DESIGN, FABRICATION AND CHARACTERIZATION OF CERAMIC - METAL COMPOSITES, AN OVERVIEW

PROJEKTOWANIE, WYTWARZANIE I CHARAKTERYSTYKA KOMPOZYTÓW METALOWO-CERAMICZNYCH: PRZEGLĄD

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Abstract: Composites are used for many different products in numerous industries applications. They satisfy the demand for new materials which combine dissimilar materials and represent properties which are not achievable by separate material. Among of them are ceramic matrix composites, especially with metal phase. Different systems have been developing. During the last years the composites with Al_2O_3, ZrO_2 ceramic matrix and metals as Ni, Mo, Cu, W, Co have been intensively studied.
Metallic particles embedded into ceramic matrix improved mechanical properties such as the toughness, the hardness and the wear resistance. Moreover, other properties as magnetic or electrical of ceramic-metal composites can be significantly changed. Because of that, ceramic- metal composites are considered as structural and functional materials.
The final properties of ceramic-metal composites depend on their microstructure, which should be optimize by the design of metal size and distribution into ceramic matrix and selection of the fabrication method.
This paper attempts to review the microstructures of ceramic-metal composites based on own experimental results. The following issues will be discussed: design the metal particles distribution into ceramic matrix, opportunities of processing in creating microstructures and fabrication of composites with complex shape and high dimensions, interactions in ceramic/metal interfaces, toughening mechanisms and properties of composites.
Keywords: ceramic-metal composite, ceramic matrix composite, microstucture, fabrication of composites, functional materials, metal particles distribution, properties of composites

Streszczenie: Z kompozytów wykonywane są elementy wykorzystywane do różnych zastosowań przemysłowych. Wynika to z możliwości uzyskania w nich właściwości nie spełnianych przez pojedynczy materiał. Wśród kompozytów o osnowie ceramicznej intensywne prace prowadzone są nad kompozytami z wprowadzoną faza metaliczną. Do częściej badanych układów zalicza się ceramikę Al_2O_3 oraz ZrO_2 w połączeniu z metalami

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jak Ni, Mo, Cu, W czy Co.
Wprowadzenie cząstek metalu do osnowy ceramicznej zapewnia zwiększenie odporności na kruche pękanie przy zachowaniu wysokiej twardości. Jednocześnie inne właściwości, jak elektryczne czy magnetyczne, mogą być modyfikowane. Dlatego też coraz częściej kompozyty ceramika-metal są postrzegane jako materiały o możliwości zastosowania jako konstrukcyjne i funkcjonalne. Właściwości kompozytów ceramika-metal zależą od ich mikrostruktury, która powinna być optymalizowana poprzez proces projektowania rozmiaru i rozmieszczenia cząstek metalu w osnowie ceramicznej oraz dobór metody wytwarzania.
W artykule przedstawiono przegląd mikrostruktury kompozytów ceramika-metal bazując na własnych pracach w tym zakresie. Analizowano sposób rozmieszczenia cząstek metalu w osnowie ceramicznej, zalety metod wytwarzania pozwalające na wykonywaniu złożonych kształtów próbek kompozytowych oraz o dużych rozmiarach. Omówiono oddziaływania na granicy międzyfazowej ceramika/metal oraz mechanizmy odpowiedzialne za wzrost odporności na kruche pękanie, jak i właściwości kompozytów.
Słowa kluczowe: kompozyt o osnowie ceramicznej, kompozyt metalowo-ceramiczny, mikrostruktura, materiał funkcjonalny, rozmieszczenie cząstek metalu, właściwości kompozytów

1. INTRODUCTION

Composites are used for many products in numerous industries applications. They satisfy the demand for new materials which combine dissimilar materials and represent properties which are not achievable by separate material. Among of them are ceramic matrix composites, especially with metal phase. Different systems have been developing. Composites with Al_2O_3, ZrO_2 ceramic matrix and metals as Ni, Al, Mo, Cu, W have been intensively studied [1–7].
Metallic particles embedded into ceramic matrix improved mechanical properties such as the fracture toughness, the hardness and the wear resistance [1,2,3,6,7]. Moreover, other properties as magnetic or electrical of ceramic-metal composites can be significantly changed [4]. Because of that, ceramic- metal composites are considered as structural and functional materials.
The final properties of ceramic-metal composites depend on their microstructure, which should be optimize by the design of metal size and distribution into ceramic matrix and selection of the fabrication method.
This paper attempts to review the microstructure of ceramic-metal composites based on own experimental results. Method of fabrication and characterization of ceramic-metal composites have been developed at Warsaw University of Technology by the Faculty of Materials Science and Engineering (group of K. Konopka) with cooperation with Faculty of Chemistry (group of M. Szafran) for more than 10 years. Most of experimental results were obtained in frame of projects of Ministry of Science and Higher Education, National Center of Science as well as in P.O.I.G.
The following issues will be discussed: design the metal particles distribution into ceramic matrix, opportunities of processing in creating microstructure and fabrication of composites with complex shape and high dimensions, influence of the ceramic/metal interfaces on the toughening mechanisms of composites.

2. TYPES OF CERAMIC-METAL COMPOSITES MICROSTRUCTURE

Three types of ceramic-metal composite microstructure can be distinguished: uniform, non-uniform and complex.

2.1. UNIFORM MICROSTRUCTURE

This type of microstructure is formed by uniformly distribution of metal particles or its clusters into ceramic matrix. It could be obtained by various methods. Own experiences based on powder metallurgy method (pressing and sintering powder mixture), colloidal methods (slip casting, tape casting and gelcasting) and infiltration of porous ceramic preforms by liquid metals. In all of these methods it is possible to create the uniform distribution of metal particles, which are embedded into ceramic matrix (Fig. 1). However, some factors in design and selection of processing must be considered.

One of them is ceramic-metal system. Combining the ceramics with metal the melting point of metal is the most important issue. For metals with a low temperature of melting (below the sintering temperature) during the sintering the metal will be turning into liquid and its distribution in solid body is controlled by size of pores. Moreover, the melted metal can be located at the boundaries of ceramic grains. For this, the type of microstructure where the metal particles are surrounded by the ceramic grains powder metallurgy method are not effective. In this case the infiltration of ceramic porous preform are recommended. Own results of infiltration porous Al_2O_3 preform by liquid Al confirmed fulfilled pores by metal. For improving ceramic wetting by metal the Ni-P coating of porous ceramic preform were proposed [8].

For all systems, where powder mixtures are prepared as the first step of processing the sedimentation and agglomeration of powder particles occurred. These two phenomena are the drawback of powder metallurgy methods. Mostly, in composites the agglomerations of a few metal particles are observed in ceramic matrix.

The process of sedimentation and agglomeration of metal powder particles can be avoided or limited by using colloidal methods as slip- or gelcasting methods. Especially, in gelcasting method the selected organic monomer and activator of polymerization are added into powder aqueous solution [9]. Prepared slurry is poured into form where the gelation process is activated. Due to polymerization the metal particles are surrounded by polymer chain which keeps the particles in the stable positions.

Own works with using gelcasting method were carried out on Al_2O_3-Ni system. As a result in sinters the even distribution of metal particles separated by ceramic grains in the whale volume are obtained (Fig. 1b). The influence of Ni particles on the gelation time was revealed. The catalytic action of metal particles, especially Ni should be considered in processing. The obtained samples were characterized by precisely reproduction of the shape of form. Moreover, the complex shapes can be produced [10].

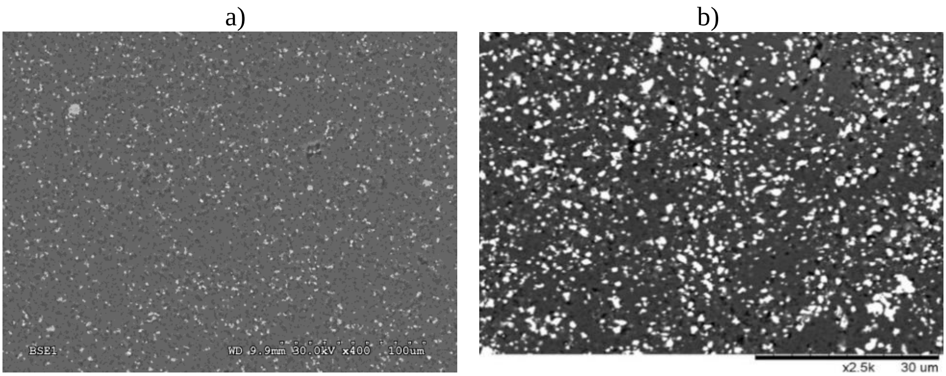


Figure 1. SEM images of composites: a) Al_2O_3-Ni composite obtained by slip casting method, b) Al_2O_3-Ni composite obtained by gelcasting method; light dots-Ni particles

2.2. NON-UNIFORM MICROSTRUCTURE

The graded structures represent the non-uniform distribution of metal particles into ceramic matrix. Similarly to uniform microstructures also the powder metallurgy and colloidal methods led to create the continuous changes of the volume contribution of metal particles. Moreover, the shape or size of metal particles in the composite can changed gradually. The models of such graded structures are presented in Figure 2.

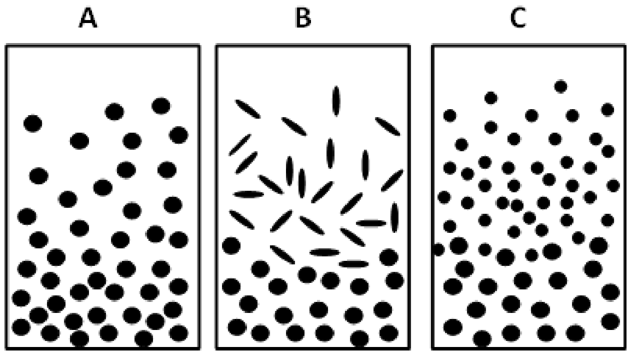


Figure 2. Schema of graded ceramic-metal composites: A. gradient of metal particles distribution into ceramic matrix, B. gradient of metal particles shape in ceramic matrix, C. gradient of ceramic particles size; black areas-metal particles

Own experimental works were concentrated on producing graded concentration of Ni or Fe particles in Al_2O_3 matrix by using slip-casting method [11,12,13]. Gravity force or magnetic field were responsible for graded distribution of metal particles. The Ni and Fe particles with higher density than alumina in a slip slurry have tendency to drop down. In addition, magnet during slip-casting stimulated to distribute metal particles according to the magnetic fields lines (Fig. 3) [11].

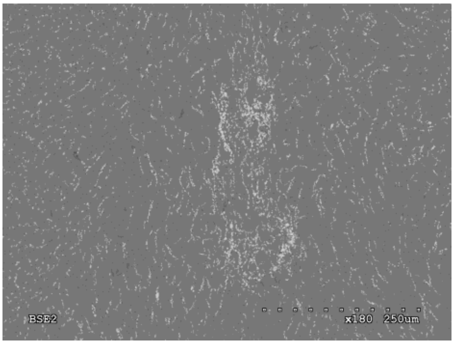


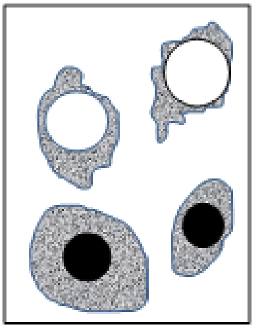
Figure 3. Al_2O_3-Ni composite obtained by slip casting method with using magnetic field; light dots- Ni particles distributed according to the magnetic lines

2.3. COMPLEX MICROSTRUCTURE

Besides the microstructure of composites with uniform or non-uniform distribution of metal particles the complex microstructure can be produced. Such microstructure is result of processing in which new phases appeared. The atmosphere of sintering can influence on the phase composition of composites. As a consequence, for example at Al_2O_3-Ni system is possible to obtained composites consisted of Ni particles embedded at Al_2O_3 matrix as well as spinel phase $NiAl_2O_4$ located in ceramic matrix [14,15]. Likewise at Al_2O_3-Fe system the spinel phase $FeAl_2O_4$ can be formed [11]. Investigation of phase composition of composites, their morphology and distribution revealed that spinel phase areas and metal phase particles can existed together in microstructure [14–17]. The characteristic morphology of spinel like “doughnut“ areas were observed (Fig. 4a,b). According to the stage of spinel forming, when the process is not fully accomplished also spinel as an layer surrounded metal particles can existed in microstructure (Fig. 4a,c) [14–17].

Second example of complex composite microstructure is ZrO_2-Ti system. The 3 mol.% Y_2O_3- stabilized ZrO_2 (3Y-TZP) is transformation-toughened ceramics. The good thermal properties, wear resistant and high fracture toughness are the most important features of this ceramics [18]. Combination with metal like Ni or Ti allows to keep these desired properties of ceramic matrix and improve the fracture toughness. It is result of a well- known mechanism of crack energy dissipation by metal particles. Moreover, in ZrO_2 ceramics the transformation t-ZrO_2 (tetragonal) into m-ZrO_2 (monoclinic) is expected. This martensitic transformation in ZrO_2 involves a shear strain and is treated as mechanism of improving the fracture toughness [19,20]. However, the volume change of material assists this transformation. There are some literature data indicated that the metal particles might have an influence on the transformation t-ZrO_2 into m-ZrO_2. The stress at the interfaces in composite, especially near the ZrO_2/metal interfaces enhance the transformation of ZrO_2 [20]. Moreover, because Ti is an active metal and easily react with O_2 or H_2 the new compounds during the sintering of composite might be formed.

a)



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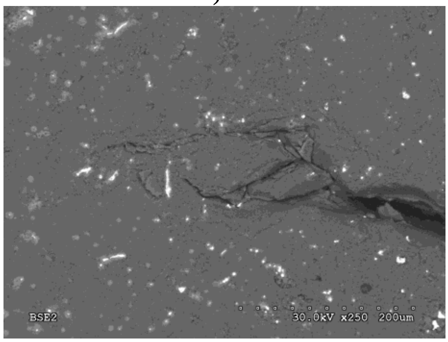

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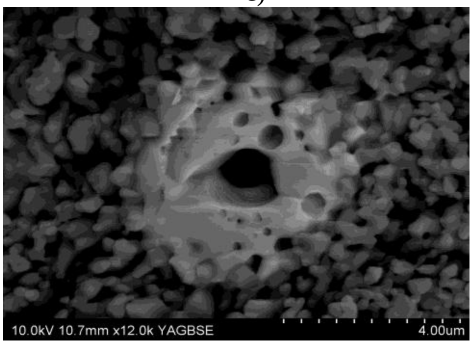


Figure 4. a) Schema of spinel distribution; grey areas – spinel distributed around metal particles (white areas) or with void inside the spinel (black dot), b) SEM image, Al_2O_3-Ni composite; white areas-Ni particles, light-grey areas spinel $NiAl_2O_4$, c) SEM image of Al_2O_3-Fe composite; void inside spinel

The preliminary research of ZrO_2-Ti composites which has been realizing in frame of project 2013/11/B/ST8/00309 finansed by National Center of Science confirmed active role of Ti in densification of composite and initiation of martensitic transformation of ceramic matrix. Together with increase of the Ti content in composites the significant increase of samples volume after sintering was noticed (Fig. 5a). Figure 5a present the photograph of the sintered composites ZrO_2-3 vol.% Ti and ZrO_2-10 vol.% Ti. Both sinters were prepared in the same way, the green bodies samples had the same diameter. After the sintering at 1400°C/1h in argon+H_2 for the composite with 10 vol.% of Ti the about 30% increased of the diameter of sample was noticed. It confirmed that the volume fraction of m-ZrO_2 increased with the Ti volume content. According to the literature, the volume change accompanied transformation of ZrO_2 from t-ZrO_2 into m-ZrO_2 are equal to about 5% [20]. Moreover, in the SEM observation the holes in microstructure of composites were observed (Fig. 5b). It can be result of reaction of Ti with atmosphere of sintering (mixture of argon and H_2). The experimental analysis of microstructure of ZrO_2-Ti composites are in progress.

a) b)

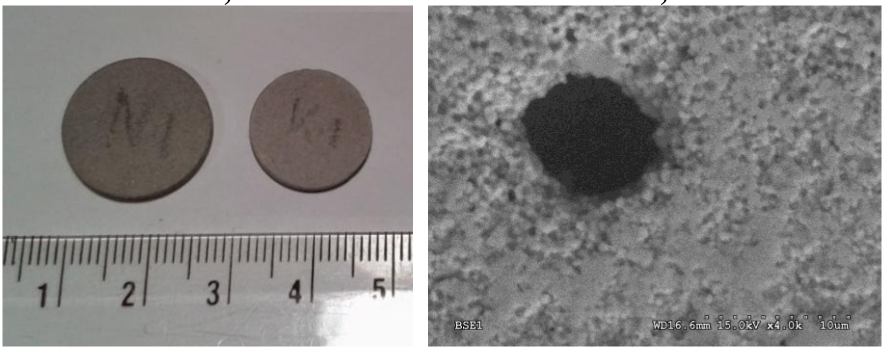


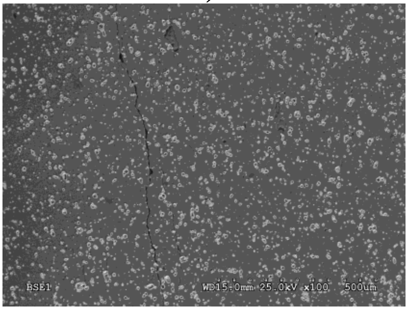
Figure 5. a) Photograph of samples, ZrO_2-3 vol.% Ti and ZrO_2-10 vol.% Ti after sintering at 1400°C/1h in argon+H_2; the larger sample – composite with 10 vol.% of Ti, the smaller sample composite with 3 vol.% of Ti, b) SEM image of ZrO_2-10 vol.% Ti; large hole visible

3. MICROSTRUCTURE AND FRACTURE TOUGHNESS

The properties of ceramic-metal composites depend on their microstructure. The most important from application point of view is the fracture toughness of composites. In all ceramic-metal systems metal phase in form of particles take part in process of crack propagation. The possible mechanisms which operate in composites are as follows: crack deflection, meandering and crack bridging. According to the literature which one of these mechanisms is dominant depend on the interfaces strength [4]. For interface ceramic/metal with firm bond the bridging is very intense. On the contrary, for weak interfaces the crack deflection mostly occurs [4]. In the complex microstructure also new phases and their interfaces are active too. In Al_2O_3-Ni and Al_2O_3-Fe systems the appearing of the spinel phase creates new interfaces, spinel/metal and spinel/ceramics. Their contribution into toughness process is also depend on their strength. The processes as crack deflection by the spinel/metal interface as well as crack cleavage are observed (Fig. 6a,b). Moreover, because of weak bonding at interface spinel/metal the pulling out of metal particles is noticed too (Fig. 6c).

Own experimental works of Al_2O_3-Ni and Al_2O_3-Fe showed that above described mechanisms of dissipation crack propagation energy could increase the fracture toughness even 2-times [10,11,16,17]. In ZrO_2-Ti system the mechanisms of improving the fracture toughness are investigated. Because of the complex microstructure in this system the fracture toughness can be result of few mechanisms active at once.

a)



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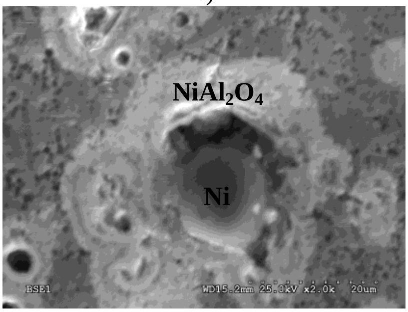

Figure 6. SEM images of Al_2O_3-Ni and Al_2O_3-Fe composites: a) crack propagation, b) crack cleavage by spinel area, c) pulling out of metal particles, after pulled out of particle the void is left

4. CONCLUSIONS

The present paper attempts to review the microstructure of ceramic-metal composites based on own experimental work done under supervising of the author. The three types of ceramic-metal microstructure can be distinguished: uniform, non-uniform and complex.

The type of microstructure depend on the many factors. Among of them are selected ceramics and metal, their difference especially melting point and density. The methods of fabrication led to produce the composites with various distribution of metal particles. Moreover, the complex microstructure depend on the possibility to form during the processing new phases. The final properties of composites, especially the fracture toughness are result of strength of interfaces which intensively take part in process of crack propagation.

At the stage of design of ceramic-metal composites consideration of above as well also others factors allow to predict the final microstructure and the fracture toughness.

ACKNOWLEDGEMENT

The author would like to thank Prof. M. Szafran and his team from The Faculty of Chemistry Warsaw University of Technology and MSc. And PhD. Students who were engaged at experimental works.

The overview paper based on own experimental date obtained in frame of projects of Ministry of Science and Higher Education and National Center of Science. Among of them there is the project which has been just realizing and supported by National Center of Science no 2013/11/B/ST8/00309.

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