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Structure of metal matrix composites with an addition of tuff

M. Łach
Materiale Engineering Institute, Cracow University of Technology, al. Jana Pawła II 37, 31-864 Kraków
Corresponding author. E-mail address: michallach85@o2.pl

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Abstract

The article presents preliminary results of tests of metal matrix composites structure which was modified by an addition of powdered volcanic tuff. Distribution and shape of ceramic particles as well as the quality of the bonding along the tuff- metal matrix interface were studied. Depth of tuff element diffusion in the matrix as well as diffusion in tuff particles were checked. Micro-hardness and porosity of the composites were also tested. The tuff from Filipowice near the town of Krzeszowice was used for the tests. Powder metallurgy was applied to obtain the composites and the matrix materials were copper and 316L steel powders. The tuff was introduced in 2, 5 and 10 % by weight. To remove water from the channels of aluminosilicates, the tuff was baked at 850 ^{o}C for 4 hours and then cooled together with the oven.

The tests revealed good quality of the bonding of the tuff particles and the matrix and their even distribution. The addition of tuff improved the hardness of the composites and reduced their porosity which has great significance because of possible applications of this kind of materials in general and copper composites in particular. This gives grounds for further studies on volcanic tuff use in metal composites

Keywords: powder metallurgy, composite materials, copper composites, ceramic particles, tuff

1. Introduction

Rapid development of metal matrix composites where ceramic particles are used as the reinforcement has contributed to a search for new materials, whose characteristic features would include great hardness, resistance to abrasion while maintaining high conductivity. These requirements are met by copper matrix composites since they contain phases, which are hard and resistant to abrasion. In recent years, various ceramic phases have been used for dispersion strengthening of copper. The most commonly used agents include oxides: Al2O3, SiO_2, ZrO_2, MgO, BeO, Cr_2O_3, carbides: SiC, TiC, Cr_7C_3, Cr_3C_2, boronides and nitrides: TiB_2, ZrB_2, CrB_2, BN. Copper composites were also produced containing the iron-aluminium or copper-aluminium-iron intermetallic phases or the like [1,2,3].

Due to the fact that copper composites with ceramic phase particles have an interesting combination of properties such as strength, plasticity, resistance to abrasion and good electrical properties, they have a wide range of applications.

They are used in: switches in low-voltage appliances, tubular heat converters, electrodes in electro-erosion machining, electrode endings in resistance welding structural elements in reactor technology [1,2,3].

The material which can be used as a ceramic addition to copper matrix composites or other metals is volcanic tuff. It has great hardness, high resistance to abrasion and a high melting point. Moreover, tuff has many features typical of zeolites, i.e. a number of very interesting physical and chemical properties, therefore tuff particles can constitute a very attractive reinforcing phase in metal matrix composites.

The aim of the study was to produce metal composites with tuff addition using powder metallurgy and to analyse their structure and hardness.

2. Materials for studies

Copper powder and water-sprayed AISI 316L austenitic stainless steel as well as ground volcanic tuff from Krzeszowice area were used in the studies.

Volcanic tuff is compact, porous sedimentary rock (figs.1) of clastic type and consisting of pyroclastic material often with an admixture of another clastic material cemented with siliceous or loamy binder. Its characteristic feature is great porosity and the resultant low specific gravity.

Fig. 1. Volcanic tuff in the form of rock

Volcanic tuff is a widespread material. In Poland it can be found in the areas where volcanoes were present: in the Sudetes or in the southern part of Wyżyna Krakowsko-Częstochowska (in. Filipowice). Basing on the lithological composition, tuff can be categorized as follows: andesitic, basaltic, rhyolitic, trachytic, radiacid and others. The tuff of Filipowice is a pink rock with light patches (unlike, e.g. basaltic tuff which has dark colour). The phase composition of the tuff is presented in table 1.

Table 1.
Phase composition of tuff [%] [4]

SiO_2	Fe_2O_3	Al_2O_3	MgO	CaO	K_2O
47,20-66,22	12,31-14,49	10,73-13,90	0,26 - 7,00	1,67 - 2,79	1,48 - 2,43
Na_2O	**TiO_2**	**MnO**	**SO_3**	**H_2O**	**P_2O_5**
0,59 - 4,87	0,21 - 1,34	0,03 - 2,43	0,09 - 0,17	5,22 - 0,43	0,03 - 2,43

Tuffs are a kind of carriers of zeolites which are aluminosilicate minerals with specific features and a very wide range of applications. In their structure there occur empty channel-like spaces whose size is several Ǻ. When heated, they give off their water content without affecting the crystal structure and, which is important, the process is reversible. They have ion exchange qualities. Zeolites have been used as molecular sieves, substances for treatment of gases and sewage (they perfectly absorb ions of heavy metals), raw material for cement production, as well as substances in food industry, petroleum industry and many others. Due to the properties of zeolites it can be assumed that they will also be an attractive strengthening material in metal composites.

Volcanic tuff in rock form was first fragmented and then ground in an ultracentrifugal RETSCH ZM1 mill for grinding ceramic materials. After milling, it was baked at 850 ^oC for 4h and then cooled with the oven. Following a sieve analysis, tuff particles measuring less than 40 μm were chosen for the studies. Tuff addition in the samples was 2, 5, 10% by weight.

Appropriate quantities of powdered tuff were mixed with metal powders and rolled samples 20x5 [mm] were made and designed for further tests. One-sided pressing was applied at a pressure of 600 MPa. Baking was carried out in a tubular sylite oven for copper at 900^{o}C and in nitrogen atmosphere and in hydrogen atmosphere at 1250^{o}C for 316L steel. Baking time in both cases was 60 minutes. After baking, the samples were cooled together with the oven.

3. Research method

Porosity evaluation was carried out using the method of weighing in the air and water according to the standard PN-EN ISO 2738:2001.

Structural and fractographic examination of sinters was performer using a Joel scanning electron microscope JSM-5510LV. Transverse microsections were prepared for studies. They were done in the plane going through the middle of the sample and parallel to the direction of the force applied during pressing. The structure examination was performed in non-pickled state. Micro hardness $\mu HV_{0,05}$ was measured using FM 700E microhardness tester.

4. Results

Table 2 demonstrates examples of microhardness values for composites with tuff. Tuff particles are characterized by great hardness. The wide range of tuff hardness values is due to diverse chemical composition of the particles. The tuff particles which are introduced are not homogenous with respect to their phase composition. The introduced tuff powder contains both complex aluminiumsilicates with different chemical composition and single particles of SiO_2.

In the case of copper matrix composite, there is an apparent increase in hardness near the tuff particles, which can result from diffusion of the elements such as Al or Si from the tuff into the matrix. The studies revealed the increase in hardness to even double in the areas near the tuff particles. The phenomenon was not observed to occur on such a scale in the case of 316L steel composites. The matrix hardness remained unchanged regardless of the distance from ceramic particles.

Table 2.
Example microhardness value ranges $\mu HV_{0,05}$ for composites with tuff

	Matrix	Tuff particles	Zones near tuff particles.
Copper-tuff	70-80	600-1250	120-160
316L+tuff	330-360	600-1250	330-370

Results of measurements of specific gravity of copper matrix sinters in relation to the quantity of tuff introduced have been presented in fig.2. An analysis of the results of the studies shows that addition of tuff has an effect on an increase in specific gravity of sinters as compared to copper sinters without tuff addition. A 2% addition of tuff increases sinter specific gravity from 87% to 94%. The highest specific gravity of some 97% was observed for copper sinter with a 5% tuff addition.

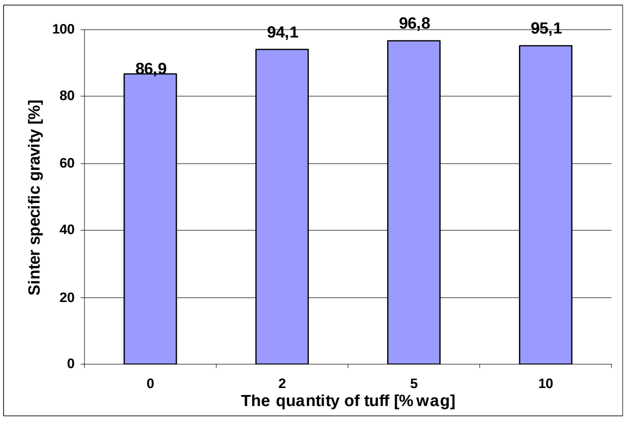


Fig. 2. Effect of tuff addition on sinter specific gravity

Examples of microstructures of the copper sinters tested have been shown in fig.3. Sinter microstructure with 2% (A) and 10% (B) tuff addition has been presented. Introduction of powdered tuff makes a significant change in the sinter structure. Tuff particles of varying size are clearly visible. The differences in particle size are due to the value range of the introduced tuff: < 40 µm.

Mixing powdered copper with powdered tuff resulted in a uniform distribution of particles in the matrix. It is a characteristic feature of the composite with a 10% tuff addition that the introduced particles combine in some areas to form longitudinal “chains”. Big clusters of particles were not observed. The introduced particles did not dissolve in the matrix during baking, which is due to a high thermal resistance of volcanic tuff.

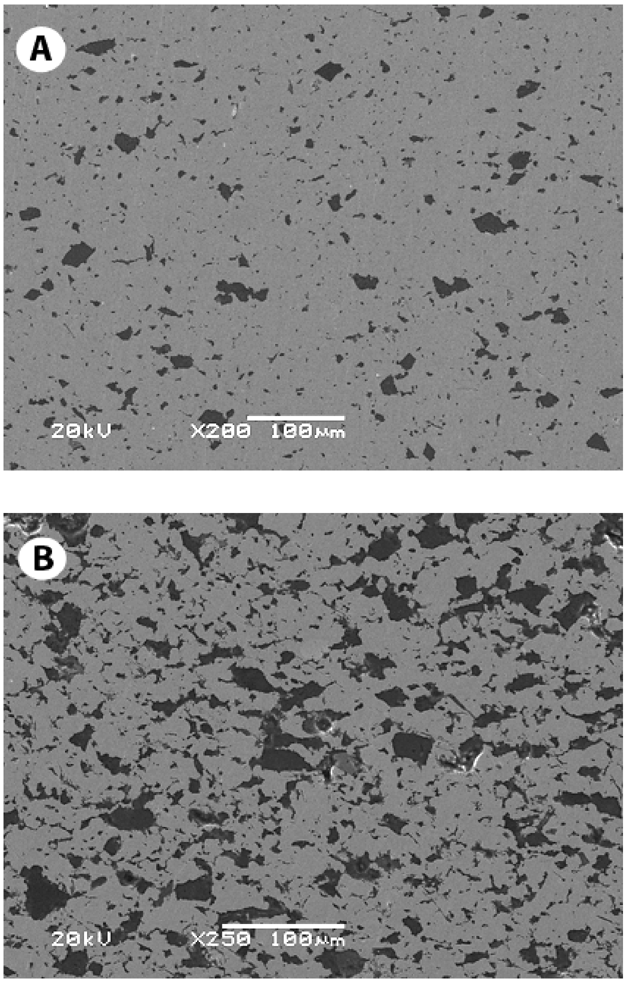
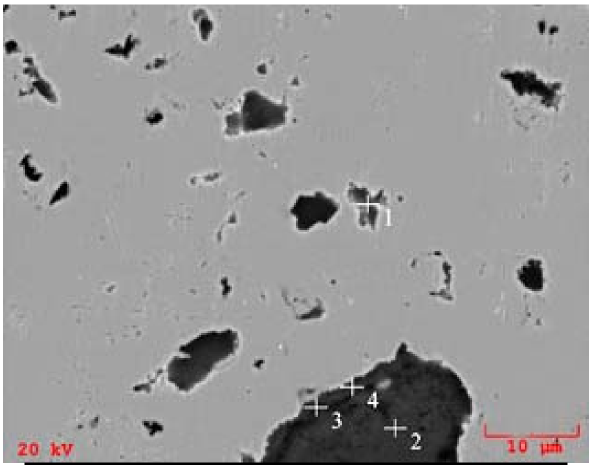


Fig. 3. Copper sinter microstructure with a) 2% tuff addition, b) 10% tuff addition. Non-pickled. SEM

Fig. 4 shows an example microstructure of copper composite with a 2% tuff addition and example percentage of Cu, Si and K in relation to the distance of the examined point from the centre of the particle. The points were marked in the picture with a cross and numbers from 2 to 4. The table shows percentage values of particular elements at these points.

The photograph shows a good bonding of tuff and copper particles. There are no discontinuities on the border of the tuff and matrix. Element percentage values at particular points may demonstrate that there occurs diffusion of copper into tuff and of tuff elements into the matrix. The copper content was observed to grow from the centre of the particle to the border line of tuff-matrix. Silicon and potassium content decreases towards the particle border which may be a proof of their diffusion into the matrix. That is why their concentration in the tuff particle decreased.



Point number	2	3	4
Cu [%]	2,16	12,46	14,46
Si [%]	41,46	34,92	27,31
K [%]	14,9	5,87	4,093

Fig. 4. Microstructure of the composite Cu-2% tuff and content of selected elements at particular points in tuff particle

Fig. 5 presents the microstructure of copper sinter with an addition of tuff which was not annealed but only milled and added in its natural form. The microstructure has been shown only to compare the quality of the tuff-matrix bonding with the above presented composites. In figures 3 and 4 there are no visible discontinuities or micro voids between the tuff particles and the matrix. In Fig. 5, on the other hand, discontinuities are clearly manifest in the form of dark areas around the tuff particles. No heat treatment of the tuff prior to composite production made the tuff crystalline water to be released only during compact baking which resulted in the tuff particles losing cohesion with the matrix.

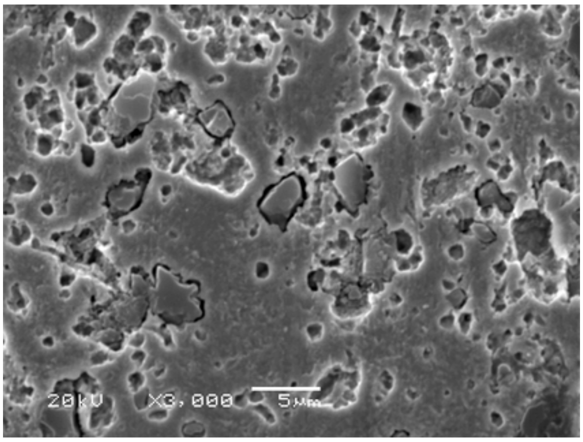


Fig. 5. Microstructure of copper composite with tuff addition, which was not annealed. SEM

Linear distribution of the elements at the matrix/tuff interface was also analysed. An example of the linear distribution of elements is presented in fig.6. The diagrams below demonstrate the distribution of particular elements along the white line marked in the figure.

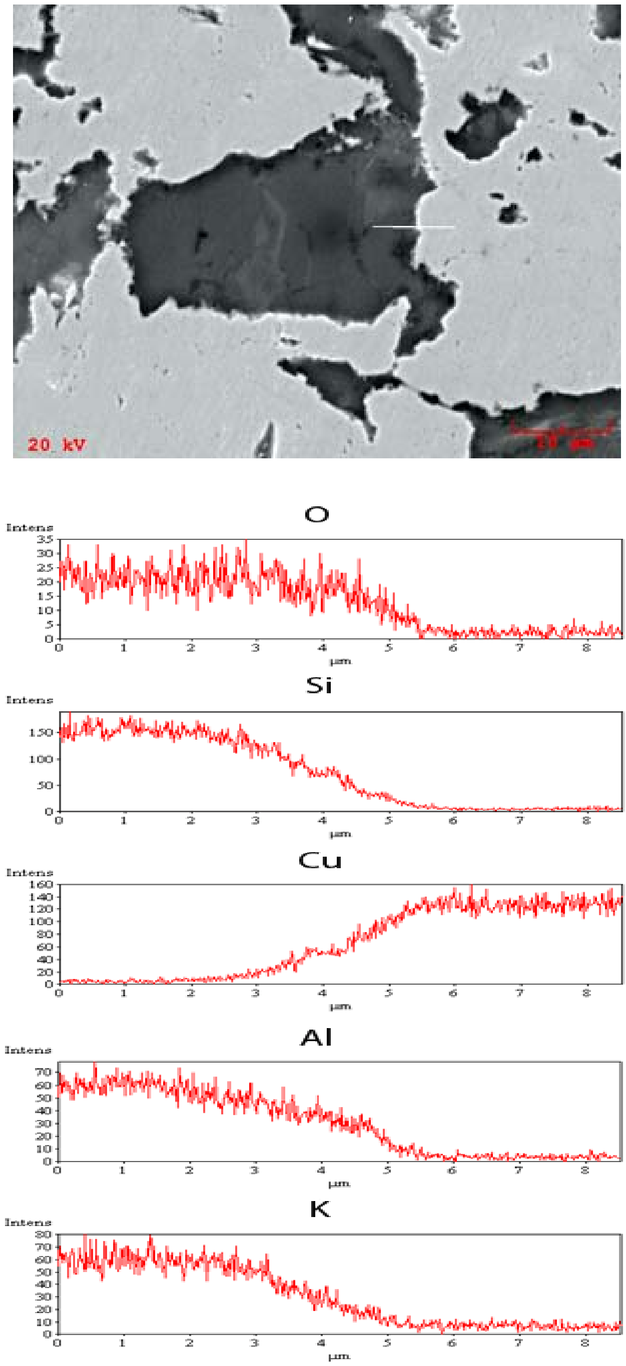


Fig. 6. Microstructure and linear distribution of elements for copper composite with tuff addition

Microanalysis results demonstrate that there might have occurred a slight diffusion of copper into the tuff particle. A trace of diffusion of the elements such as Al, K, Si, O into the copper matrix can also be seen. The results prove a good quality of the bonding between the introduced ceramic particles and the matrix. In this kind of composites, considering their application potential, the purity of copper is very important as it can affect its electric conductivity. The insignificant diffusion obtained in the composites produced is not likely to cause deterioration of the composite conductivity but can improve the quality of the bonding between the particles and the matrix.

Beside copper composites, 316L steel composites were also made to check how tuff particles would behave in another kind of matrix. Figure 7 shows 316L steel and 2% tuff composite microstructure.

Like in the case of copper sinter, there is an apparent variation of tuff particle size and even distribution of tuff particles. The tuff particles did not diffuse in spite of the fact that baking temperature was higher than in the case of copper. The introduced particles were not observed to aggregate. The analyses did not reveal changes in the chemical composition of the matrix which would prove that diffusion had taken place. Good quality of bonding between the particles and the matrix was obtained. No micro voids or discontinuities were observed at the interfaces. The particles noticed in the composite structure differ significantly in their border shapes as compared to the copper matrix composite. The tuff particles in steel 316L composites have a more regular shape. In the copper matrix composites the surface of the particles is more irregular.

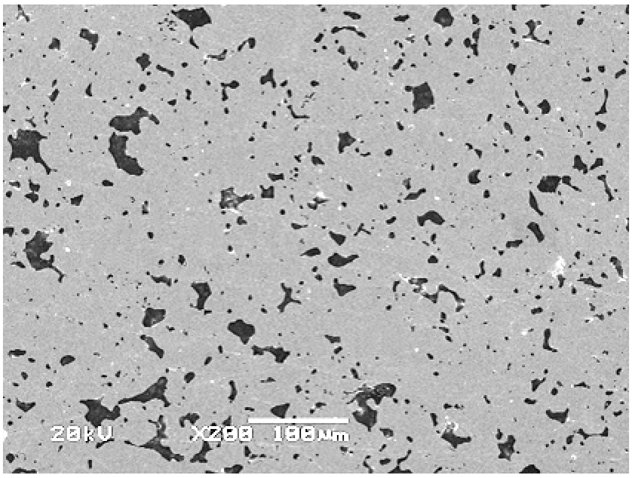


Fig. 7. 316L steel-2% tuff composite microstructure. SEM

Figure 8 shows 316L steel and 10% tuff composite microstructure. There is a clearly visible difference in the shape of the particles as compared to the copper composite microstructure presented in fig. 6.

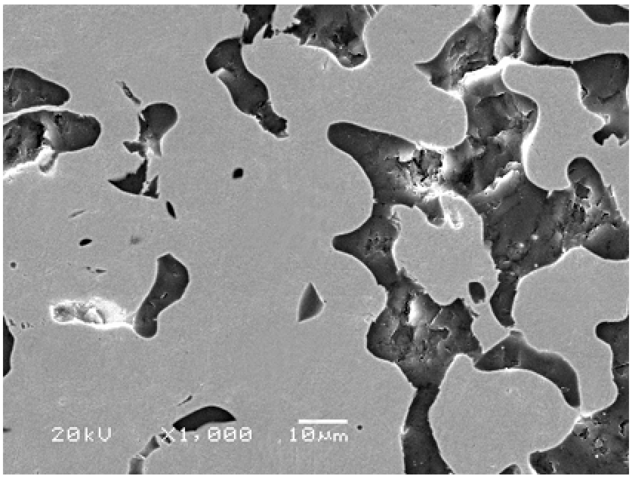


Fig. 8. 316L steel and 10% tuff composite microstructure. SEM

Figures 9 and 10 demonstrate the surface of fractures in copper and 316L steel sinters with 2% volcanic tuff. The fractures clearly differ in appearance. Tuff particles in the copper composite have particularly interesting forms. They look as if they were 'coated' with tiny particles. A microanalysis of these areas revealed them to be copper. The phenomenon proves a very good quality of the bonding between the tuff particles and the matrix to have been obtained.

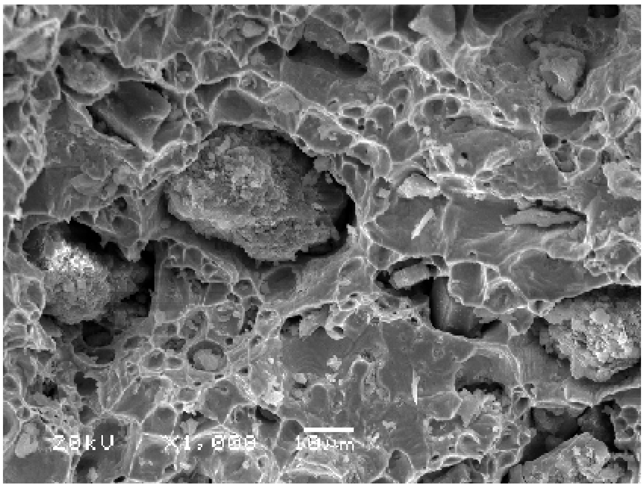


Fig. 9. Cu-2% tuff sinter fracture topography SEM

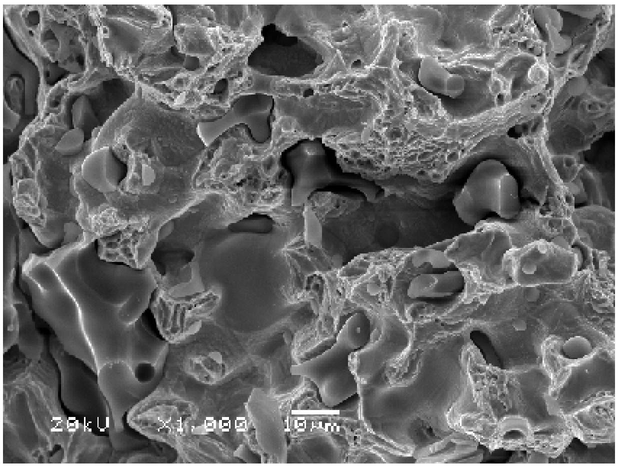


Fig. 10. 316L-2% tuff sinter fracture topography. SEM

5. Conclusion

The results of preliminary studies allow the following conclusions to be drawn:

1. It is possible to produce good quality metal-tuff composites using powder metallurgy.
2. Annealing tuff particles at 850°C for 4 hours improves the quality of the bonding between the tuff and the matrix. The particles introduced are coherent with the metal matrix.
3. The addition of tuff diminishes the porosity of copper composites. Specific gravity of the composites increases as the introduced particles participation increases.
4. Owing to the fact that tuff particles can be easily mixed with metallic powders their distribution in the matrix after sintering is even.
5. The particles do not decompose or diffuse in the matrix at the baking temperature of the composites produced.

The studies which were carried out and their analysis show that powdered volcanic tuff introduced into metal matrix improves the hardness and diminished the porosity of the composite and can be used as a strengthening material. The particle distribution is even and the quality of the bonding with the matrix is satisfactory.

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