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COMPOSITES THEORY AND PRACTICE

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FABRICATION OF GRADED ALUMINA-NICKEL COMPOSITES BY CENTRIFUGAL SLIP CASTING

Microstructural graded hollow cylinders were processed and characterized using the developed method called centrifugal slip casting (CSC). Water based slurries containing alumina and nickel powders were tested in this work. Two series were produced by the CSC technique at mould rotational speeds of 1000 rpm. The samples were prepared with a solid content of 50 vol.% with 10 and 20 vol.% nickel particles with respect to the total solid volume, respectively. The microstructure along the radial direction of a cross-sectional sample was presented. No new phases were observed after sintering. Microstructural observation and EDS analysis showed two zones with various concentrations of Ni particles in both series along the radial direction of the cylinders.

Keywords: composites Al_2O_3-Ni, centrifugal slip casting technique, microstructure

WYTWARZANIE KOMPOZYTÓW GRADIENTOWYCH Al_2O_3-Ni METODĄ ODLEWANIA ODŚRODKOWEGO

Z użyciem metody odlewania odśrodkowego mas lejnych wytworzono próbki w kształcie walca, które następnie scharakteryzowano. W niniejszej pracy badano zawiesiny wodne zawierające tlenek glinu oraz nikiel. Dwie serie zostały wytworzone techniką odlewania odśrodkowego z mas lejnych przy prędkości obrotowej 1000 rpm. Przygotowano próbki o zawartości 50% objętościowo zawartości fazy stałej, w tym udział fazy metalicznej wynosił odpowiednio 10 oraz 20% objętościowo. Przeprowadzono obserwacje mikrostruktury wzdłuż kierunku promieniowego na przekroju poprzecznym próbek. Nie zaobserwowano nowych faz po spiekaniu. Obserwacja mikrostruktury i analizy EDS wykazały obecność dwóch stref o różnej koncentracji cząstek Ni w obydwu seriach wzdłuż kierunku promieniowego cylindra.

Słowa kluczowe: kompozyty Al_2O_3-Ni, metoda odlewania odśrodkowego z mas lejnych, mikrostruktura

INTRODUCTION

For several years now we have seen the rapid development of new technologies associated with the demand for new materials. The skilful combination of materials and their manufacturing techniques allow one to obtain increasingly better materials with unique properties and microstructure. Examples of such materials may be functionally graded materials (FGM). These materials may be an alternative to conventional materials. FGM belongs to a group of advanced materials characterized by variation in their properties as the dimension varies [1-6]. Due to the fact that FGM is characterized by a change in the physical and chemical properties of the whole volume, they may be successfully used in many applications [5]. These materials can be used as a structural material, and also as functional materials in biomedical applications, in the aerospace or military industries.

FGM can be fabricated by a variety of methods, such as: tape casting [7, 8], slip casting and filtration [9, 10], centrifugal casting [11-13], centrifugal slip casting (CSC) [14,15], pulsed laser deposition (PLD) [16, 17], self-propagating high-temperature synthesis (SHS) [18] and electrophoretic deposition (EDP) [19-22].

Literature data and the authors' own research has shown that the centrifugal slip casting technique is much cheaper and easier to use compared to the others [14, 15, 23, 24]. The CSC method allows one to obtain composites in the shape of a hollow cylinder. Al_2O_3-Ni composites with a gradient concentration of Ni particles may be an example of FGM obtained by the centrifugal slip casting method. The location of the metal particles gradient may be controlled through several parameters such as: speed or time of mixing, the size of the used powders, solids content or the amount of metallic phase.

Therefore, this study focused on the research of Al_2O_3-Ni FGMs containing different amounts of metallic phase. The tested Al_2O_3-Ni composites contained

10 and 20 vol.% the metallic phase, respectively. Furthermore, ceramic and metallic powders of significantly different sizes were applied.

MATERIALS AND METHODS

CSC is a method that combines traditional slip casting with centrifugal casting. The advantage of the connected operation of the two processes - centrifugation casting and slip casting, is that the fluid removal process and particle orientation may occur at the same time [14, 25, 26]. Figure 1 shows the schematic diagram of centrifugal slip casting equipment for the functionally gradient composites used in our study.

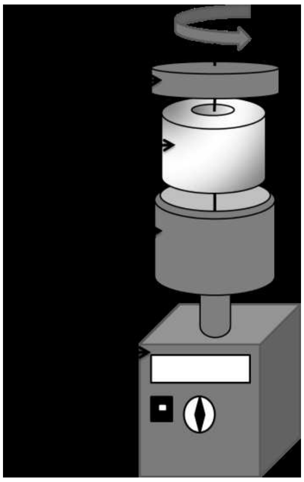

Fig. 1. Diagram of CSC equipment for fabricating Al_2O_3/Ni composites

Rys. 1. Schemat urządzenia CSC do wytwarzania kompozytów Al_2O_3/Ni

A commercially available α-Al_2O_3 powder (TM-DAR, Taimei Chemicals) of an average particle size of 133 nm and density of 3.96 g/cm^3, and Ni powder (Alfa Aesar) of an average particle size of 27 μm and density of 8.9 g/cm^3 were used as the starting materials. Particle size measurements were conducted by a Laser Particle Size Analyzer LA-960. The Al_2O_3 and Ni are highly pure powders (99.99%). In Figure 2, SEM micrographs of the starting powders are shown. The microstructure of the starting powders reveal that they have the tendency to agglomerate. Citric acid (≥ 99.5% Sigma-Aldrich) and diammonium hydrocitrate (puriss, POCh) were used as the dispersant in the composite slurries. Ceramic aqueous slurries were prepared with a solid content of 50 with 10 vol.% (Series I) or 20 vol.% (Series II) nickel particles with respect to the total solid volume. The sedimentation characteristics of the prepared suspensions were monitored in 20 ml tubes and the settling height in time was recorded up to 48 h. The scheme of the composite preparation process is shown in Fig. 3. The samples after sintering were 40 mm in length and the outer diameter was 20 mm with a wall thickness of ~16.5 mm. Figure 4 shows the schematic diagram of typical samples obtained by centrifugal slip casting.

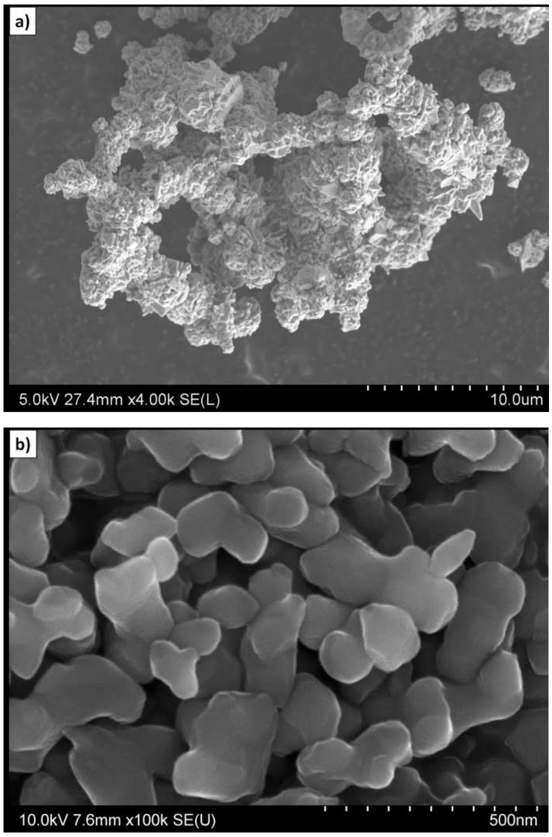

Fig. 2. Morphology of starting powders: a) nickel, b) alumina

Rys. 2. Morfologia proszków wyjściowych: a) niklu, b) tlenku glinu

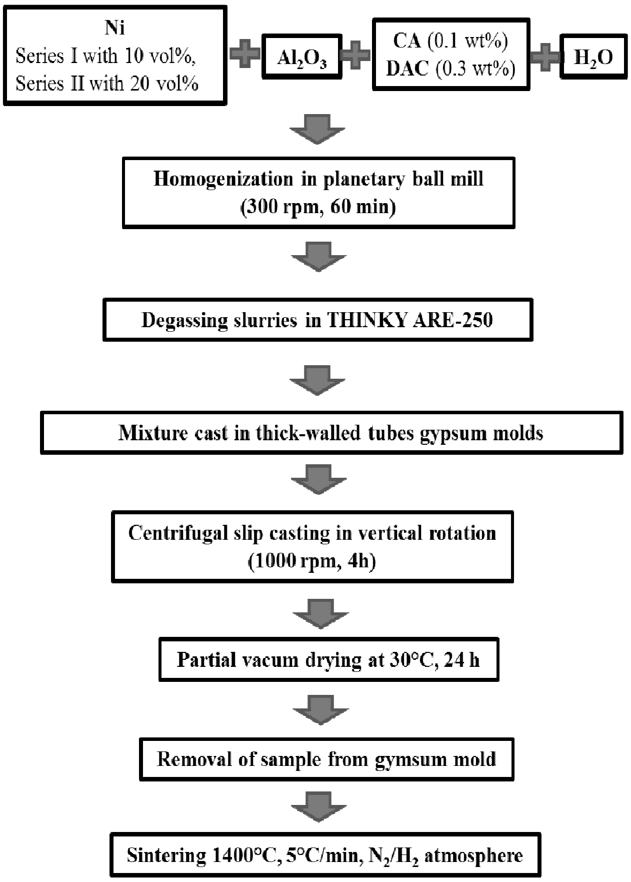

Fig. 3. Scheme of Al_2O_3-Ni composite fabrication

Rys. 3. Schemat wytwarzania kompozytów Al_2O_3-Ni

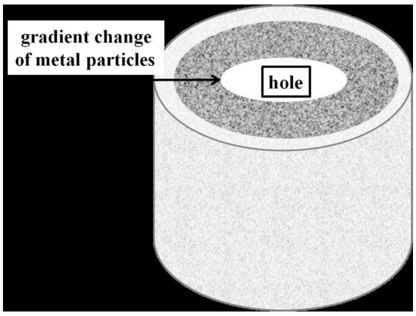

Fig. 4. Diagram of FGM composites obtained by centrifugal slip casting

Rys. 4. Schemat próbki kompozytu z gradientem stężenia cząstek uzyskanych metodą CSC

The phase composition of the sintered samples was examined by X-ray diffraction. XRD analyses were conducted on composite cross-sections with a Rigaku MiniFlex II diffractometer using CuK_{α} radiation (1.54178 Å), in the range 10° to 100° 2θ, with a step size of 1° 2θ and an acquisition time per step of 1 min.

The microstructure of the sample cross-sections after sintering was examined by SEM (HITACHI SU-70). The chemical compositions in microareas from the cross-sections were determined by EDS (HITACHI S-3500N).

RESULTS AND DISCUSSION

Sedimentation experiments were carried out to assess the effect of metal addition on the stability and settling of particles in the slurry. The results are presented in Figure 5.

Fig. 5. Sedimentation behaviour of suspension after 48 h: a) Al_2O_3 + 10 vol.% Ni, b) Al_2O_3 + 20 vol.% Ni

Rys. 5. Proces sedymentacji zawiesiny po 48 godzinach: a) Al_2O_3 + 10% obj. of Ni, b) Al_2O_3 + 20% obj. of Ni

For single spherical particles in a liquid, the sedimentation rate can be calculated using Stokes law. In the case of an Al_2O_3-Ni system, the density of alumina is much less than the density of nickel. As a result, Ni would likely segregate from the Al_2O_3 powder due to differential sedimentation rates driven by the difference in density. However, for slurries with a high solid content, the motion of the particles is hindered by interaction between the particles in the liquid medium. In our research, we are not dealing with the movement of individual particles but their agglomerates. For both slips, moving the metallic particles is accompanied by movement of ceramic particles. For that reason, after 48 hours the slurry with 10 vol.% Ni (Fig. 5a) is divided into two parts: composite (dark) and alumina (light). When increasing the amount of nickel in the slurry to 20 vol.%, sedimentation did not occur. Presumably, the entire ceramic phase has been linked to the nickel particle. In the slurry with the higher vol.% of Ni, the distance between the agglomerates was smaller, and the particles were intensively blocked by each other thus preventing their movement. The use of centrifugal force can significantly speed up the process of producing composites with gradients.

The XRD analysis of the Series I and Series II samples after sintering showed the presence of Ni and Al_2O_3 as the only crystalline phases. The presence of an alumina nickel spinel phase ($NiAl_2O_4$), sometimes observed in such materials [27], was not detected in our investigations. The application of a reductive atmosphere (H_2/N_2) during sintering prevented the formation of $NiAl_2O_4$.

Typical SEM microphotographs of FGM samples for Series I and Series II are shown in Figure 6. The light spots dispersed in the grey matrix were identified as Ni particles. In each series both zones were observed, with different metal particle contents. It can be clearly seen that zone A is composed almost entirely of alumina. The whole volume of the Ni particles was situated in the second as a result of the centrifugal process. It was found that the distribution of nickel particles in zone B in Series II was more homogeneous than in Series I. Furthermore, the width of the zones were different in both series. In the case of Series I, the width of zone A was 0.44 mm, while the width of zone B was 2.75 mm and Series II demonstrated a 0.14 mm width of zone A while zone B was 3.40 mm, respectively. No sedimentation of Ni particles in the slurry was probably responsible for the more homogeneous distribution of them and the narrower width of zone A. The use of centrifugal force during the casting of slurries with different amounts of nickel caused a similar movement of particles to that during gravitational sedimentation. The applied centrifugation speed was too low to increase the velocity of the particles in the suspension and prevented achieving a more pronounced gradient.

Furthermore, by using the EDS technique, successful property gradation was confirmed. The research consists in finding the chemical composition in several (four) regions along the gradation direction. The areas used for making the EDS measurement are shown in Figure 7. As illustrated by Table 1, the chemical composition gradual changes from alumina with oxygen to nickel in each series. The lower nickel content in area 1 is due to the capillary force of the mold and deposition of a casting slip on the wall of mold before the process started.

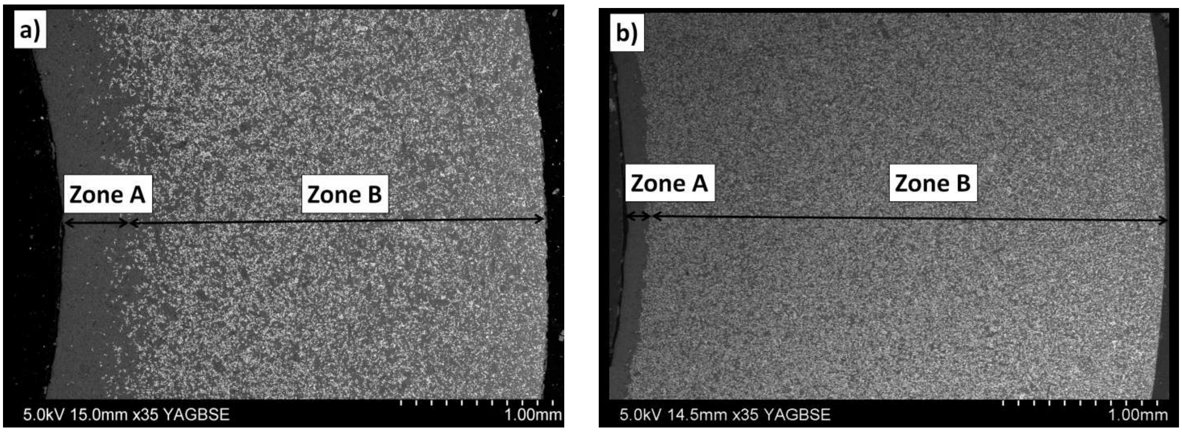

Fig. 6. SEM micrograph of composite microstructures: a) Al_2O_3-Ni with 10 vol.% metallic phase, b) Al_2O_3-Ni with 20 vol.% metallic phase

Rys. 6. Zdjęcia SEM mikrostruktur kompozytów: a) Al_2O_3-Ni z 10% obj. zawartością fazy metalicznej, b) Al_2O_3-Ni z 20% obj. zawartością fazy metalicznej

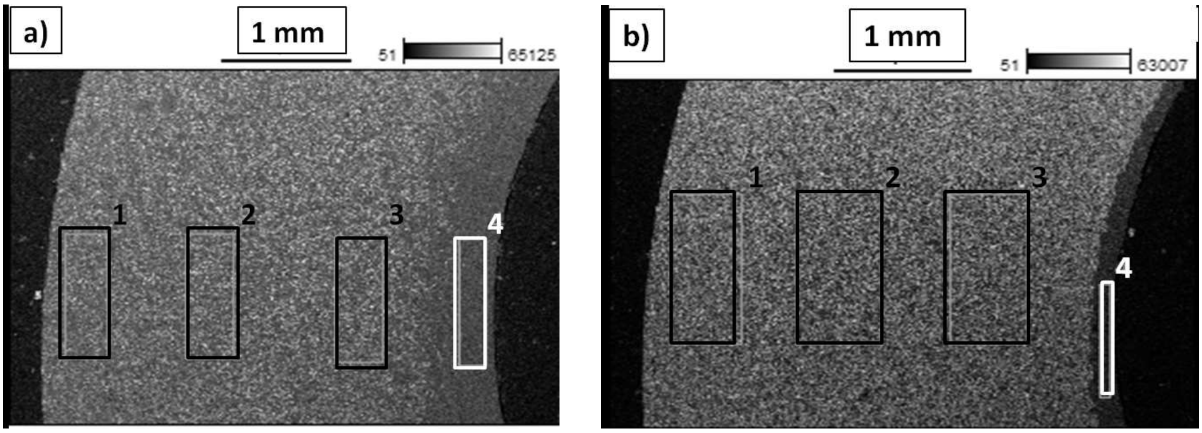

Fig. 7. EDS for samples: a) Al_2O_3-Ni with 10 vol.% metallic phase b) Al_2O_3-Ni with 20 vol.% metallic phase

Rys. 7. Analiza EDS składu chemicznego próbek: a) Al_2O_3-Ni z 10% obj. zawartością fazy metalicznej b) Al_2O_3-Ni z 20% obj. zawartością fazy metalicznej

TABLE 1. **Weight, atomic and volume [%] content of samples from different areas**
TABELA 1. **Zawartość masowa, atomowa i objętościowa [%] próbek w różnych obszarach**

Al_2O_3-Ni composites							
Series I with 10 vol.% Ni (Fig. 6a)							
	Weight %			Atom %			Volume %
	O	Al	Ni	O	Al	Ni	Ni
area 1	26.61 ±0.87	52.60 ±0.77	20.79 ±2.86	41.93 ±1.37	49.15 ±0.72	8.93 ±1.23	11
area 2	25.22 ±0.76	50.47 ±0.70	24.31 ±2.67	48.45 ±1.23	48.45 ±0.68	10.73 ±1.18	13
area 3	23.67 ±0.73	50.07 ±0.67	26.26 ±2.53	39.11 ±1.20	49.07 ±0.65	11.83 ±1.14	14
area 4	36.63 ±0.98	63.37 ±0.80	0.00 ±0.00	49.36 ±1.32	50.64 ±0.64	0.00 ±0.00	0
Series II with 20 vol.% Ni (Fig. 6b)							
	Weight %			Atom %			Volume %
	O	Al	Ni	O	Al	Ni	Ni
area 1	18.67 ±0.76	45.99 ±0.73	35.34 ±3.07	33.59 ±1.37	49.08 ±0.78	17.33 ±1.51	19
area 2	18.62 ±0.69	41.93 ±0.65	39.46 ±3.14	34.33 ±1.27	45.84 ±0.72	19.83 ±1.58	22
area 3	18.56 ±0.66	42.22 0.62	39.22 ±2.95	34.19 ±1.21	46.12 ±0.68	19.69 ±1.48	22
area 4	34.59 ± 0.99	62.77 ± 0.83	2.64 ± 0.95	47.69 ± 1.37	51.32 ± 0.68	0.99 ± 0.36	1

SUMMARY AND CONCLUSIONS

Al_2O_3-Ni graded hollow cylinders were effectively fabricated by the centrifugal slip casting technique. No additional phases were present when sintering was carried out in reducing atmosphere. SEM micrographs showed that there was a uniform gradient of Ni from the Al_2O_3 rich region in an outwards direction. In both series both zones were observed. The width of the zones and distribution of Ni particles inside the zone depend on the vol.% content of metal particles in the slurry. The higher content of Ni particles in the slurry resulted in no sedimentation which gave the narrow width of zone A where Al_2O_3 with a 2.64 wt.% of Ni was found. Moreover, in this case zone B was characterized by a more homogeneous distribution of Ni particles. For Series I where the Ni particles sedimented in the slurry and the region, only Al_2O_3 was distinguished in the composite and zone A with 100 wt.% alumina was obtained. All the Ni particles were distributed inside zone B.

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