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THE INFLUENCE OF SINTERING TECHNIQUE ON MICROSTRUCTURE AND PROPERTIES OF ZrO_2/Al_2O_3 COMPOSITE

The following paper presents the results of investigations on the microstructure and mechanical properties of sintered composites in the zirconia-alumina system, fabricated by various sintering techniques. The investigations were performed for a particulate composite consisting of two continuous ceramic phases – zirconia (TZP) and alumina (α-Al_2O_3), 50 vol.% each. Two different methods were used to produce the samples: pressureless sintering and the U-FAST technique. The microstructure of the obtained sintered composite samples was evaluated using a scanning electron microscope. In addition, the density of the sintered bodies, their hardness and fracture toughness were investigated to evaluate the mechanical properties. Based on the obtained results of the investigations, the influence of the sintering technique on the microstructure and mechanical properties of the sintered composites was determined.

Keywords: zirconia/alumina composite, sintering, U-FAST technique, microstructure, grain size

WPŁYW TECHNIKI SPIEKANIA NA MIKROSTRUKTURĘ I WŁAŚCIWOŚCI KOMPOZYTU ZrO_2/Al_2O_3

Niniejsza praca prezentuje wyniki badań mikrostruktury oraz właściwości mechanicznych spieków kompozytowych na bazie tlenku cyrkonu i tlenku glinu, otrzymanych różnymi technikami spiekania. Materiał wykorzystany do badań to kompozyt ziarnisty składający się z dwóch ciągłych faz ceramicznych - tlenku cyrkonu (TZP) i tlenku glinu (α-Al_2O_3), po 50% objętościowych każda. Próbki zostały wytworzone za pomocą dwóch różnych metod, tj. spiekania swobodnego oraz techniki U-FAST. Dokonano oceny mikrostruktury otrzymanych spieków kompozytowych za pomocą skaningowego mikroskopu elektronowego. Dodatkowo przeprowadzono badanie ich stopnia zagęszczenia oraz ich twardości i odporności na kruche pękanie w celu określenia właściwości mechanicznych. Na podstawie otrzymanych wyników określono wpływ techniki spiekania na mikrostrukturę spieków kompozytowych, a co za tym idzie na ich właściwości mechaniczne.

Słowa kluczowe: kompozyt tlenek cyrkonu/tlenek glinu, spiekanie swobodne, technika U-FAST, mikrostruktura, wielkość ziarna

INTRODUCTION

Ceramic particulate composites can be designed in two ways: as materials with a dominant content of the matrix phase and isolated inclusions of the second phase or as materials composed of two continuous phases (a duplex structure) [1]. The second of the mentioned composite types is more difficult to densify during sintering due to the limited intensity of diffusion processes caused by the presence of the second phase [2]. Achieving a high level of densification in these composites by pressureless sintering demands a high sintering temperature, which provokes intensive grain size growth [3]. The scale of this phenomenon could be decreased by utilizing a sintering technique assisted by an electrical field (U-FAST) [4].

Composite materials in the zirconia (TZP)-alumina (α-Al_2O_3) system show good mechanical properties (strength, hardness) and very good abrasive wear in different environmental conditions [5, 6]. Additionally, their high value of the Weibull modulus [7], indicates their high reliability and raises interest in using them as versatile structural ceramics in many technical applications.

The presented paper shows the possibility of manufacturing a dense sintered composite in the ZrO_2/Al_2O_3,

system with a volumetric ratio of phases 1:1. It means that both constituent phases are present in the same proportion. The microstructures and properties of two ZrO_2/Al_2O_3 materials were compared: one pressureless sintered and the second one, sintered using the U-FAST (upgraded field assisted sintering technology) technique.

EXPERIMENTAL PROCEDURE

The composite material was composed of commercial powders: zirconia (TZ-3Y, Tosoh) and alumina (TM-DAR, Taimei Chemicals Co. Ltd). The mean grain size of the powders was about 160 nm for alumina and 100 nm for zirconia (according to the producers' data). The volumetric phase content was 50 vol.% of both phases. In the present work this material is described as ZA50.

The composite powder was prepared by intensive, 30-minute mixing of suitable amounts of the constituent zirconia and alumina powders in an attritor mill. Mixing was conducted in ethyl alcohol using zirconia milling media 2 mm in diameter. One part of the powder was shaped by uniaxial pressing (50 MPa) and consequently by isostatic pressing (300 MPa). These samples were pressureless sintered at 1300, 1400, 1500 and 1650°C with a 2-hour soaking time. The rate of temperature increase was 3°C/min. The other part of the composite powder was used to manufacture sintered bodies by the U-FAST technique utilizing GeniCore equipment. Sintering was conducted under a load of 50 MPa at temperatures of 1250, 1300 and 1400°C. The soaking time at the maximum temperature was 2 mins. The sintered samples were cut and polished for the planned experiments.

The apparent density (g/cm^3) of the sintered samples was determined by hydrostatic weighing at 21°C. The relative densities (% of theoretical) were calculated as a ratio of the apparent density and the theoretical one.

The theoretical density was accepted as the mean value of the theoretical density of zirconia (6.10 g/cm^3) and alumina (3.99 g/cm^3). Vickers hardness (*HV*) was measured using a 9.81 N indenter load. The fracture toughness values (K_{Ic}) were determined using the indentation procedure as well. In this case the indenter load was 29.42 N. The calculations were conducted utilizing Niihara's equation, elaborated for the Palmqvist crack model [8]. Microstructure observations were conducted using a Nova Nano SEM 200. Additionally, the mean grain size of the constituent phases was calculated using micrographs of thermally etched microstructures. Calculations were performed using the ImageJ v. 1.35c programme.

RESULTS AND DISCUSSION

The results of some of the physical property measurements are shown in Table 1. For the pressureless sintered materials, the sintering process was not effective at the lowest temperature (1300°C). The highest density was registered for the material densified at the highest applied temperature (1650°C). Applying the U-FAST technique did not assure high densities at the lowest temperatures (1200 and 1300°C) either. However, at the highest sintering temperature (1400°C) the U-FAST process assured a density 98% higher than that of theoretical one.

Regardless of the applied sintering technique, an increase in hardness with a higher temperature of the process can be observed. This phenomenon is more intensive for the U-FAST process. The highest hardness value (17.9 GPa) was achieved for the material sintered by U-FAST at 1400°C.

By analysing the critical stress intensity factor values (K_{Ic}) which describe the fracture toughness of the material, it can be stated that the investigated composites showed a higher fracture toughness than traditional monophase materials [9]. The highest K_{Ic} value (12.9 MPa·m$^{0.5}$) was measured for the material pressureless sintered at 1300°C. The most probable reason was that this material was still distinctly porous. The presence of pores could increase the K_{Ic} value.

TABLE 1. **Properties of tested samples**
TABELA 1. **Zestawienie właściwości badanych próbek**

Sintering conditions		App. dens. [g/cm^3]	Theo. dens. [%]	Mean grain size [nm]		Hardness *HV* [GPa]	K_{Ic} [MPa·m$^{0.5}$]
Technique	Temperature [°C]			Al_2O_3	ZrO_2		
Pressureless sintering	1300	4.03	79.8	250 ±100	180 ±60	7.3 ±0.1	12.9 ±1.0
	1400	4.903	97.2	420 ±120	250 ±100	16.5 ±0.4	9.4 ±0.3
	1500	4.978	98.7	510 ±180	420 ±160	16.6 ±0.2	9.2 ±0.2
	1650	4.982	98.8	770 ±280	550 ±220	16.7 ±0.3	10.5 ±0.2
U-FAST	1250	4.13	82.0	120 ±40	100 ±30	8.7 ±0.2	9.8 ±0.5
	1300	4.62	91.6	140 ±40	110 ±30	13.0 ±0.4	10.0 ±0.3
	1400	4.93	98.4	160 ±70	120 ±50	17.9 ±0.4	9.5 ±0.4
Density theoretical. 5.045 g/cm^3							

The microstructures of the investigated samples are presented in Figures 1-6. M The marker in five of the micrographs indicate 500 nm. Chemical contrast allows easy distinction between the zirconia (light) and alumina (dark) phases. Only in the micrograph of the microstructure of the material pressureless sintered at 1650°C (Fig. 3) does the marker indicate 1 μm due to intensive grain growth at this temperature. Such a high temperature was necessary to assure a high level of densification. Stereological calculations of the mean size of the grains performed for the composites were also collected in Table 1. They indicated that application of the U-FAST technique enabled the grain growth process to be limited. Dense sintered materials after the U-FAST process still have a very fine-grained microstructure (Figs. 4-6).

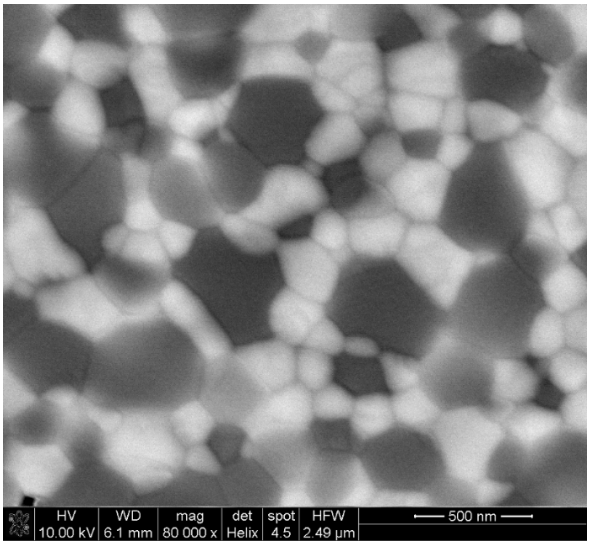

Fig. 1. Microstructure of sample sintered at 1300°C

Rys. 1. Mikrostruktura próbki spiekanej swobodnie w 1300°C

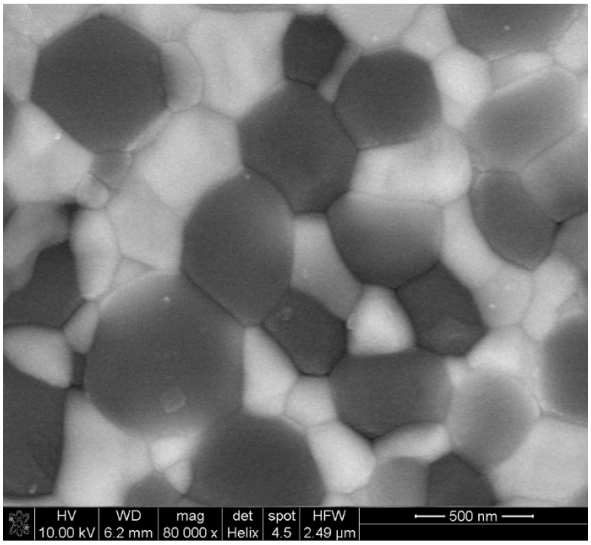

Fig. 2. Microstructure of sample sintered at 1400°C

Rys. 2. Mikrostruktura próbki spiekanej swobodnie w 1400°C

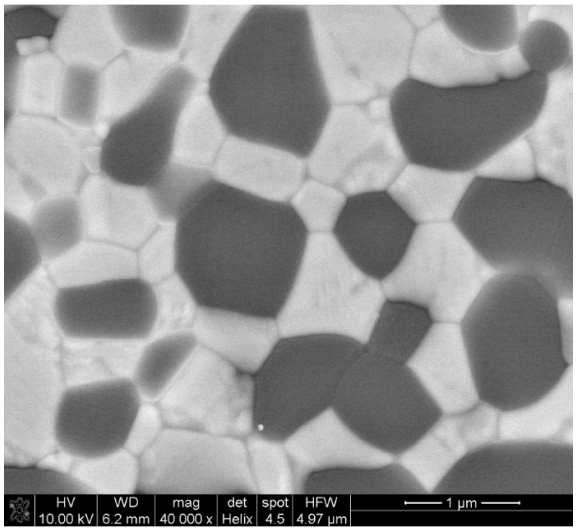

Fig. 3. Microstructure of sample sintered at 1650°C

Rys. 3. Mikrostruktura próbki spiekanej swobodnie w 1650°C

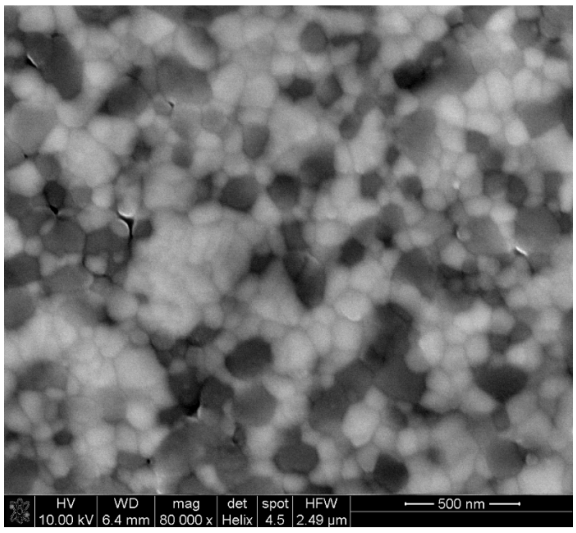

Fig. 4. Microstructure of sample sintered by U-FAST at 1250°C

Rys. 4. Mikrostruktura próbki spiekanej techniką U-FAST w 1250°C

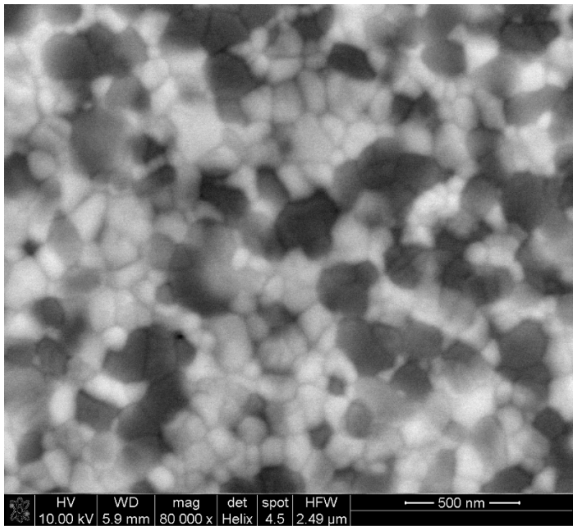

Fig. 5. Microstructure of sample sintered by U-FAST at 1300°C

Rys. 5. Mikrostruktura próbki spiekanej techniką U-FAST w 1300°C

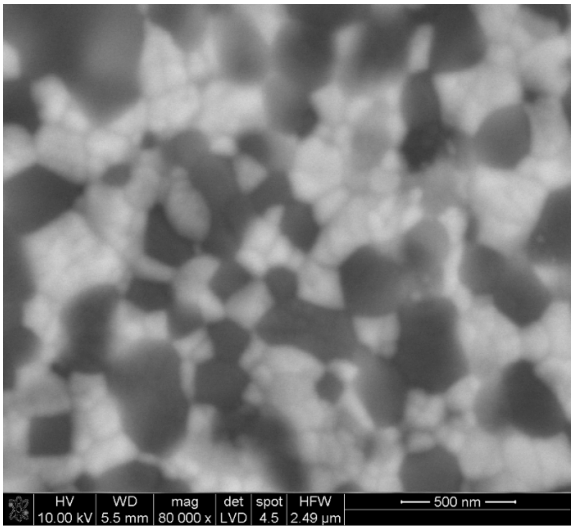

Fig. 6. Microstructure of sample sintered by U-FAST at 1400°C

Rys. 6. Mikrostruktura próbki spiekanej techniką U-FAST w 1400°C

The mean size of the alumina grains after sintering at 1400°C was reduced from ~420 nm (pressureless sintering) to ~160 nm (U-FAST). Similarly, the mean size of the zirconia grains at the mentioned sintering temperature was reduced from ~250 to ~120 nm. It is worth explaining that the measured grain sizes of the U-FAST densified materials were equal to or even slightly smaller than the grain sizes of the raw materials. Most probably, it could be explained by a reduction in the grain sizes of the starting powders during the homogenization (mixing) process, which was very intensively conducted in the attritor mill. The scale of this reduction is unknown, but it must have influenced the final grain size after sintering. In practise, one can observe that after the U-FAST process, the final size of grains (in the sense of its mean value) is the same as in the starting powders.

SUMMARY

- The conducted experiments allowed the conditions of sintering by the U-FAST technique which resulted in dense zirconia/alumina (1:1) composite bodies, to be determined. The level of densification (> 98%) was assumed as a reference level. Such densification is possible in the investigated system utilizing pressureless sintering at 1500°C or at higher temperature conditions.
- The same level of densification using U-FAST equipment demanded the application of the temperature of 1400°C. Under such conditions the grain growth process was significantly limited. Such a temperature did not influence the hardness or fracture toughness of the composites. The values of the HV and K_{Ic} parameters was similar for dense materials regardless of the applied manufacturing technique.

Acknowledgements

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