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Structural and mechanical properties of graded composite Al_2O_3/Ni obtained from slurry of different solid content

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Abstract

In this work, an alumina-nickel graded hollow cylinders were prepared by the centrifugal slip casting. In the paper, the results for samples formed from slurries with different solid content: 45 vol.%, 40 vol.% and 35 vol.% are presented. The structure of the samples after sintering was examined by X-ray diffraction (XRD). The microstructure of the composite, especially the nickel particle size distributions were investigated by using scanning electron microscopy (SEM). An image analyzer has been used for the measurement of volume fraction of the nickel particles in the composites. The hardness was measured by using a Vickers hardness-testing. Based on hardness measurements K_{IC} value were determined. The XRD results confirmed only two phases: Ni and α-Al_2O_3 in all samples. The preliminary macroscopic observation as well as SEM showed, that the microstructure of the sample cross-section is not homogeneous. Microstructural characterization revealed the gradation of nickel content along the radial direction of hollow cylinder. Three zones were distinguished, from outer surface towards the inner side of the tube. The maximum of volume fraction of nickel particles was obtained at the middle zone of the composites. The results of hardness-testing revealed that the maximum hardness values were observed in region at the inner edge of the casting due to an absence of nickel particles.

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Keywords: Functionally graded materials (FGM), Ceramic-Metal Composites, Microstructure, Microhardness

1. Introduction

The demand for the Functionally Graded Materials (FGMs) have increased significantly in recent years due to their unique properties, which are not achieved by the conventional materials. FGM consisting of two or more phases, in

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which properties change at least in the one spatial direction (Miyamoto 1999; Ogawa 2006; Hirai 1996; Suresh 1998; Neubrand 1997; Tomsia 1998; Mortensen 1995). These materials have successfully been introduced in many applications field due to their unique physical and mechanical properties (Suresh et al. 1998).
Example of the FGMs are Al_2O_3-Ni composites with a gradient of Ni concentration obtained by the centrifugal slip casting (Zygmuntowicz et al. 2015). Own experiments and research showed that the tree zones with different Ni particles concentration can be distinguished (Zygmuntowicz et al. 2015). Depending on the content of the metal particle in the zone, the composites have a different microstructure, and hence different mechanical properties. The zones containing metal particles are characterized by increased fracture toughness compared to the zones with lower content of metal particles (Moya 2007; Konopka 2001; Konopka 2003; Diaz 2000; Sun 1996).

The metal particles in a ceramic matrix can disperse the energy of propagating cracks (Yeomans, 2008). In the case, when the metallic and ceramic phases are firmly connected together, important role in increasing the fracture toughness has mechanism of bridging an advancing cracks by the ductile particles (Konopka 2003; Yeomans 2008). The zones composed almost entirely of ceramic are characterized by higher brittleness as compared to areas containing the metal particles, although these zones have a higher hardness.

The width of each zone can be controlled through the selection of technological parameters such as: solids content, the amount of the metallic phase, speed of the mixing or the size of used powders (Zygmuntowicz et al. 2015).

The purpose of the present work is to prepare Al_2O_3-Ni functionally gradient materials from the slurry of different solid content (35 vol.%, 40 vol.%, 45 vol.%). The dependency between the microstructures and mechanical properties (the fracture toughness and hardness) of the composites are discussed.

2. Experimental procedure

Functionally gradient composites with 45 vol.%, 40 vol.% and 35 vol.% content of solid phase in an aqueous based slurries containing alumina and nickel powder (10 vol.% with respect to total volume) were tested. The following commercially available powders were used for this work: Al_2O_3 TM-DAR from Taimei Chemicals of an average particle size 133 nm and density 396000 kg/m^3, and Ni powder from Alfa Aesar of an average particle size 27 μm and density 89000 kg/m^3. Citric acid (≥ 99.5% Sigma-Aldrich) and diammonium hydrocitrate (puriss, POCh) were applied as dispersant in the composite slurries.

Composites were fabricated by centrifugal slip casting method. Fig. 1. and Fig. 2. show the schematic diagram of used vertical centrifugal casting equipment and the summarized processing route applied in this work for the fabrication of graded hollow cylinders. Ceramic suspensions were prepared by mixing of powders with deflocculates and water as a solvent in a planetary ball mill PM100 (Retsch) for 1 hours with a speed of 300 rpm. The slurry were poured in a gypsum mold with the inner diameter of 0.02 m. The cylindrical mold was centrifuged along the radial direction with a speed of 1000 rpm for 4 hours. After centrifugation, the sample together with a gypsum mold was removed from metal mold and the gypsum mold with sample was dried in vertical position inside vacuum chamber at room temperature for 24 hours. The dried and shrunk sample could be removed from the gypsum mold easily. The sample was sintered at 1400°C in an atmosphere of H_2/N_2 with H_2 of 20 vol. % and rest N_2. The heating and cooling rate was taken as 1°C/min through sintering process. The dimensions of the fabricated tubes after sintering were 0.04 m length and 0.020 m outer radius with a wall thickness falling between 0.016 and 0.018 m. The sintered functionally graded hollow cylinder composites are show in the Fig. 3.

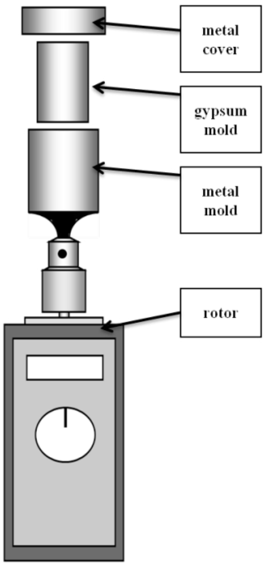

Fig. 1. Schematic diagram of vertical centrifugal slip casting equipment for preparation functionally gradient composites.

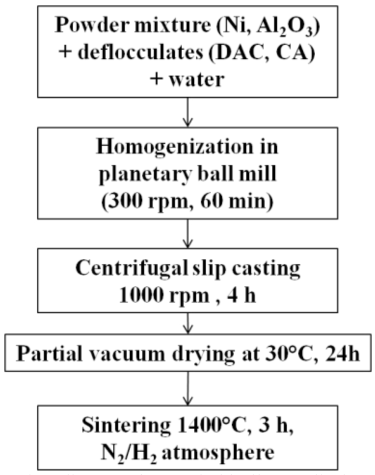

Fig. 2. Processing diagram for graded hollow cylinder preparation.

Fig. 3. Typical functionally graded hollow cylinder composites.

The microstructures of the sintered samples were observed on the cross-sectioned surfaces using a scanning electron microscope (HITACHI SU-70). The cross-section were prepared by cutting the samples parallel the axial direction with diamond saw. The polished cross-section were prepared by grinding and polishing with diamond paste up to 1 µm. The XRD study was performed to determine the bulk crystalline phases of composites. It was conducted using a Rigaku MiniFlex II diffractometer with $CuK_{\alpha 1.54}$ (λ = 1.54178 Å). The spectra were scanned at a rate of 1 deg/min in the range 2θ = 10 - 100°. The analyses were done at the cross-section of composites.

Quantitative description of the microstructure of the FGM was made on the basis of electron micrographs of randomly selected areas on the cross-section using computer image analyzer (Michalski et al., 2005). The object of the study was to use stereological analysis to determine the volume fraction of nickel particles in the composites. Vickers hardness was measured in the polished surface along radial direction with the given load of 9.8 N in equal interval of distance (330 μm) to confirm the compositional changes. Based on measurements length of cracks propagated from the corner of the hardness indentation,the fracture toughness of the material (K_{IC}) were determined. In this study, a Vickers hardness indenter was applied to propagate what are called median cracks on the surface. The K_{IC} values in this case can be estimated using the equation (Niihara, 1983):

$$K_{IC} = 0.067 \cdot \left(\frac{E}{H_V}\right)^{0.4} \cdot \left(\frac{c}{a}\right)^{-1.5} \cdot H_V \cdot \sqrt{a} \quad (1)$$

where E - Young's modulus; H_V - Vickers hardness, c - crack length [μm], a - one half of the indent diagonal length [μm].

3. Resultants and discussion

X-ray diffraction patterns showed no other reflections than those of nickel and alumina at the all samples (35 vol.%, 40 vol.%, 45 vol.%). Fig. 4. presents an exemplary diffraction pattern for samples with 45 vol.% content of solid phase. Application of reductive atmosphere during sintering led to avoid the formation of alumina nickel spinel phase ($NiAl_2O_4$), which may affect the properties of the composite.

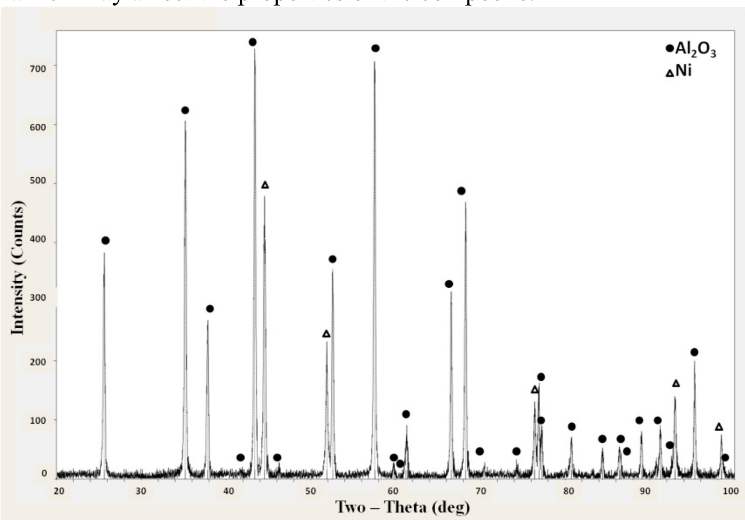

Fig. 4. X-ray phase analysis from samples with 45 vol.% content of solid phase.

The microstructure of three functionally graded materials, obtained with solid phase concentration 35 vol.%, 40 vol.% and 45 vol.% are show in Fig. 5. In this micrographs the bright area is Ni and the grey area is Al_2O_3.

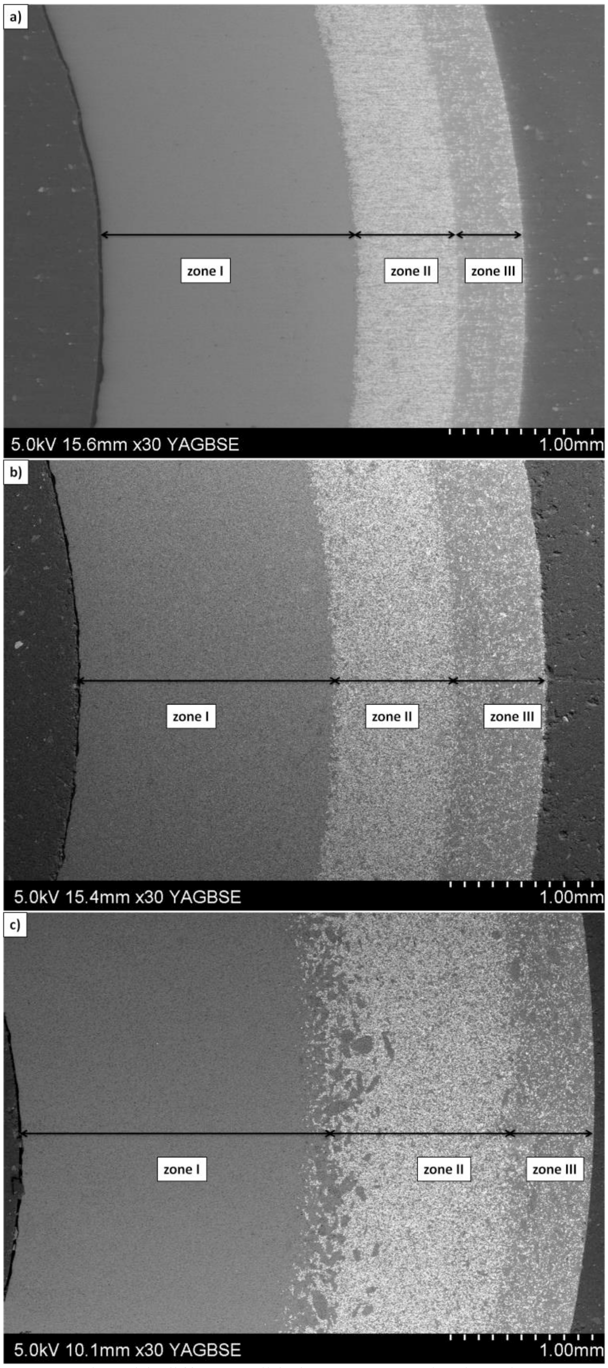

Fig. 5. SEM microphotographs of FGM samples with different solid phase concentration in a slurry: (a) 35 vol.% (b) 40 vol.%, (c) 45 vol.%.

It was found that the samples are characterized by a similar structure. The differences in the widths of the zones in each sample were shown in Fig. 6. Microscopic observation reveal that the each composites with different solid content have the three zones, changing of the Ni particles at the cross-section. The observation of graded hollow cylinders reveal that with the increase in solid content was increased the width of the zones in each the sample. It was found that the sample containing 45 vol.% of the solid phase was characterized by the widest zones in relation to other composites. In addition, it was observed that the zone boundaries in this sample (45 vol.%) are not sharp, however gradually connected to one another.

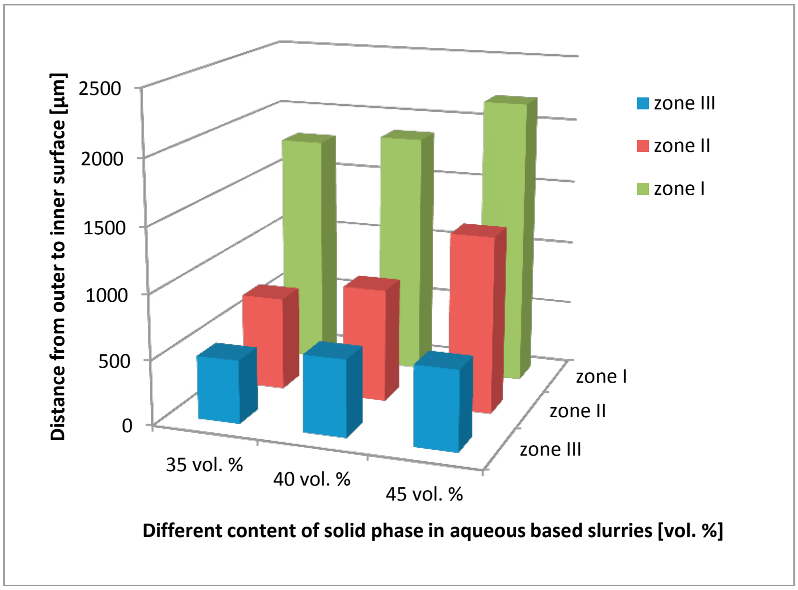

Fig. 6. The differences in the widths of the zones of samples with different solid content (35 vol.%, 40 vol.% and 45 vol.%).

An image analysis equipped in the Micrometer program (Michalski et al., 2005) has been used for measurement of volume fraction of nickel particles in the graded region in the composites. The results are presented in Table 1.

Table 1. Changes in Al_2O_3-Ni composites of relative Ni particles content from outer zone to inner.

relative Ni content [%]			
	zone I	**zone II**	**zone III**
35 vol.%	0	36	11
40 vol.%	0	34	10
45 vol.%	0	27	10

The first zone in all cases was composed of alumina oxide. For the second zone nickel content is highest for sample formed from the lowest solid content (35 vol.%) and was 36 %. With the increase solid content share of nickel in these zone decreases to 34 % and 27 % for samples 40 vol.% and 45 vol.% respectively. This is due to the distance between the particles in the suspension. The greater part of the solvent in the slurry , the distances are larger and the movement of particles with higher density (Ni) is facilitated. This region of FGM was produced as a result of centrifugal acceleration. In zone III relative nickel content is at similar level, about 10%. This part of composites was a result of removing fluid through capillary action in the gypsum mold.

The Vickers hardness measurements were realized on the polished surface from the outer to the inner direction. The results of Vickers hardness reveal that a hardness profile for each samples have similar curve (Fig. 7). In each composites in zone I where is an absence of nickel particles the Vickers hardness has maximum values. In the area near to the inner edge of the zone I in all samples the hardness value amounted of range of 1910 - 2180 HV, and then were decreased to value 652 - 789 HV. The decrease in the hardness values is the result of increases in the increase content of Ni particles in the centrifuged hollow composites. In the zone II are the maximum amount of nickel particles in all composites and this correspond to the lowest value of hardness of in the range of 633 - 842 HV. In graph, the area between zone II and III slightly higher hardness is observed, which are due to the decrease nickel particles in the samples. In region III corresponds to the hardness value of in the range of 1200 - 1550 HV for all composites.

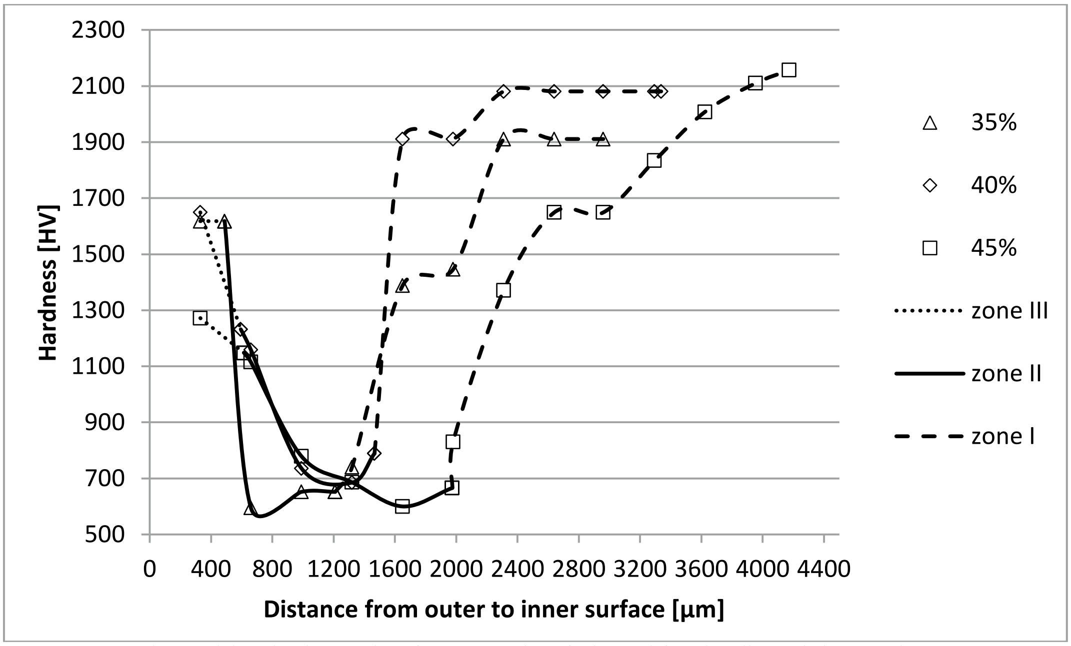

Fig. 7. Vickers hardness values from outer edge of Al_2O_3-Ni functionally graded composites.

Table 2. shows the fracture toughness obtained by the indentation fracture method for the composite fabricate with different solid content. For the all samples the K_{IC} has been calculated for the three distinguished zones described above. The area consisting of 100% Al_2O_3 (zone I) for each composite were characterized by the K_{IC} in the range 5.4 - 5.9 MPa*m$^{1/2}$ with an error ratio measurement 0.3 - 0.5 MPa*m$^{1/2}$. The resulting value is high for samples fabricated from submicron Al_2O_3. For example Li in his work with the use of the same alumina powder compacted by high speed centrifugal casting received a K_{IC} = 4.8 MPa*m$^{1/2}$ (Li et al., 2003). High value obtained for alumina in our work may be related to high green density of composites which resulted in high relative density after sintering. In case of zone II fracture toughness depends on the content of the metal phase and reaches the maximum value about 8 MPa*m$^{1/2}$ for a sample prepared with 35 vol.% solid content in a slurry. Improving the resistance to cracking is related to crack deflection by large volume of ductile nickel phase in this area. In zone III the fracture toughness is slightly higher than in zone I.

Table 2. Fracture toughness values of FGM composites with different solid content (35 vol.%, 40 vol.%, 45 vol.%).

fracture toughness values (K_{IC}) [MPa·m^{½}]			
	zone I	**zone II**	**zone III**
35 vol.%	5.72 ± 0.54	7.98 ± 0.44	6.02 ± 0.43
40 vol.%	5.93 ± 0.42	6.88 ± 0.51	5.98 ±0.48
45 vol.%	5.40 ± 0.35	7.06 ± 0.43	5.78 ±0.51

4. Conclusions

Centrifugal slip casting allows to produce the composite material with gradient concentration of the Ni particles.

This investigation confirmed that the change of the width of the zones in the FGM composites can be controlled by the volume fraction of the solid phase in the slurry. The presented results revealed that with the increase of solid content increases the width of the zones. Macroscopic observation showed that the gradation of nickel content from outer surface towards the inner side of hollow cylinder. For samples containing 35 vol.% and 40 vol.% solid phase the evident boundary between zones was observed, opposite to the sample with 45 vol.% of solid phase. For sample with 45 vol.% of solid phase in slurry the small distance between the particles was responsible for the continuous changing between the zones.

The maximum hardness was obtained at the inner edge of the zone I in all samples. The highest value of hardness correspond to the areas where were absence of nickel particles in the FGM materials. The lowest hardness values were noticed in zone II due to the maximum amount of nickel particles in samples.

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