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COMPOSITES THEORY AND PRACTICE

Aleksandra Miazga[1]*, Katarzyna Konopka[1], Agnieszka Idźkowska[2], Mikołaj Szafran[2]

[1] Warsaw University of Technology, Faculty of Materials Science and Engineering, ul. Wołoska 141, 02-507 Warsaw, Poland
[2] Warsaw University of Technology, Faculty of Chemistry, ul. Noakowskiego 3, 00-664 Warsaw, Poland
**Corresponding author. E-mail: aleksandramiazga86@gmail.com*

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APPLICATION OF GELCASTING METHOD IN CERAMIC-METAL COMPOSITE FABRICATION

An attempt has been made to prepare Al_2O_3/Ni composites with 1 vol.% metal particles by the gelcasting method. Alumina matrix composites with an addition of nickel particles, already used in industry, are produced by various methods. Unfortunately, there is a problem to obtain elements of a complex shape characterized by a uniform distribution of metal particles in a ceramic matrix. For this reason, the authors have attempted to apply the gelcasting method to produce an Al_2O_3 + +1 vol.% Ni composite. The electrokinetic properties of the composite slurries and the influence of the presence of nickel have been studied. The microstructure of green and sintered samples were examined with a scanning electron microscope. Selected physical properties of the composite have been described. The material in the green state was characterized by a density of about 57%, and 97% after sintering, of the theoretical density. Nickel particles were uniformly distributed in the ceramic matrix. The performed studies have confirmed the possibility of using the gelcasting method to produce an alumina-nickel composite.

Keywords: ceramic composites, nickel, gelcasting

WYKORZYSTANIE METODY GELCASTING W WYTWARZANIU KOMPOZYTÓW CERAMIKA-METAL

Celem wytwarzania kompozytów ceramika/metal jest poprawa odporności na pękanie. Kompozyty o osnowie tlenku glinu z dodatkiem niklu są już obecnie materiałem stosowanym. Materiał ten wytwarzany jest różnymi metodami. Problemem niestety jest uzyskanie elementów o skomplikowanym kształcie charakteryzujących się jednorodnym rozmieszczeniem cząstek metalu w ceramicznej osnowie. Z tego powodu autorzy podjęli próbę aplikacji metody gelcasting do wytworzenia kompozytu Al_2O_3 + 1% obj. niklu. Przeprowadzone zostały badania właściwości elektrokinetycznych, wpływu obecności niklu oraz ilości incjatora na czas polimeryzacji. Próbki w stanie surowym oraz po procesie spiekania badano z wykorzystaniem skaningowego mikroskopu elektronowego. Określone zostały także wybrane właściwości fizyczne kompozytu. Materiał w stanie surowym charakteryzował się gęstością ok. 57%, natomiast po procesie spiekania 97% gęstości teoretycznej. Cząstki niklu były jednorodnie rozmieszczone w ceramicznej osnowie. Wykonane badania potwierdziły możliwość wykorzystania metody gelcasting w wytwarzaniu kompozytów Al_2O_3\Ni.

Słowa kluczowe: kompozyty ceramiczne, nikiel, metoda odlewania żelowego

INTRODUCTION

Till now, different processing routes have been examined to produce alumina-nickel composites such as: pressing [1], coating powder pressing [2], the sol gel method [3], slip casting [4] and other methods. It has been reported that there is a major problem associated with obtaining large-sized elements of complex shapes [5]. Other problems include achieving a uniform microstructure because of the great differences in density of the ceramic and metallic particles. The authors have made an effort to use the gelcasting method to obtain an alumina/nickel composite. Gelcasting is a near-net-shape forming method for advanced ceramics and composites [6]. The process involves a slurry prepared from a powder, a solvent, a mixture of deflocculant and monomer solution which is poured into a mold, polymerized in-situ to immobilize the particles in the gelled part, removed from the mould while still wet, then dried and sintered. The suspension of ceramic/metal powders is a very complex system because particles with a different surface charge may interact with each other and heteroflocculation effect takes place [7]. Interaction between the particles can prevent segregation of the heavier materials.

EXPERIMENTAL PROCEDURE

Materials

The raw materials used in gelcasting are listed in Table 1.

TABLE 1. **Raw materials used in gelcasting process**
TABELA 1. **Substancje użyte w procesie gelcasting**

Raw materials	Function
Alumina [TM-DAR] D_{50} = 0.21 μm	Ceramic powder
Nickel [Sigma-Aldrich] D_{50} = 10.21 μm	Metal powder
Deionized water	Solvent
Diammonium citrate (DAC)	Deflocculant
Citric acid (CA)	Deflocculant
2-hydroxyethyl acrylate	Monomer
N,N`-methylenebisacrylamide	Cross-linking agent
N,N,N,`N`,-tetramethylenediamine (TEMED)	Activator
Ammonium persulfate	Initiator

Slurries with a 50 vol.% solid content were prepared with 1 vol.% nickel powder. The mixture of deflocculant (DAC-0.3 wt.%; CA-0.1 wt.%), monomer (3 wt.%), cross-linking agent (2 wt.% with respect to quantity of monomer) was dissolved in water. The powder slurries were mixed in a planetary ball mill with a rotating speed of 300 r.p.m. for 60 min. The homogenized slurries were degassed for 15 min under low pressure in a vacuum desiccator. Subsequently, the activator (1 wt.% with respect to amount of monomer) and initiator (0.5÷1 wt.% with respect to amount of monomer) were added. The next step was casting into rubber moulds. The composite samples were cylindrical with a diameter of about 18 mm and height of 5 mm. The samples were dried for 72 h (at room temperature - about 25°C) and sintered in a reducing atmosphere (mixture of argon and hydrogen: 80 and 20 vol.% respectively at a temperature of 1400°C for 5 hours.

Test method

The zeta potential measurements in this study of the pure powders and powders mixed with of all the components for all the suspension and agglomerate distributions were conducted on a Zetasizer 3000 (Malvern Instruments). The pH of the suspension was adjusted using a 0.1 mol/dm^3 HCl or NaOH solution and varied from 2 to 12. The rheological measurements of the ceramic and composite slurries were examined using a Brookfield DV II Pro.

The density of the obtained bodies was measured by the Archimedes method in water. Each measurement was carried out on 15 samples.

The microstructure of the green and sintered specimens was observed with a HITACHI SU-70 scanning electron microscope. Based on the SEM images of two parallel cross-sections of polished samples, measurement of nickel particle size d (average equivalent diameter of circle whose area is equal to measured cross sectional area of the particle) was carried out using a computer image analyzer [8]. Calculations were performed for more than 200 nickel particles.

RESULTS AND DISCUSSION

Flowable, yet stable composites slurries were produced. Figure 1 shows the change of the zeta potential of the alumina powder, nickel powder and their mixture with additives (water, deflocculant, monomer, cross-linking agent). The isoelectric zero charge points of alumina and nickel were 4 and 9 respectively. This means that in the range of pH 4÷9, the surface charge of the alumina powder and nickel powder in water are opposite to each other. In such a suspension, the heteroflocculation effect takes place [9].

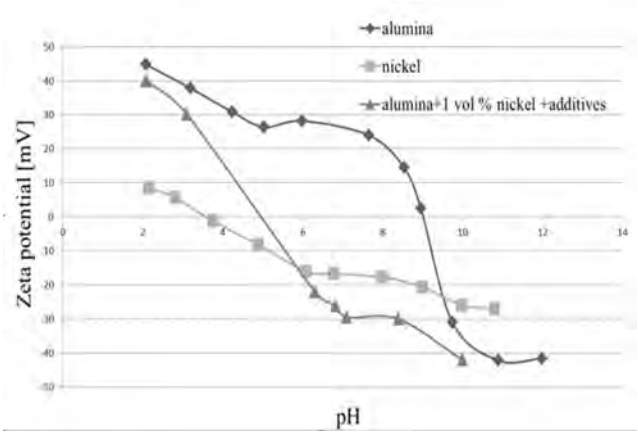

Fig. 1. Zeta potential as a function of pH of Al_2O_3, Ni powders and their mixture in presence of additives

Rys. 1. Potencjał zeta w funkcji pH proszku tlenku Al_2O_3, Ni oraz ich mieszaniny wraz z dodatkami

The zeta potential measurement for the composite slurry with the addition of additives showed a zero charge point at pH 5. Alumina-nickel agglomerates were formed. The interaction between the particles prevents segregation of the heavier material. Measurement of the particle size distribution of the composite slurry showed two maximum points: at about d = = 200 nm indicating a good degree of dispersion of alumina in a suspension and at d = 10 μm corresponding to particles of nickel and Al_2O_3/Ni agglomerates created as a result of the electrostatic attraction between the particles of opposite electrical charge .

It is very important to control the time after which the gelation process occurs. The amount of activator in the composite slurries was constant and fixed as 1 wt.% based on the monomer amount. The quantity of initiator was therefore matched. It was noticed that the presence of nickel particles accelerated the polymerization reaction four times in comparison to the alumina slurry. It can be due to the catalytic action of nickel with the monomer that was used [10]. For this reason, the quantity of initiator was reduced and the polymerization time was examined (Fig. 2). The optimum amount of initiator was determined as 0.5 wt.% in respect to the monomer. In this case the time lag was about 20 minutes.

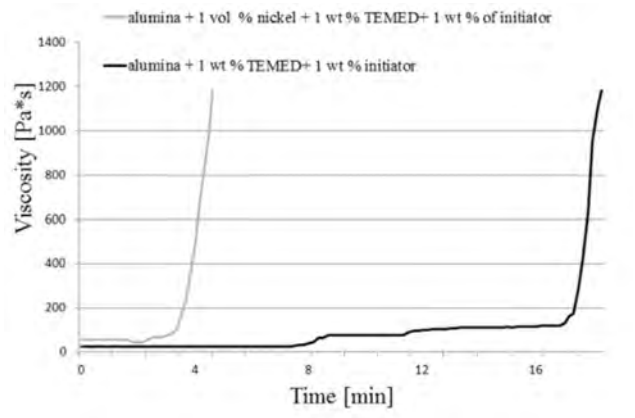

Fig. 2. Influence of presence of nickel on slurry viscosity

Rys. 2. Wpływ obecności niklu na lepkość masy lejnej

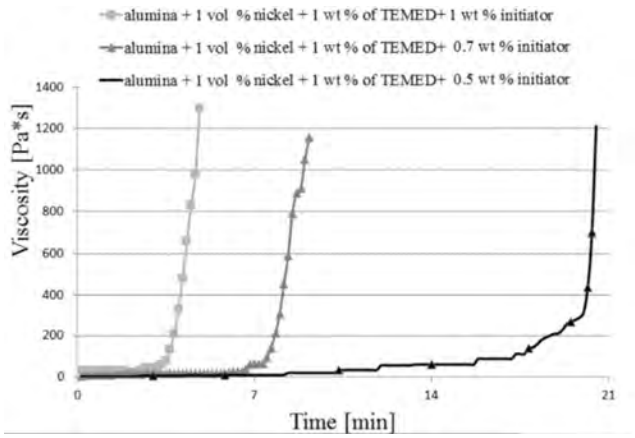

Fig. 3. Influence of quantity of initiator on composite slurry viscosity

Rys. 3. Wpływ ilości inicjatora na zmianę lepkości w czasie kompozytowych mas lejnych

The SEM microstructure of the green body (Fig. 4) confirms that the composite specimens are homogenous and the monomer can form a polymer network. No clusters of polymeric chains are visible. The nickel particles are covered with alumina particles. This confirms good wettability of the ceramic and metallic grains by the monomer.

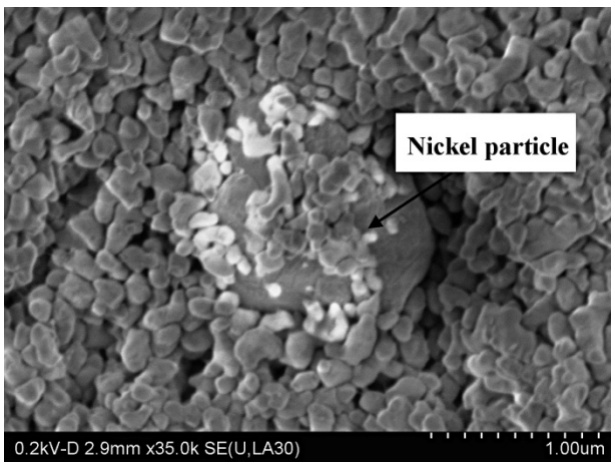

Fig. 4. SEM microstructure of composite green body fracture surface

Rys. 4. Zdjęcie SEM mikrostruktury kompozytu w stanie surowym

Characterization of the composite samples obtained by gelcasting are summarized in Table 1. The green density was about 55% of the theoretical value. After the sintering process, the relative density was about 97% respectively. The reproducibility of the results can be considered as high.

TABLE 2. **Selected properties of composite material**
TABELA 2. **Wybrane właściwości kompozytu**

Property	Value	Confidence interval with probability 0.99
Green body relative density [%]	55.4	1.6
Relative density [%]	97.3	1.5
Open porosity [%]	1.83	0.88
Linear shrinkage [%]	12.13	1.5
Soaking [%]	1.36	0.58

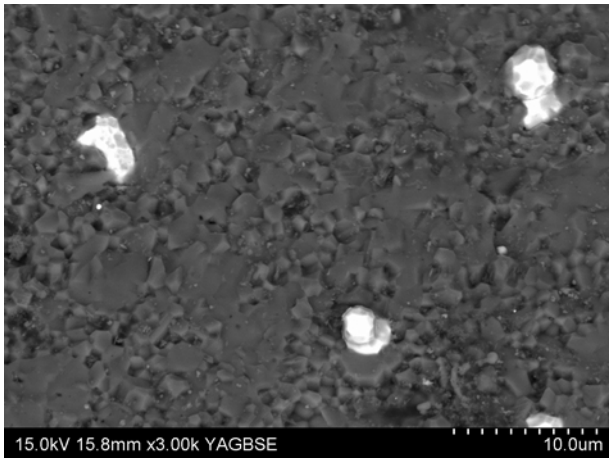

Fig. 5. SEM image of composite fracture surface (white fields - nickel particles)

Rys. 5. Zdjęcie SEM powierzchni przełomu kompozytu (jasne pola - cząstki niklu)

Figure 5 shows the SEM micrographs of a typical fracture of the obtained composites. The nickel particles are dispersed uniformly in the ceramic matrix. The microstructure was homogeneous, with no visible pores or cracks. The nickel particle size ranged from about 1 to 10 microns. The average particle diameter (expressed as average spherical equivalent d2) evaluated by stereological analysis was 2.86 μm (with standard deviation of ±3.77 μm), which is smaller than the value of the average particle diameter of the used powder. It can be caused by the fact that the initial nickel powder was agglomerated and during the mixing process in water, some agglomerates separated. Furthermore, the morphology of the metallic powder was spherical with some ridges. On the cross-section they can look like small single particles, hence the low d2 value.

Figure 6 shows a photograph of a sintered composite sample fabricated by gelcasting.

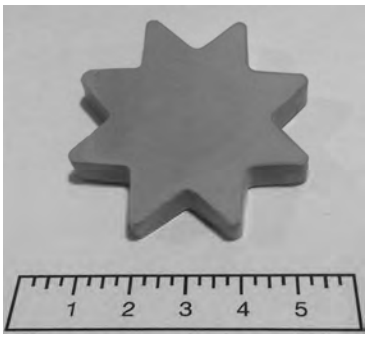

Fig. 6. Photograph of sintered composite sample fabricated by gelcasting
Rys. 6. Zdjęcie kształtki kompozytowej wykonanej metodą gelcasting

CONCLUSIONS

An alumina/nickel composite with 97% relatively density of the sintered body was successfully fabricated by the gelcasting method. The nickel showed the effect of a catalyzing polymerization reaction in case of the used monomer. For this reason, it was necessary to reduce the amount of initiator. In the composite slurry, the heteroflocculation effect occurred therefore distribution of the nickel particles in the ceramic matrix was homogeneous. The SEM micrographs showed that the particles were surrounded by ceramic grains with no visible cracks on the border between the ceramic and metal.

Acknowledgements

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