alumina content. The hardness of

the composite containing 3 vol.% Al_2O_3 was approx. 48 MPa, whereas that of the composite containing 10 vol.% of ceramic equaled ca. 87 MPa.

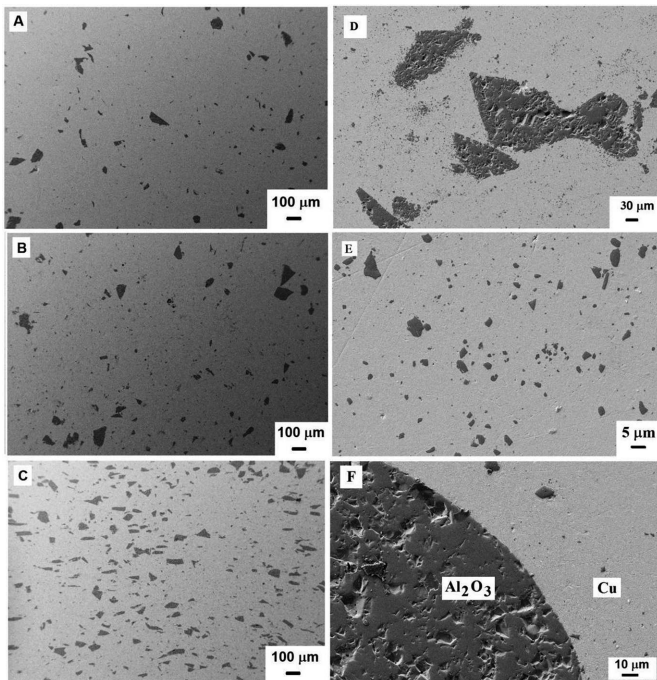


Fig. 3. Sample image (SEM) of the microstructure of copper composites with Al_2O_3 from foams; A) 3 vol.% Al_2O_3 , B) 5 vol.% Al_2O_3 , C) 10 vol.% Al_2O_3 , D) larger ceramic fragments in the composites, E) the smallest ceramic fragments in the composite, F) a phase boundary

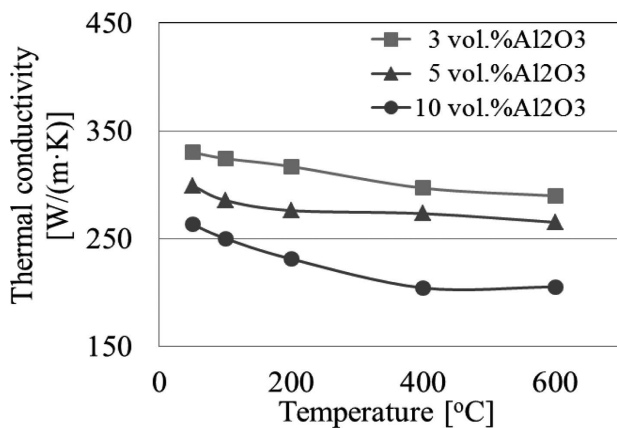


Fig. 4. The thermal conductivity of $\text{Cu-Al}_2\text{O}_3$ composite materials vs. the temperature

Thermal properties of the composites are shown in Fig. 4. The produced composites are characterized by high thermal conductivity ($330 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for $\text{Cu} - 3 \text{ vol.}\% \text{ Al}_2\text{O}_3$), which decreases with increasing both the content of the ceramic phase ($260 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for $\text{Cu} - 10 \text{ vol.}\% \text{ Al}_2\text{O}_3$), as well as the temperature. This behaviour is typical for this type of materials.

The abrasion resistance of the composites was also tested. Changes in the coefficient of friction diagrams presented in Figure 5 indicate that after several cycles of friction (equivalent to a test lasting approximately 20-50 s) the friction coefficient stabilizes at 1.3-1.4. This value is much higher than in

the case of composites prepared analogously with the use of commercial alumina powder as the ceramic phase, for which the friction coefficient of 0.3-0.4 was obtained. At the same time, regardless of the content of the ceramic phase in the composite, the profiles of traces are very similar in size and shape, as shown in the images presented in Fig. 6.

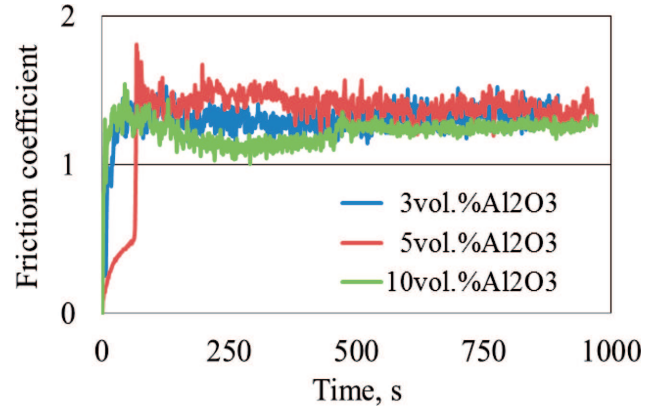


Fig. 5. The friction coefficient of copper-alumina composites

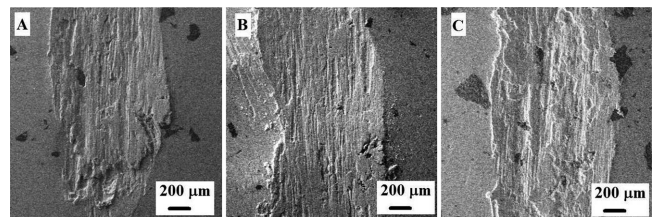


Fig. 6. SEM images of friction traces of $\text{Cu-Al}_2\text{O}_3$ composites after 15 min: a) 3 vol.% Al_2O_3 , b) 5 vol.% Al_2O_3 , c) 10 vol.% Al_2O_3

In general, there are no significant differences between the tribological properties of materials with the Al_2O_3 content ranging from 3 to 10 vol.%. Probably, the greatest discrepancy can be observed when the ceramic phase content in composites is even higher. We will focus on such composites in the next phase of our investigation.

5. Conclusions

Alumina derived from crushing of foams used as the ceramic phase allowed obtaining composite materials with advantageous properties. As shown, foams used in this study were homogeneous and their microstructure was free of voids. That is how porosity was eliminated in the composite in the area of the reinforcing phase, which is unattainable during the manufacture of composites using alumina powder which undergoes agglomeration. Additionally, using low pressure (30 MPa) when producing the composite resulted in obtaining the relative density of approximately 98-99%. This translated into other measured values. Materials with high thermal conductivity of $330 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for $\text{Cu} - 3 \text{ vol.}\% \text{ Al}_2\text{O}_3$ were obtained. The higher the content of the ceramic phase, the greater the hardness of the composites (for $\text{Cu} - 10 \text{ vol.}\% \text{ Al}_2\text{O}_3$ is close to 90 MPa). The resulting composites have high resistance to abrasion. Regardless of the type of the composites, the friction coefficient is in the range 1.3-1.4.

Acknowledgements

The work is partially financed by the Polish Science Funds in the years 2010-2013 as a research project no. N N507 4312 39.

The results presented in this paper have been partially obtained within the project "KomCerMet" (contract no. POIG.01.03.01-14-013/08-00 with the Polish Ministry of Science and Higher Education) within the framework of the Operational Programme Innovative Economy 2007-2013.

REFERENCES

- [1] M.V. Rajkovic, D. Bozic, M. Popovic, M.T. Jovanovic, *Science of Sintering* **41**, 185-192 (2009).
- [2] V. Rajkovic, D. Bozic, M.T. Jovanovic, *Materials and Design* **31**, 1962-1970 (2010).
- [3] K. Pietrzak, K. Jach, D. Kaliński, M. Chmielewski, J. Morgiel, *Proceedings of the Euro PM2011 Congress and Exhibition* **2**, 1-6 (2011).
- [4] N.A. Travitzky, *Materials Letters* **36**, 114-117 (1998).
- [5] K. Pietrzak, K. Jach, D. Kaliński, M. Chmielewski, *Proceedings of PM 2010 World Congress and Exhibition* **2**, 361-367 (2010).
- [6] L. Olmos, Ch.L. Martin, D. Bouvard, *Powder Technology* **190**, 134-140 (2009).
- [7] A. Lara-Guevara, I. Rojas-Rodriguez, R. Velazquez-Hernandez, R.A. Bernal-Correa, A. Sierra-Gutierrez, A. Herrera-Ramos, M.E. Rodriguez-Garcia, *Materials and Manufacturing Processes* **28**(2), 157-162 (2013).
- [8] Y.Z. Wan, Y.L. Wang, G.X. Cheng, M.M. Tao, Y. Cao, *Powder Metallurgy* **41**, 59-63 (1998).
- [9] M.F. Zawrah, H.A. Zayed, R.A. Essawy, A.H. Nassar, M.A. Taha, *Materials and Design* **46**, 485-490 (2013).
- [10] Zuhailawati Hussain, Leong Chee Kit, *Materials and Design* **29**, 1311-1315 (2008).
- [11] D.W. Lee, G.H. Ha, B.K. Kim, *Scripta Materialia* **44**, 2137-2140 (2001).
- [12] Zhao Zheng, Li Xiao-jie, Tao Gang, *Journal of Alloys and Compounds* **478**, 237-239 (2009).
- [13] F. Shehata, A. Fathy, M. Abdelhameed, S.F. Moustafa, *Materials and Design* **30**, 2756-2762 (2009).
- [14] M. Korać, Ž. Kamberović, Z. Andjić, M. Filipović, M. Tasić, *Science of Sintering* **42**, 81-90 (2010).
- [15] K. Dash, B.C. Ray, D. Chandra, *Journal of Alloys and Compounds* **516**, 78-84 (2012).
- [16] K. Jach, K. Pietrzak, D. Kalinski, M. Chmielewski, *Advances in Science and Technology* **65**, 164-169 (2010).