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COMPOSITES THEORY AND PRACTICE

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ALUMINA MATRIX CERAMIC-NICKEL COMPOSITES FORMED BY GELCASTING METHOD

Currently, much attention has been focused on the development of ceramic composites with metallic particles due to their higher strength as well other properties compared to monolithic ceramic. Traditional methods of powder metallurgy are not capable of obtaining composite materials products of any shape in an uncomplicated way. For this reason, attempts have been made to adapt the gelcasting method (initially used only for ceramic powder) to obtain cermets. In the paper, the results concerning alumina-nickel ceramic composites fabrication via the gelcasting method are presented. The paper presents the effect of the addition of nickel on the polymerization time of composite slurries. The characteristics of selected physical and mechanical properties of composites containing 5 vol% and 10 vol% of nickel have been described and compared to the results achieved for 100% Al_2O_3. Tests were conducted for the green body and for sintered samples. Observations of the microstructure of the produced Al_2O_3-Ni composites are also presented. Computer image analysis was performed for the nickel particles in the composites. The obtained results confirmed the possibility of applying the gelcasting method for the preparation of alumina-nickel composites.

Keywords: **cermetals, alumina, nickel, gelcasting**

KOMPOZYTY Al_2O_3-Ni FORMOWANE METODĄ ODLEWANIA ŻELOWEGO (GELCASTING)

Obecnie wiele uwagi skupiane jest na rozwoju ceramicznych kompozytów z cząstkami metalicznymi ze względu na wyższe właściwości wytrzymałościowe w porównaniu do ceramiki monolitycznej oraz inne właściwości. Tradycyjne metody metalurgii proszków nie dają możliwości w sposób mało skomplikowany otrzymania wyrobów z materiałów kompozytowych o dowolnym kształcie. Z tego względu podjęte zostały próby zaadaptowania do otrzymywania cermetali metody odlewania żelowego (gelcasting), początkowo stosowanej tylko dla proszków ceramicznych. Metoda ta polega na bezpośredniej konsolidacji proszków na drodze reakcji polimeryzacji monomeru organicznego wewnątrz ceramicznej masy lejnej. W niniejszej pracy przedstawione zostały wyniki badań nad otrzymywaniem kompozytów Al_2O_3-Ni metodą gelcasting. W artykule zaprezentowano wpływ dodatku niklu na czas polimeryzacji kompozytowych mas lejnych. Opisana została charakterystyka wybranych właściwości kompozytów o zawartości niklu 5% obj. i 10% obj. oraz dla porównania wyniki uzyskane dla 100% Al_2O_3. Badania przeprowadzone zostały zarówno dla próbek surowych, jak i po procesie spiekania. Przedstawione zostały również obserwacje mikrostruktury wytworzonych kompozytów oraz wykonana została analiza stereologiczna dla cząstek niklu w kompozycie. Uzyskane wyniki potwierdziły możliwość wykorzystania metody gelcasting do otrzymywania kompozytów Al_2O_3-Ni.

Słowa kluczowe: **cermetale, tlenek glinu, nikiel, odlewanie żelowe**

INTRODUCTION

Currently, much attention has been focused on the development of ceramic composites with metal particles because these materials offer good mechanical and multifunctional properties [1].

Classical methods of powder metallurgy [2] used for ceramic-metal composites are not capable of fabricating a product of complex shape while maintaining the high homogeneity and mechanical strength of the material. The review of scientific publications shows that methods based on colloidal processes - such as gelcasting - for advanced composite materials fabrication have gained much attention nowadays [3-5].

Gelcasting is a combination of the conventional moulding method from slips with polymerization reaction, at first applied to ceramic materials [6]. This method is based on the fact that an organic monomer, a mixture of dispersants and polymerization activator are added to the aqueous slurry of the powder. Then the slurry is thoroughly mixed and degassed. After adding the initiator, the slurry is poured into the mold. Then the slurry undergoes a gelation process. The resulting green body shapes are characterized by good reproduction of form and high mechanical strength [7].

EXPERIMENTAL PROCEDURE

Preparation of suspension

The composites were fabricated from an α-Al_2O_3 powder (TM-DAR, Tamei Japan) of an average particle size D_{50} = 0.21 μm and density 3.8 g/cm^3 and a nickel powder (Sigma-Aldrich) with an average particle size D_{50} = 10.45 μm and density 8.9 g/cm^3 (Fig. 1). The powder sizes were determined by the dynamic light scattering method.

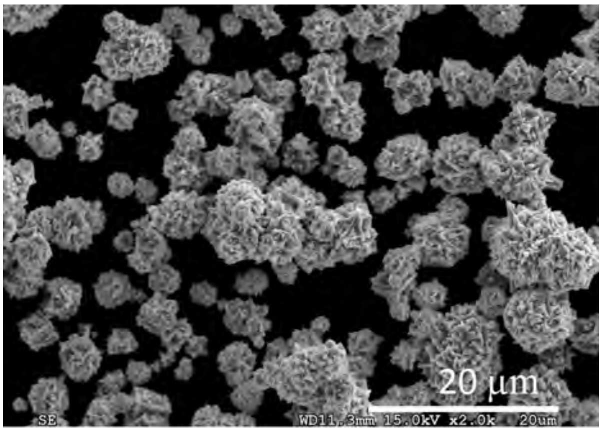

Fig. 1. Scanning electron microscopy image of nickel powder

Rys. 1. Zdjęcie z mikroskopu skaningowego użytego proszku niklu

Water-based slurries with 54 vol. % solid content were prepared with respectively 0, 5 and 10 vol. % of nickel powder. A mixture of diammonium citrate (0.3 wt.%) and citric acid (0.1 wt.%) was added in the role of deflocculant, which was dissolved in distilled water. As a monomer and cross-linking agent in the gelcasting process, 2-hydroxyethyl acrylate (3 wt.%) and N,N`-methylenebisacrylamide (2 wt.% with respect to the quantity of monomer) were used. The activator was N,N,N,`N`,-tetramethylenediamine (1 wt.% with respect to the amount of monomer), and ammonium persulfate (0.3 wt.% with respect to the amount of monomer) served as the polymerization initiator.

The ingredients (without initiator) were homogenized in planetary ball mill with a rotating speed of 250 r.p.m. for 80 min. Subsequently, the slurries were degassed for 10 min under low pressure in a vacuum desiccator with a magnetic stirrer. Immediately before casting, the polymerization initiator was added and the slurry was mixed by a magnetic stirrer for 1 min. The casted samples were cylindrical with a diameter of 17 mm and height of about 4 mm. The specimens were dried for 96 h at room temperature (25ºC) and sintered in a hydrogen atmosphere at a temperature of 1350°C for 2 hours.

Characterization techniques

Rheological measurements of the ceramic and composite slurries were examined using a Brookfield DV II Pro.

The tensile strength of the green bodies was performed by the “Brazilian test” [8] using a Universal Testing Machine TINIUS OLSEN TOROPOL H10KS. The tensile strength was calculated based on the following equation:

$$\sigma_t = (2P/\pi hD) \quad (1)$$

where P is force, and h, D are the height and diameter of the obtained sample.

The physical parameters of the sintered samples (such as relative density) were estimated by means of the hydrostatic method. The linear shrinkage was calculated from a change in the sample dimensions.

The microstructure was examined by scanning electron microscope (SEM). In order to confirm uniform nickel particle distribution, microscopy observations were carried out on two cross-sections of the samples as in Figure 2.

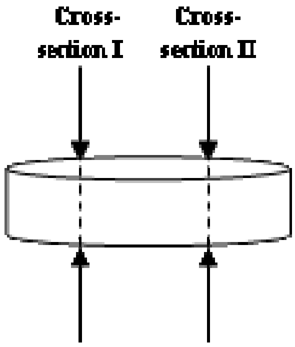

Fig. 2. Cutting cross-section of composites samples for microscopic observation

Rys. 2. Płaszczyzny cięcia próbek kompozytowych do obserwacji mikroskopowych

Measurements of the nickel particles geometry were carried out using a computer image analyzer [9]. The following parameters of the particles have been measured [10]:

- d - average equivalent diameter of circle whose area is equal to measured cross-sectional area of grain,
- A - average equivalent surface area of cross section of Ni particles,
- d_{max} - longest particle chord,
- d_{max}/d - parameter used to describe shape of particle sections called shape factor (is sensitive to grain elongation), which equals 1 for a circle.

RESULTS AND DISCUSSION

The results of viscosity changes in time for the ceramic and composites slurries have been presented in Figure 3.

Alumina and nickel composites slurries show great differences compared to alumina slurries when the time of gelation (time after which viscosity increases sharply) is taken into account. The gelation time for

Al_2O_3-Ni is about 5 minutes while for alumina it is about 35 minutes. This is caused by the catalytic action of nickel powder on the polymerization process. For the used quantity of nickel, an addition of activator (N,N,N,`N`,-tetramethylenediamine) was required, because without it the polymerization process for composite materials was very long, over 120 minutes.

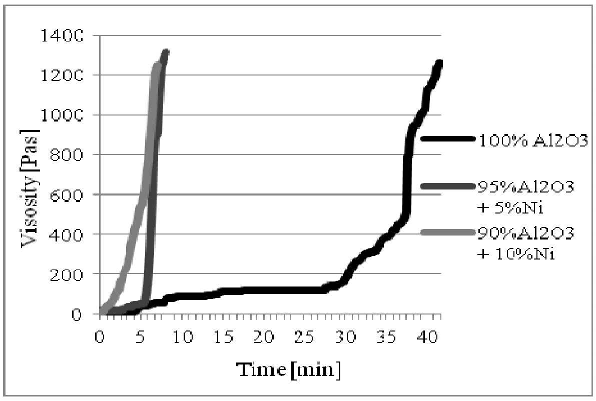

Fig. 3. Influence of quantity of nickel powder on viscosity of ceramic slurries

Rys. 3. Wpływ dodatku niklu na zmianę lepkości ceramicznej masy lejnej

In Table 1, the properties of the obtained materials are listed. Both alumina and composite green bodies are comparably well-densified and their measured relative density is about 60% for all the tested samples. With an increasing amount of nickel, a decrease in tensile strength was observed. This might be caused by worse adhesion of the polymer to nickel particles than to alumina particles, which is the subject of further investigation.

The total linear shrinkage of the specimens was about 12.5% for alumina samples and about 14% for composites. The final relative density was about 97% for all the specimens.

TABLE 1. **Physical and mechanical properties of green body and sintered samples of Al_2O_3 and Al_2O_3 + Ni composites**

TABELA 1. **Wybrane właściwości fizyczne oraz mechaniczne dla kształtek i spieków Al_2O_3 oraz kompozytów Al_2O_3 +Ni**

Nickel content	0 vol. %	5 vol. %	10 vol. %
Properties of green body			
relative density d_{rel} [%]	57.8 ± 1.5	61.1 ± 0.1	60.0 ± 0.2
tensile strenght [MPa]	1.65 ± 0.18	0.90 ± 0.10	0.49 ± 0.1
Properties of sintered samples			
relative density d_{rel} [%]	97.5 ± 0.9	96.2 ± 1.0	96.6 ± 1.3
linear shrinkage S_l [%]	12.5 ± 1.5	14.5 ±0.7	13.9 ± 0.4
open porosity Po [%]	1.2 ± 0.6	1.79 ± 0.8	2.7 ± 1.0

± confidence interval with probability 0.99

In Figures 4 and 5, typical Al_2O_3-Ni composite microstructures with 5 vol.% and 10 vol.% of nickel are presented. The nickel particles are not distributed uniformly in the Al_2O_3 matrix. Metallic particle agglomerates were observed, as shown in Figure 4. Their presence may be due to a too short mixing time of the slurry or sedimentation of the nickel particles in the composite suspension.

Table 2 presents the results of computer image analysis performed for two parallel cross-sections of the sample (see Figure 2). Similar results for both cross-sections suggest that the cermets have a uniform structure throughout the volume.

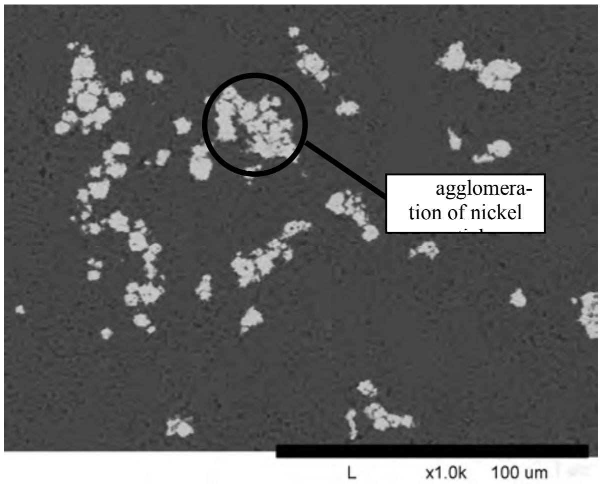

Fig. 4 SEM of sintered body of Al_2O_3+5% Ni composite [light field - nickel]

Rys. 4. Zdjęcie z mikroskopu skaningowego spieczonej kształtki kompozytu Al_2O_3+5% Ni [pola jasne - nikiel]

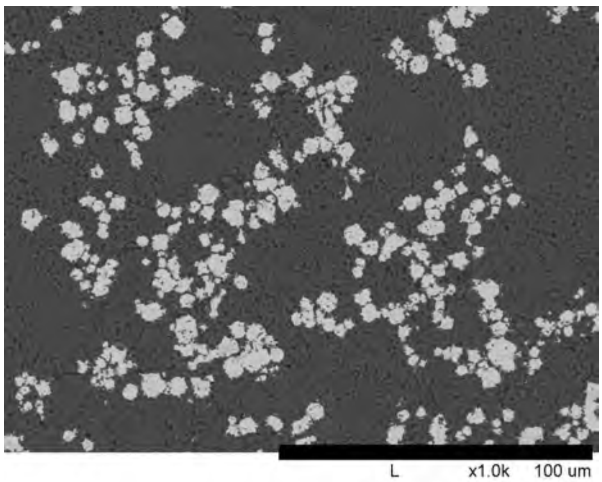

Fig. 5. SEM of sintered body of Al_2O_3+10% Ni composite [light field- nickel]

Rys. 5. Zdjęcie z mikroskopu skaningowego spieczonej kształtki kompozytu Al_2O_3+10% Ni [pola jasne - nikiel]

For both the obtained composite materials the average particle diameter was about 3.5 µm with a standard deviation of more than 2 µm. The wide range of results is due to the presence of nickel particle agglomerates.

The value of parameter d_{max}/d, which describes the shape of the Ni particles for both the tested cermets is close to 1, which shows the circular geometry of the nickel particle. Greater elongation was observed in the composite containing 5 vol.% Ni.

TABLE 2. **Stereological parameters of nickel particles**
TABELA 2. **Parametry stereologiczne cząstek niklu**

Ni content	5 vol. %		10 vol. %	
	I cross-section	II cross-section	I cross-section	II cross-section
d [μm]	3.22±2.00	3.49±2.25	4.24±2.64	3.34±2.25
A [μm^2]	11.27±13.26	13.54±17.67	19.57±26.03	12.71±15.20
d_{max}/d	1.41	1.40	1.35	1.35

± standard deviation

CONCLUSION

Alumina-nickel composites were fabricated successfully by the gelcasting technique. The resulting material replicated the casting form well. The composites were characterized by even distribution of nickel particles in the entire volume of the material.

However, the presence of nickel particles has an influence on the gelcasting process. Nickel has a catalytic influence on the polymerization process of 2-hydroxyethyl acrylate, as it was observed that the gelation time for composite slurries was much shorter than the time measured for the slurries without nickel particles. On the other hand, the polymerization process does not occur, unless an activator/initiator is added. This means that the catalytic action of nickel is not enough large and the gelcasting method can be applied to ceramic-metal composite fabrication. However, the catalytic action of nickel should be taken into account and the slurry composition should be carefully designed in order to control the gelation time.

A decrease in tensile strength was observed for composite green bodies in comparison to a ceramic obtained in the same conditions.

Acknowledgments

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