

Monika Gwoździk*, Zbigniew Bałaga

Czestochowa University of Technology, Institute of Material Engineering, al. Armii Krajowej 19, 42-200 Czestochowa, Poland

**Corresponding author. E-mail: gwozdzik.monika@wip.pcz.pl*

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MICROSTRUCTURE OF SINTERED Cu/SiC COMPOSITES

The paper presents the preliminary results of studies on obtaining copper-based composite materials strengthened with silicon carbides. Electrolytically obtained copper powders were used as the matrix. The sinters were characterized by different SiC contents (0, 5, 10, 15 wt.%). The materials were consolidated by one-sided pressing followed by sintering ($T = 800^{\circ}\text{C}$, $t = 1\text{ h}$). One-sided pressing was carried out at the pre-set pressing pressure of 60 kN and at the rate of 200 N/s. The research was performed on powders, mouldings and sinters. Investigations of the composites comprised: microscopic examinations (SEM), chemical composition analysis (SEM), XRD measurements, the density of the composites was determined, and a quantitative evaluation of the porosity of the composites was carried out.

Keywords: sinters Cu/SiC, porosity, SEM, XRD

MIKROSTRUKTURA SPIEKANYCH KOMPOZYTÓW Cu/SiC

Przedstawiono wyniki badań otrzymywania materiałów kompozytowych na podstawie miedzi umacnianych węglikiem krzemu. Osnowę kompozytów stanowiły proszki miedzi otrzymane metodą elektrolityczną. Spieki charakteryzowały się różną zawartością SiC (0, 5, 10, 15% wag. SiC). Konsolidacja materiałów następowała poprzez jednostronne prasowanie i następujące po nim spiekanie ($T = 800^{\circ}\text{C}$, $t = 1\text{ h}$). Prasowanie odbywało się przy założonym nacisku prasowania 60 kN i szybkości 200 N/s. Badania przeprowadzone zostały na proszkach, wypraskach oraz spiekach. Badania obejmowały: badania mikroskopowe (SEM), analizę składu chemicznego (SEM), pomiary rentgenowskie (XRD), określenie gęstości kompozytów, wykonanie ilościowej analizy porowatości w materiałach kompozytowych.

Słowa kluczowe: laminaty spieki Cu/SiC, porowatość, SEM, XRD

INTRODUCTION

Composite materials are becoming increasingly more popular among many centers in the country and in the world [1-8]. Ever more research is concerned with powder materials [9, 10], mainly used for sintered materials [11-13].

Powder metallurgy is a well-established field of technology, which includes methods for producing sinters. The process starts with homogeneous mixing of the reinforcement in the powder matrix. After mixing, the samples are compacted, which are then sintered under fixed conditions. Forming products from powders with a specific shape can take place as a result of powder paste pressing or extrusion, powder slurry casting, and powder rolling.

Copper and its alloys are used in many industrial and marine applications due to their electrical properties and good heat transfer. Copper is, however, susceptible to many forms of corrosion, including localised corrosion, such as crevice corrosion [12]. Cu based metal matrix composites are used in many electrical contacts, thermal and electronic packaging applications as they

possess high thermal and electrical conductivity as well as good corrosion resistance [13].

Copper-based composites are of particular interest to many scientific centers around the world [12-22]. Copper-based composites strengthened with metallic precipitates such as Fe and Cr or ceramic particles like SiC and Al_2O_3 have received a great deal of attention during recent years [19]. Metal matrix composites provide a novel way of strengthening materials. Metal matrix composites reinforced with ceramics impart both metallic properties like high toughness and ductility as well as ceramic properties [13].

In paper [20], micro-sized Cu and Cu-SiC composite powders were consolidated by powder metallurgy (PM), followed by sintering or high-pressure torsion (HPT) to study the effect of the different processing methods on the microstructure evolution and mechanical properties. Cu-Fe and Cu-Fe-SiC nanocomposite powders were synthesized by a two step mechanical alloying process in paper [19]. A supersaturated solid-solution of Cu-20 wt.% Fe was prepared by ball milling

of elemental powders up to 5 and 20 h and subsequently the SiC powder was added and then the mixture was milled for an additional 5 h [19].

MATERIAL AND METHODS

The first stage consisted in preparing an appropriate amount of copper powders and silicon carbide powders to produce compacts of varying mass compositions (0, 5, 10, 15 wt.% SiC content). The granularity of the copper and SiC powders were $10\div 30\text{ }\mu\text{m}$ and $20\div 50\text{ }\mu\text{m}$, respectively. The mixture of copper powder components and SiC powder was compacted. One-sided pressing without protective atmosphere was used (Fig. 1), at the pre-set pressing pressure of 60 kN and at the rate of 200 Ns^{-1} . The specimens were pressed on a Zwick Roell Z100 testing machine. The next stage comprised compact sintering at 800°C for 1 hour.

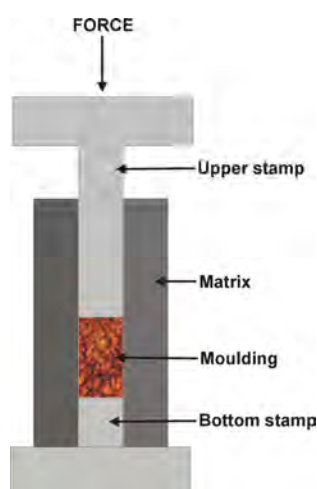


Fig. 1. Diagram of single-sided molding

Rys. 1. Schemat prasowania jednostronnego

Thorough examinations of the composites comprised:

- microscopic examinations performed using a Jeol JSM-6610LV scanning electron microscope (SEM),
- chemical composition analysis using a Jeol JSM-6610LV scanning electron microscope (SEM) working with an Oxford EDS electron microprobe X-ray analyzer, quantitative evaluation of the porosity of the composites. The microstructure of the composites was analyzed by image analysis software (Image Pro Plus). Then the average pore size and their volume share were calculated.

RESULTS AND DISCUSSION

The particles of the copper powder used for the research had a dendritic shape, obtained by the electrolytic method (Fig. 2a). The SiC particles were characterized by a polyhedral shape of a much larger dimensions than the copper particles (Fig. 2b).

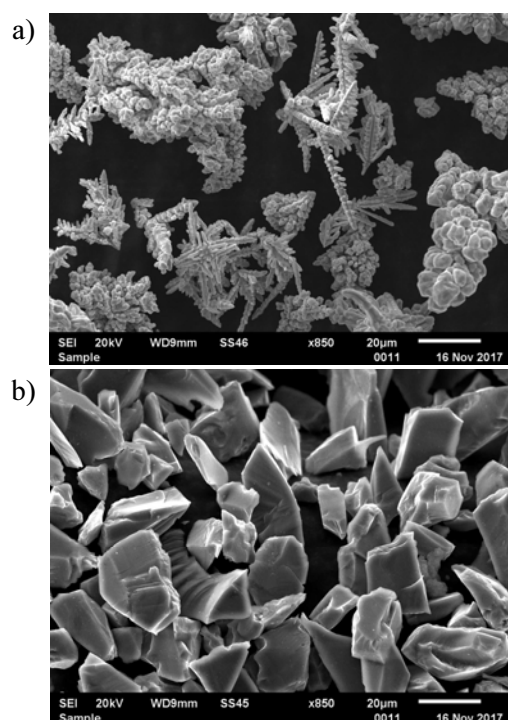


Fig. 2. Powder particle shapes (SEM): a) copper, b) silicon carbide

Rys. 2. Kształty proszków (SEM): a) miedzi, b) węgliku krzemu

Lower porosity was observed in the upper parts of the moulding (Figs. 3-6), i.e. in the layers adjacent to the upper punch during pressing, through which the pressure on the material is transferred. The greater the distance from the upper punch, the greater porosity of the material was observed, which is a consequence of the pressure drop.

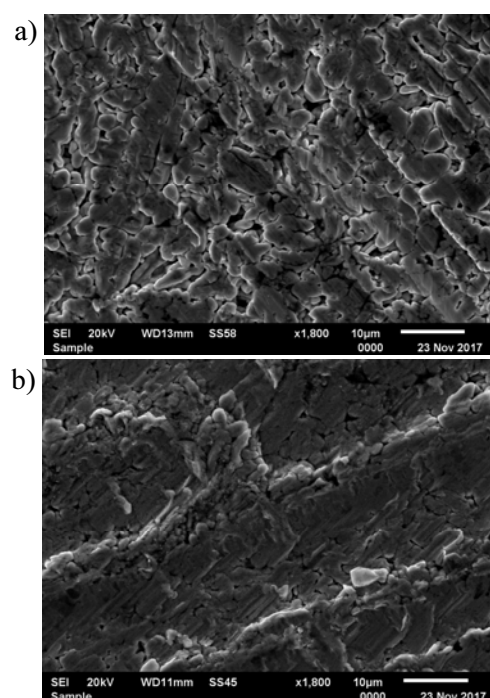


Fig. 3. Copper mouldings: a) from bottom stamp side, b) from upper stamp side

Rys. 3. Wypraski miedzi: a) od strony stempla dolnego, b) od strony stempla górnego

In the case of copper doped with 5% SiC, the lowest porosity was observed. In this case, microcracks in the silicon carbide are visible. The addition of SiC (15%) caused significant porosity, which is particularly visible

between the SiC particles, where gaps are formed in places.

The element distribution maps of the mouldings (Fig. 7) show uneven distribution of the SiC additive.

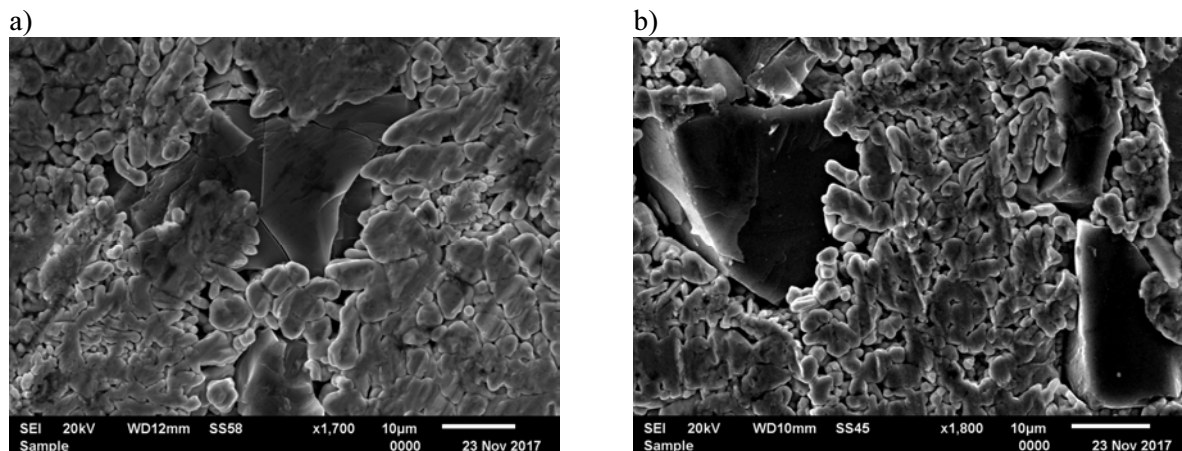


Fig. 4. Composite mouldings: 95% Cu, 5% SiC: a) from bottom stamp side, b) from upper stamp side

Rys. 4. Wypraski kompozytów: 95% Cu, 5% SiC: a) od strony stempla dolnego, b) od strony stempla górnego

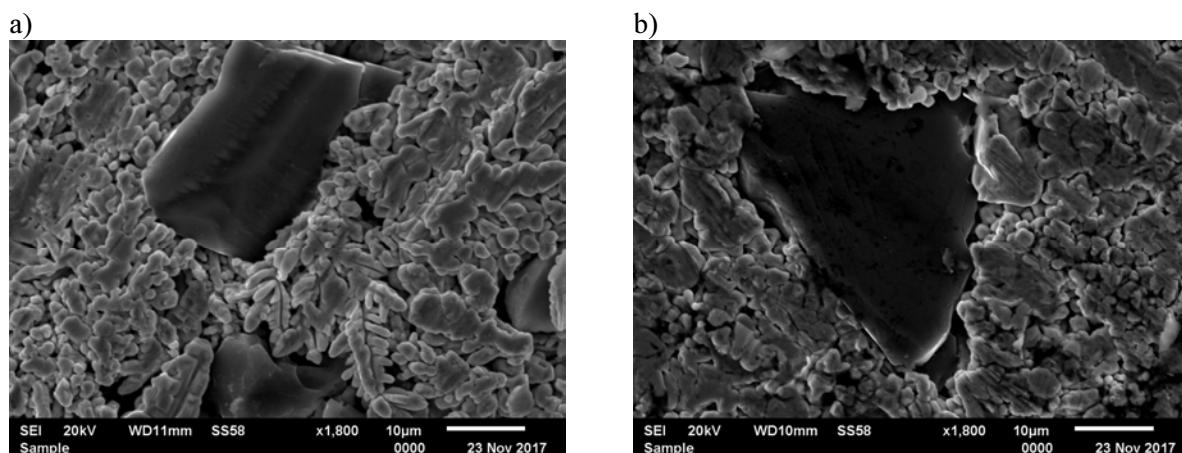


Fig. 5. Composite mouldings : 90% Cu, 10% SiC: a) from bottom stamp side, b) from upper stamp side

Rys. 5. Wypraski kompozytów: 90% Cu, 10% SiC: a) od strony stempla dolnego, b) od strony stempla górnego

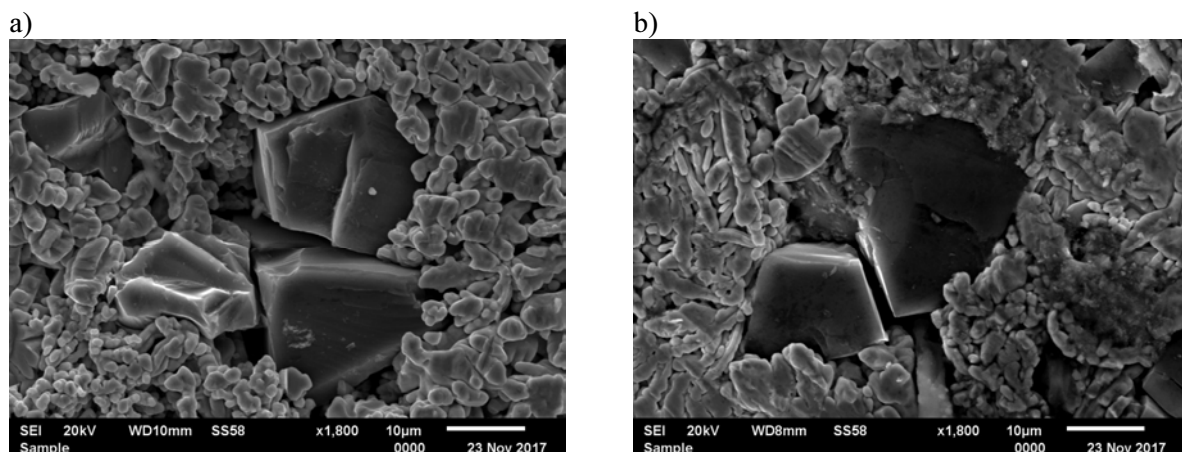


Fig. 6. Composite mouldings : 85% Cu, 15% SiC: a) from bottom stamp side, b) from upper stamp side

Rys. 6. Wypraski kompozytów: 85% Cu, 15% SiC: a) od strony stempla dolnego, b) od strony stempla górnego

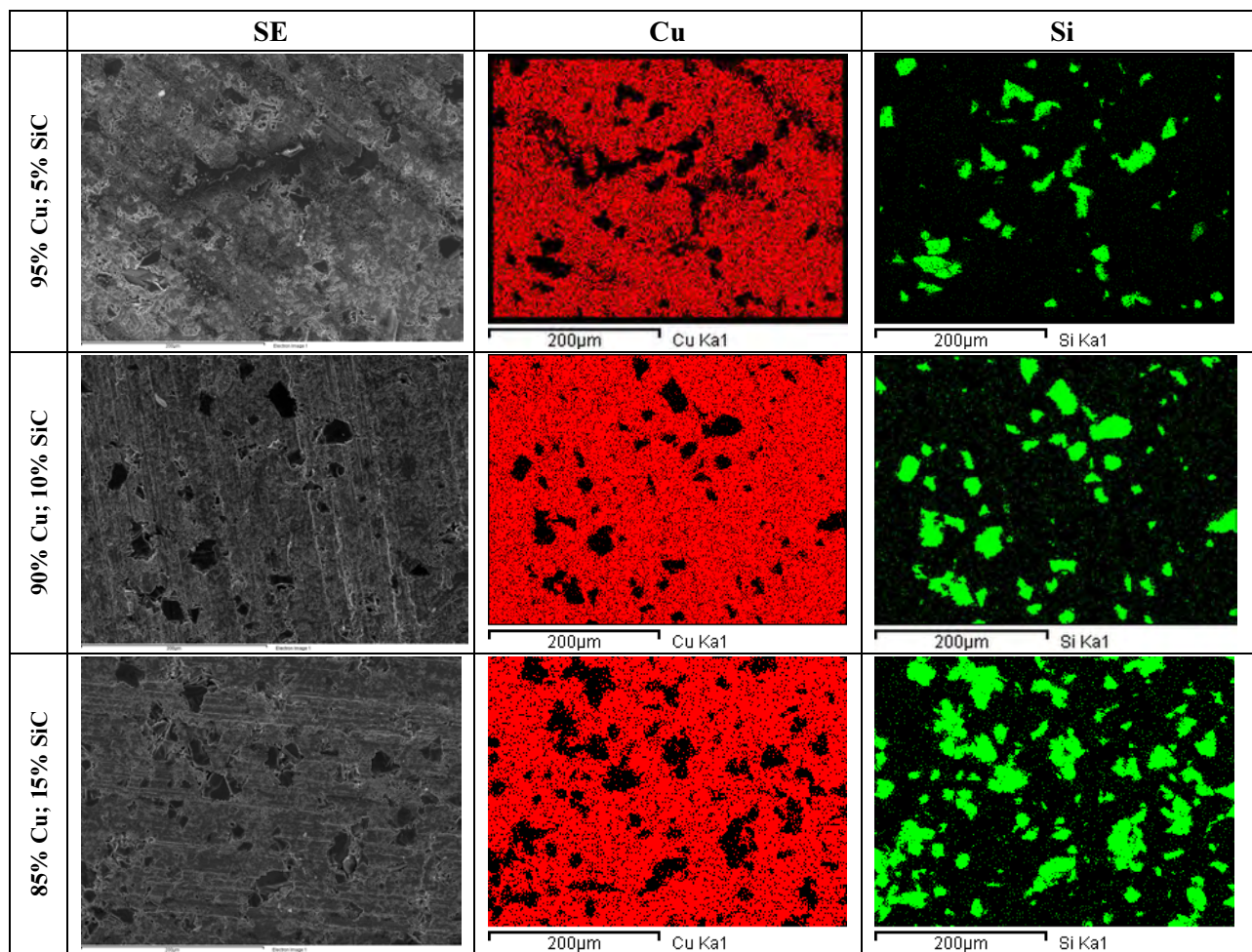


Fig. 7. Element distribution maps (SEM), composite mouldings

Rys. 7. Mapy rozkładu pierwiastków (SEM), wypraski kompozytów

As a result of the sintering process, the pore volume in the material decreased. Large pores disappeared to form a larger quantity but with a smaller volume, which is especially visible in the case of the copper powder (Fig. 8). A smaller amount of porosity was observed from the upper stamp on the Cu-SiC sintered samples (Figs. 9-11), this situation takes place within the copper

matrix. However, in the area of silicon carbide, the reverse effect is visible.

Directly from the side of the upper stamp, the porosity is larger. There are quite large pores entering the fissures at the interface between the copper matrix and SiC particles. This is most evident in the case of the addition of silicon carbide in the amount of 15%.

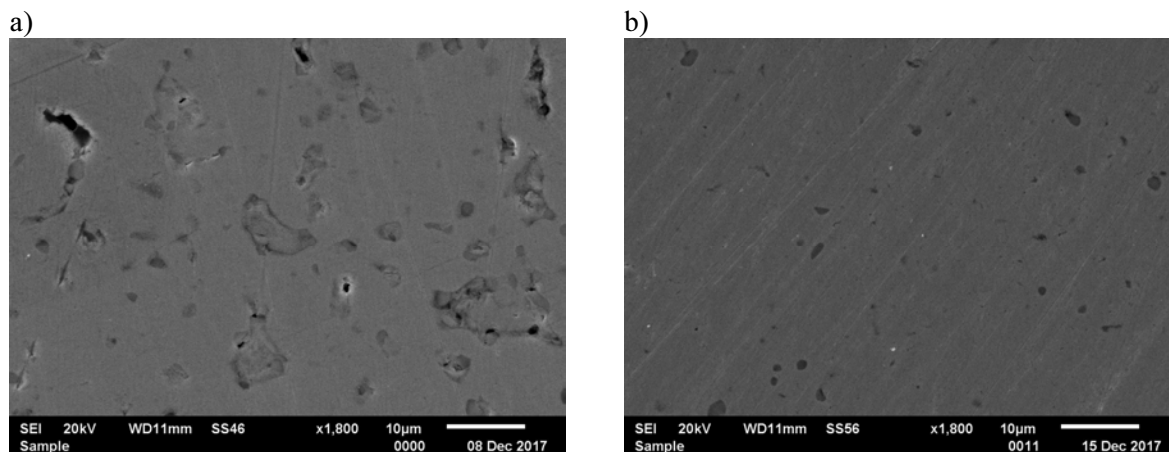


Fig. 8. Copper sinters: a) from bottom stamp side, after polishing, b) from upper stamp side, after polishing

Rys. 8. Spiek miedzi: a) od strony stempla dolnego, po polerowaniu, b) od strony stempla górnego, po polerowaniu

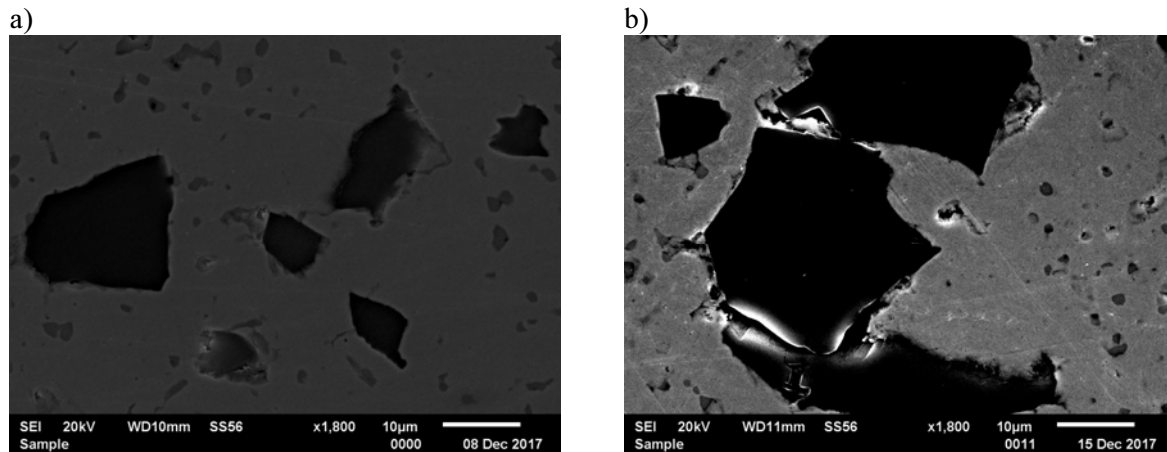


Fig. 9. Composite sinters: 95% Cu, 5% SiC: a) from bottom stamp side, after polishing, b) from upper stamp side, after polishing

Rys. 9. Spieki kompozytów: 95% Cu, 5% SiC: a) od strony stempla dolnego, po polerowaniu, b) od strony stempla górnego, po polerowaniu

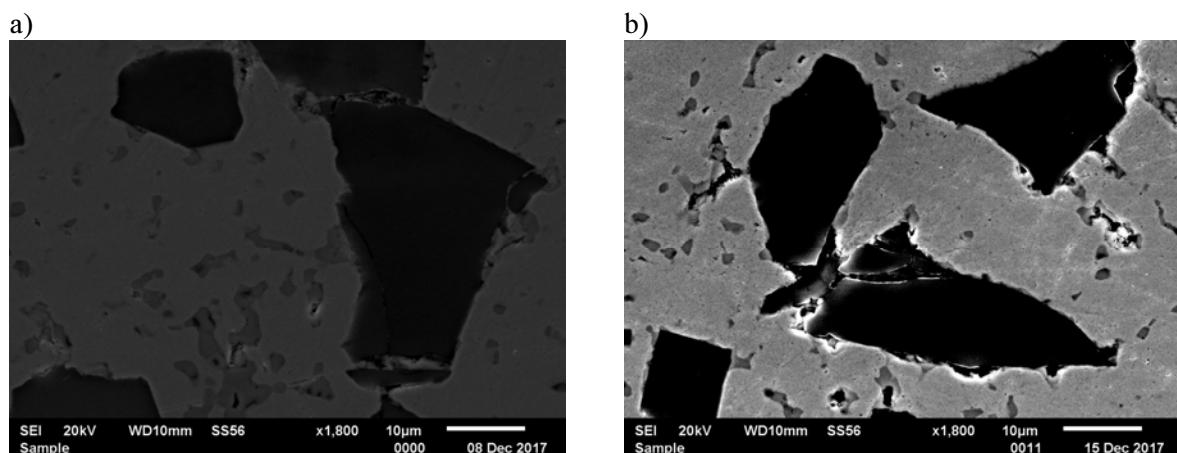


Fig. 10. Composite sinters: 90% Cu, 10% SiC: a) from bottom stamp side, after polishing, b) from upper stamp side, after polishing

Rys. 10. Spieki kompozytów: 90% Cu, 10% SiC: a) od strony stempla dolnego, po polerowaniu, b) od strony stempla górnego, po polerowaniu

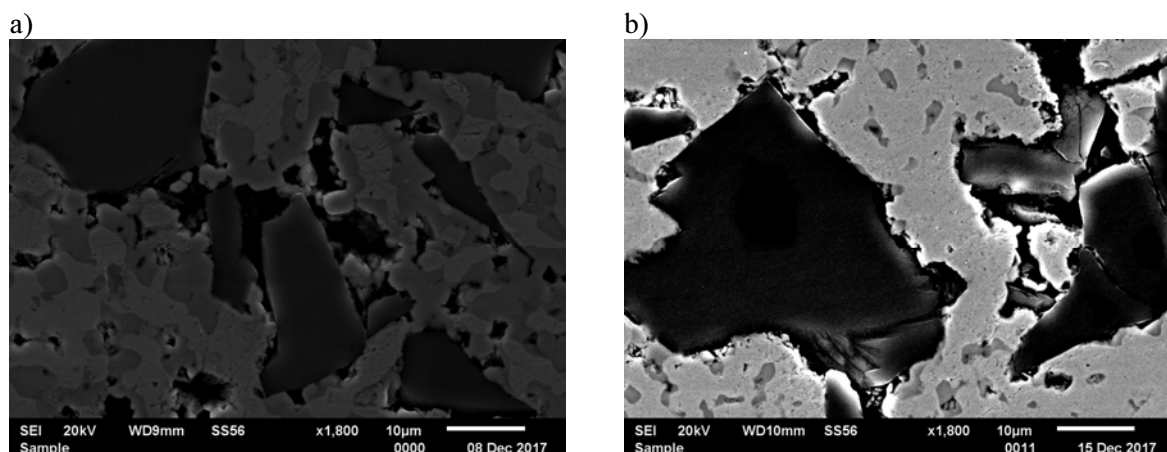


Fig. 11. Composite sinters: 85% Cu, 15% SiC: a) from bottom stamp side, after polishing, b) from upper stamp side, after polishing

Rys. 11. Spieki kompozytów: 85% Cu, 15% SiC: a) od strony stempla dolnego, po polerowaniu, b) od strony stempla górnego, po polerowaniu

The obtained porosity results (Figures 12-14) showed that in the case of pure copper from the bottom side (bottom stamp) the porosity is higher. As the addition of SiC increases, the proportion of porosity in the tested sinters increases. It was observed

that despite the lower porosity from the upper side (upper stamp) for the copper, the proportion of porosity for the whole sinter (Cu + SiC) is significantly higher owing to the addition of increasingly more SiC.

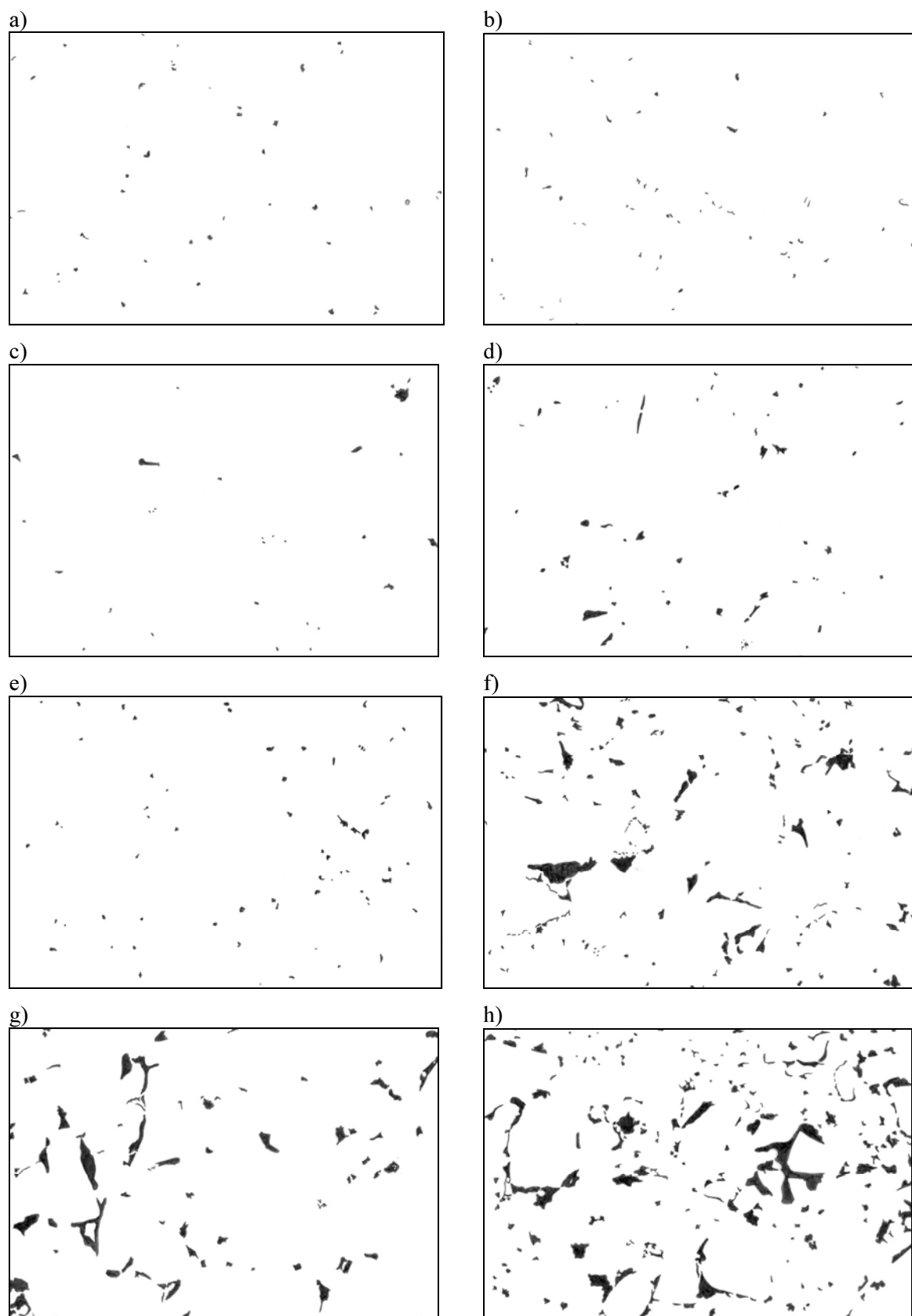


Fig. 12. Composite sinters after binarization: a) 100% Cu, from bottom stamp side, b) 100% Cu, from upper stamp side, c) 95% Cu, 5% SiC, from bottom stamp side, d) 95% Cu, 5% SiC, from upper stamp side, e) 90% Cu, 10% SiC, from bottom stamp side, f) 90% Cu, 10% SiC, from upper stamp side, g) 85% Cu, 15% SiC, from bottom stamp side, h) 85% Cu, 15% SiC, from upper stamp side

Rys. 12. Spieki kompozytów po binaryzacji: a) 100% Cu, od strony stempla dolnego, b) 100% Cu, od strony stempla górnego, c) 95% Cu, 5% SiC, od strony stempla dolnego, d) 95% Cu, 5% SiC, od strony stempla górnego, e) 90% Cu, 10% SiC, od strony stempla dolnego, f) 90% Cu, 10% SiC, od strony stempla górnego, g) 85% Cu, 15% SiC, od strony stempla dolnego, h) 85% Cu, 15% SiC, od strony stempla górnego

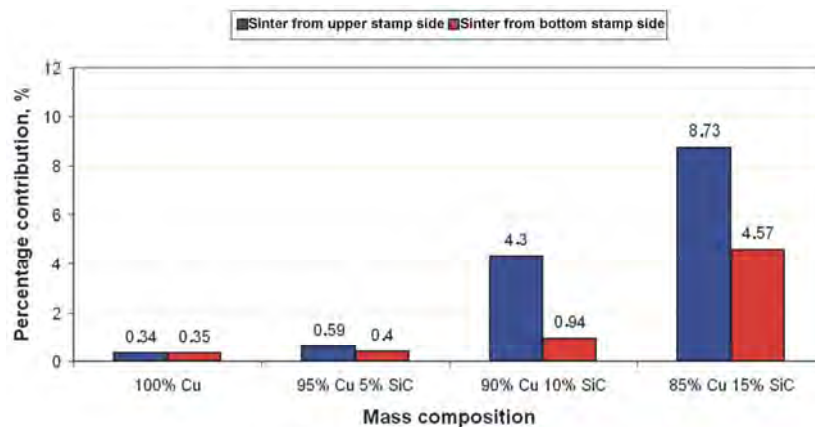


Fig. 13. Percentage of porosity in composites

Rys. 13. Udział procentowy porowatości w kompozytach

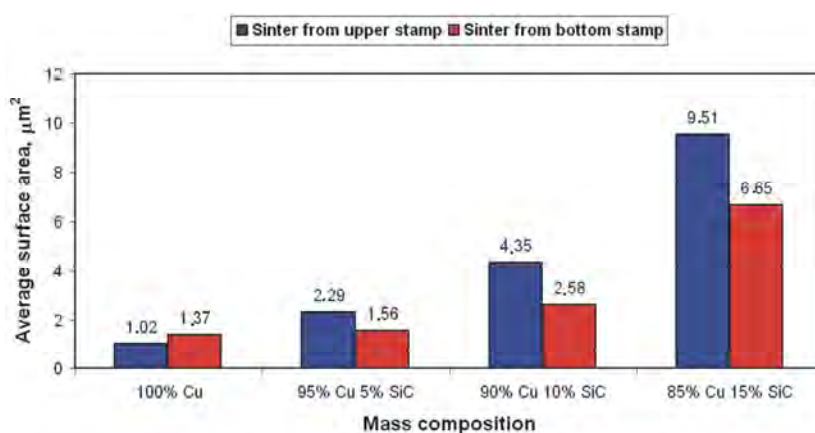


Fig. 14. Average surface area of pores in composites

Rys. 14. Średnie pole powierzchni pory w kompozytach

CONCLUSIONS

On the basis of the conducted research, the following statements and conclusions were formulated:

- the metallographic examination showed an uneven distribution of SiC powder in the copper powder,
- together with an increase in silicon carbide content at a pressing force of 60 kN, its damage is greater,
- larger cracks in the silicon carbide were observed from the upper stamp side,
- lower porosity was observed in the upper parts of the moulding for pure copper,
- the addition of SiC caused the reverse situation. Despite the lower porosity in the copper, the porosity in the entire sinter (Cu + SiC) is greater from the upper stamp side, caused by the higher pressure, which causes damage to the SiC particles.

REFERENCES

- [1] Rinaldi M., Puglia D., Dominici F., Cherubini V., Torre L., Nanni F., Melt processing and mechanical property characterization of high-performance poly(ether ether ketone)-carbon nanotube composite, *Polym. Int.* 2017, 66, 1731-1736.
- [2] Strobel H.A., Calamari E.L., Beliveau A., Jain A., Rolle M.W., Fabrication and characterization of electrospun polycaprolactone and gelatin composite cuffs for tissue engineered blood vessels, *J. Biomed. Mater. Res. Part B* 2018;106B:817-826.
- [3] Shuo Zhao, Yiwei Zhang, Yuming Zhou, Kaibo Qiu, Chao Zhang, Jiasheng Fang, Xiaoli Sheng, Reactable polyelectrolyte-assisted preparation of flower-like Ag/AgCl/ BiOCl composite with enhanced photocatalytic activity, *Journal of Photochemistry and Photobiology A: Chemistry* 2018, 350, 94-102.
- [4] Gwoździk M., Bałaga Z., Wróbel D., Nitkiewicz Z., Evaluation of wear degree of rotational instruments with diamond coat, *Composites Theory and Practice* 2017, 17(4), 216-220.
- [5] Zygoń P., Gwoździk M., Peszke J., Nitkiewicz Z., The effect of water acrylate dispersion on the properties of polymer-carbon nanotube composites, *Archives of Metallurgy and Materials* 2015, 60(4), 2716-2720.
- [6] Zygoń P., Gwoździk M., Peszke J., Nitkiewicz Z., Comparison of properties of polymer composite materials reinforced with carbon nanotubes, *Archives of Metallurgy and Materials* 2015, 60(1), 193-198.
- [7] Bałaga Z., Przybycin A., Wawrzyniak J., Gnatowski A., Examinations of the effect of montmorillonite on selected properties and structure of polybutylene terephthalate, *Composites Theory and Practice* 2017, 17(3), 175-179.
- [8] Xiangwen Ma, Peng Zhang, Yuanyuan Zhao, Ying Liu, Jian Li, Jin Yuan Zhou, Xiaojun Pan, Erqing Xie, Role of N doping on the electrochemical performances of ZnCo₂O₄ quantum dots/reduced graphene oxide composite nanosheets, *Chemical Engineering Journal* 2017, 327, 1000-1010.

- [9] Zygoń P., Peszke J., Gwoździk M., Nitkiewicz Z., Malik M., Characteristic of carbon nanotubes modified with cobalt, copper and bromine, *Archives of Metallurgy and Materials* 2014, 59(2), 675-679.
- [10] Szafarska M., Iwaszko J., Laser remelting treatment of plasma-sprayed Cr_2O_3 oxide coatings, *Archives of Metallurgy and Materials* 2012, 57(1), 215-221.
- [11] Dudek A., Microstructure and properties of the composites: hydroxyapatite with addition of zirconia phase, *Journal of Engineering Materials and Technology, Transactions of the ASME*, 2011, 133(2), 021006-1-021006-5.
- [12] Zygoń P., Gwoździk M., Nitkiewicz Z., Jagielska-Wiaderek K., Corrosion resistance of sinters Cu/CNT, *Solid State Phenomena* 2015, 227, 51-54.
- [13] Arabinda Meher, Debasis Chaira, Effect of graphite and SiC addition into Cu and SiC particle size effect on fabrication of Cu-graphite-SiC MMC by powder metallurgy, *Trans Indian Inst Met* 2017, 70(8), 2047-2057. DOI 10.1007/s12666-016-1026-1.
- [14] Gongjun Cui, Jian Ren, Zhangxiang Lu, The microstructure and wear characteristics of Cu-Fe matrix, Friction Material with Addition of SiC. *Tribol Lett* 2017, 65, 108. DOI 10.1007/s11249-017-0890-0.
- [15] Liu Meng, Bai Shuxin, Li Shun, Zhao Xun, Xiong Degan, Microstructure and thermal properties of SiCp/Cu composites with Mo coating on SiC particles, *Journal of Wuhan University of Technology-Mater. Sci.* 2017, 32, 5 103-1018. DOI 10.1007/s11595-017-1704-9.
- [16] Chmielewski M., Pietrzak K., Strojny-Nędza A., Jarząbek D., Nosewicz S., Investigations of interface properties in copper-silicon carbide composites, *Arch. Metall. Mater.* 2017, 62, 2B, 1315-1318, DOI: 10.1515/amm-2017-0200.
- [17] Prosviryakov A.S., SiC content effect on the properties of Cu-SiC composites produced by mechanical alloying, *Journal of Alloys and Compounds* 2015, 632, 707-710.
- [18] Jarząbek D.M., Milczarek M., Wojciechowski T., Dziekoński C., Chmielewski M., The effect of metal coatings on the interfacial bonding strength of ceramics to copper in sintered Cu-SiC composites, *Ceramics International* 2017, 43, 5283-5291.
- [19] Rabiee M., Mirzadeh H., Ataie A., Processing of Cu-Fe and Cu-Fe-SiC nanocomposites by mechanical alloying, *Advanced Powder Technology* 2017, 28, 1882-1887.
- [20] M. Ibrahim Abd El Aal, Effect of high-pressure torsion processing on the microstructure evolution and mechanical properties of consolidated micro size Cu and Cu-SiC powders, *Advanced Powder Technology* 2017, 28, 2135-2150.
- [21] Shabani M., Hossein Paydar M., Zamiri R., Goodarzi M., Mohsen Moshksar M., Microstructural and sliding wear behavior of SiC-particle reinforced copper matrix composites fabricated by sintering and sinter-forging processes, *J. Mater. Res Technol.* 2016, 5(1), 5-12.
- [22] Yanguang Zhou, Ming Hua, Mechanical behaviors of nanocrystalline Cu/SiC composites: An atomistic investigation. *Computational Materials Science* 2017, 129, 129-136.