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Particulate Composites in the Al_2O_3-YAG System

Radosław Lach*, Krzysztof Haberko

AGH University of Science and Technology, Faculty of Materials Science and Ceramics, Kraków, Poland
*e-mail: radek.lach@poczta.fm

Abstract

Contrary to the composites in the Al_2O_3–ZrO_2 system, materials with Al_2O_3 matrix and YAG ($Y_3Al_5O_{12}$) inclusions are much less recognized. YAG is one of the three compounds in the Y_2O_3-Al_2O_3 system of the highest alumina content. It is well known that YAG polycrystals belong to the materials of especially high creep resistance. Since coefficients of thermal expansion of YAG and alumina do not differ essentially from each other, the YAG inclusions in the alumina matrix seem to lead to a material of interesting mechanical properties.

Only a few publications concern the preparation of such composites by powder sintering. In the present work Y_2O_3 was introduced to alumina by yttrium hydroxide precipitated within the alumina powder suspension. By low temperature calcinations a homogenous alumina/yttria mixture was received. Yttria content corresponded to 20 vol.% YAG after the reaction. It was found that at 1500ºC YAG and α-alumina were the only phases present in the system. The investigation of the resulting material microstructure revealed a uniform distribution of sub-micrometer YAG particles. Hardness of the material was similar to that of dense alumina and fracture toughness ($K_{Ic} \approx 6$ MPa·m$^{1/2}$) was close to the observed in the case of the 3-YTZP material.

Keywords: Al_2O_3, YAG, Ceramic matrix composites, Powder preparation, Sintering

KOMPOZYTY ZIARNISTE W UKŁADZIE Al_2O_3-YAG

W przeciwieństwie do kompozytów Al_2O_3-ZrO_2, materiały na bazie tlenku glinu z wtrąceniami YAG ($Y_3Al_5O_{12}$) są słabo rozpoznane w literaturze. YAG (granat itrowo-glinowy) jest fazą o najwyższej zawartości glinu wśród znanych glinianów itru. Jak dobrze wiadomo polikryształy YAG zaliczają się do materiałów o szczególnie wysokiej odporności na pełzanie. Ze względu na niewielką różnicę wartości współczynników rozszerzalności cieplnej granatu itrowo-glinowego i tlenku glinu, materiał zawierający wtrącenia YAG w matrycy Al_2O_3 może się charakteryzować interesującymi właściwościami mechanicznymi.

Jedynie nieliczne doniesienia literaturowe dotyczą preparatyki kompozytów Al_2O_3-YAG drogą spiekania proszków. W prezentowanej pracy prekursor Y_2O_3 został wytrącony w zawiesinie Al_2O_3. Następnie zawiesina została wysuszona, a otrzymany proszek poddano prażeniu, dzięki czemu uzyskano jednorodną mieszaninę ziaren Al_2O_3 i Y_2O_3. Zawartość tlenku itru odpowiadała 20 % obj. YAG po reakcji. Stwierdzono, że α-Al_2O_3 i YAG jako jedyne fazy występują w materiale spiekanym w temperaturze 1500ºC. W mikrostrukturze uzyskanych spieków obserwuje się submikronowe cząstki YAG. Wtrącenia cząstek YAG w osnowie Al_2O_3 podnoszą twardość materiału w porównaniu do spieków czystego tlenku glinu. Natomiast odporność na kruche pękanie ($K_{Ic} \approx 6$ MPa·m$^{1/2}$) jest porównywalna z wartością obserwowaną w przypadku materiału 3-YTZP.

Słowa kluczowe: Al_2O_3, YAG, kompozyty ziarniste, preparatyka proszków, spiekanie

1. Introduction

An unquestionable advantage of the ceramic oxide materials, also for high temperature applications, is constituted by their resistance to oxidation. Alumina is one of the most popular materials of this kind. However, its relatively low mechanical properties, especially fracture toughness, limit its application. Secondary phase inclusions are usually applied to improve these properties. The YAG particle inclusions in the alumina matrix seem to be one of the solutions. Since YAG is the phase of the highest alumina content among other phases in the Al_2O_3–Y_2O_3 system, the reaction between alumina and yttria should result in the system rich in alumina in the YAG inclusion synthesis in the alumina matrix.

Studies on the materials in the alumina-YAG system concern mainly the method of directional crystallization [1-5]. This is an expensive and technically difficult technique due to very high temperatures of the process. Only a few papers concern the alumina-YAG materials prepared by the ceramic method [6-11]. One of the recent papers concerns the triple alumina-zirconia-YAG system [12].

The aim of the present study is to find proper preparation conditions resulting in dense Al_2O_3–YAG composites. Mixtures of submicrometer α-Al_2O_3 and nanometric Y_2O_3 powders were used. It was assumed that the reaction between yttria and alumina should lead to the formation of YAG particle inclusions within the alumina matrix.

2. Experimental

In our investigation α-Al_2O_3 (TM-DAR Taimei Chemicals, Japan) and Y_2O_3 (4N Aldrich) were applied. Yttria was dis-

solved in nitric acid of analytical quality and the solution was added to the alumina powder aqueous suspension whose concentration corresponded to 50 vol.% Al_2O_3. Ammonium carbonate was applied as a precipitation agent. It was added to the vigorously stirred suspension until pH = 8.5 was reached. Such conditions give quantitative yttrium compound precipitation. After the completion of the process, a clear liquid over the sediment was observed. It suggests good homogenization of the system. Yttrium proportion as recalculated to Y_2O_3 corresponded to 20 vol.% YAG after the reaction with alumina.

The suspension was dried at 100ºC, and the resultant powder calcined at 600ºC. A DTA/TG measurement (Derivatograph Q, MOM Hungary) was used find this calcinations temperature. One part of the powder was homogenized by attrition milling for 30 min in the aqueous suspension under pH = 8 brought about by the $(NH_4)_2CO_3$ additive, using 2 mm zirconia (TOSOH) balls. The selection of the suspension pH results from the zeta potential measurements (Zetasizer Nano-Z, Malvern). Another part of the powder was not homogenized. The resulting powders were compacted into discs of 25 mm in diameter and 3 mm thickness with no lubricant additive. Uniaxial compaction under 50 MPa was followed by the isostatic pressing under 300 MPa. Sintering at 1400, 1500 and 1600ºC for 2 h (a heating rate of 5ºC/min) allowed us to prepare samples for material characterization. Hydrostatic weigh was used to determine sample density. X-ray diffraction (CuKα_1 irradiation, X'Pert Pro, Philips PANalytical) showed the phase composition of the systems. The pore size distribution in the powder compacts by Hg-porosimetry (PoreMaster, Quantachrome) and the powder specific surface area using Nova 1200 (Quantachrome) were determined. Vickers hardness and K_{Ic} (Palmquist crack model) [13, 14] were measured using HighTec (Japan) equipment. The loads of 3 and 10 kG were applied, respectively.

3. Results and discussion

A DTA/TG analysis helped us to find proper calcinations temperature of the composite powder precursor. Fig. 1 indicates that no reactions related to the yttrium part of the system occur at temperatures of 600ºC and higher. X-ray diffraction revealed yttria and alumina as the only phases existing in the powder calcined at 600ºC.

The best conditions for homogenization of the bi-phase system correspond to the hetero-floculation state in which particles of the same kind repel each other and the particles of different phases attract each other [15]. Fig. 2 shows the results of the zeta potential measurements for the α-Al_2O_3 and Y_2O_3 powders. The plots indicate that the conditions, which meet the hetero-floculation requirement, occur at pH = 8 in the system under discussion. Such conditions were applied during attrition homogenization of one part of the studied powder (Alumina-20 vol.% YAG/homogenized).

Pore size distributions of the composite powder compacts shown in Fig. 3 indicate that the attrition homogenization leads to the mono-modal distribution and higher densification of the sample, contrary to the compact derived from the powder non-subjected to this process. The possible reason of this behaviour is a formation of agglomerates in the powder during calcination at 600ºC. Presumably the agglomerates

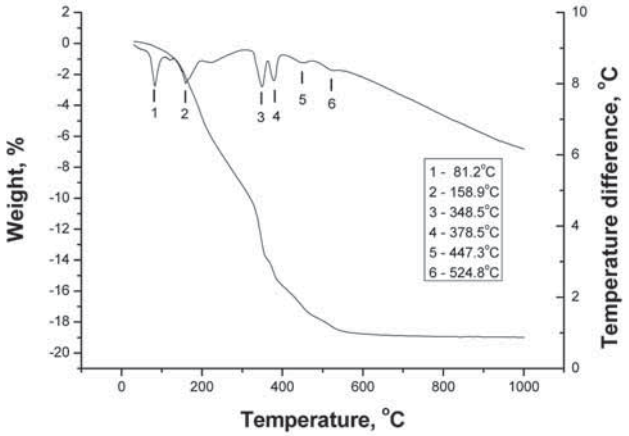

Fig. 1. DTA/TG curves of composite powder with yttrium compound precipitated with $(NH_4)_2CO_3$. Rate of temperature increase in air was equal to 10ºC/min.

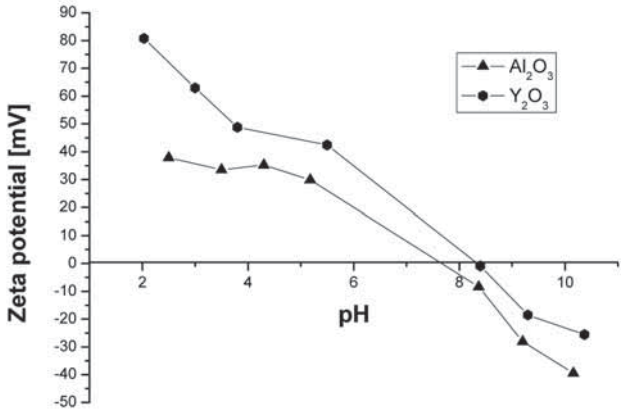

Fig. 2 Zeta potential vs. pH for the alumina and yttria powders.

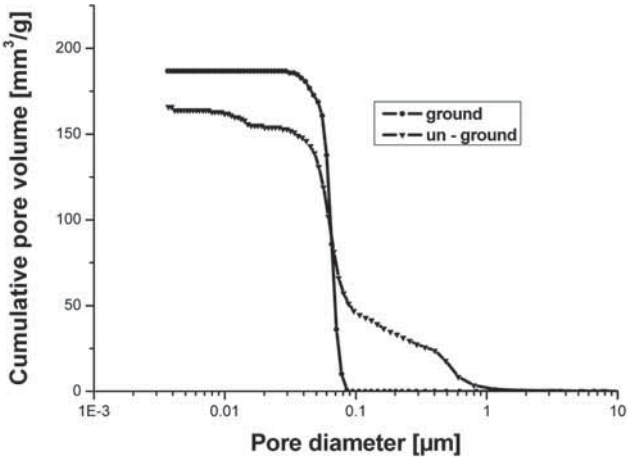

Fig. 3. Cumulative curves of pore size distribution of the compacts originated from the powder calcined at 600ºC as a function of preparation conditions.

are sufficiently hard to withstand the compaction process. Thus smaller pores correspond to the inter-agglomerate porosity and the bigger to the intra-agglomerate space. It is well known that mono-modal and narrow pore size distribution leads to the better conditions of densification during sintering in contrast to the bi-modal pore size distribution [16]. The data of Table 1 illustrate an essential difference in densification of the compacts derived from the attrition homogenized powder and the powder non-subjected to this operation. It

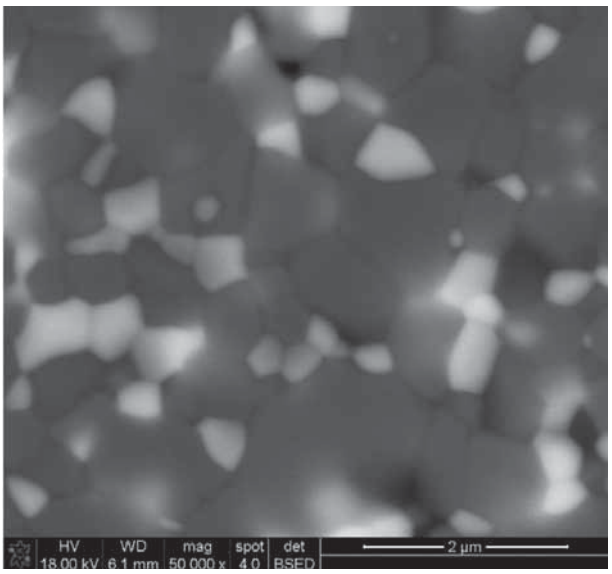

Fig. 4. SEM micrograph of the material derived from the composite powder suspension homogenized at pH = 8 and sintered at 1500ºC.

is clear that the attrition milling essentially improves density of the material. Undoubtedly, this effect can be attributed to the agglomerate destruction by the attrition milling. SEM micrographs corroborate the effect of milling on the material microstructure (Figs. 4 and 5).

X-ray diffraction indicated that materials heat treated at 1500ºC and 1600ºC show α-Al_2O_3 and YAG as the only phases; Fig. 6 provides an example of the typical diffraction pattern. However, the material fired at 1400ºC (not presented in Fig. 6) except of alumina and YAG, showed small amounts of Y_2O_3, YAM ($Y_4Al_2O_9$), and YAP ($YAlO_3$). The related phase composition was as follows: 0.7 wt.% Y_2O_3 15.6 wt.% YAG, 0.2 wt.% YAM, 0.6wt.% YAP, and the rest of α-alumina, as determined by the Rietveld method.

Hardness and fracture toughness of the samples derived from the powder attrition homogenized and sintered at 1500ºC and 1600ºC are shown in Table 2. The data clearly indicate that 20 vol.% YAG particles essentially improve fracture toughness of the alumina material. The values are comparable to the observed in the case of the 3Y-TZP material.

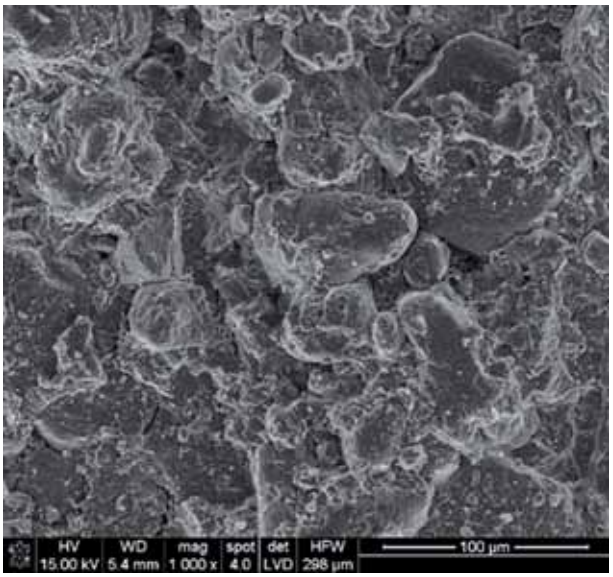

Fig. 5. SEM micrograph of the material derived from the composite powder non-homogenized and sintered at 1500ºC.

Table 1. Relative density [% TD] of alumina and alumina/YAG composite sintered at indicated temperatures.

Material	1500ºC	1600ºC
Al_2O_3	99.30 ± 0.12	99.05 ± 0.11
Al_2O_3-YAG/non-homogenized	88.57 ± 0.09	89.98 ± 0.03
Al_2O_3-YAG /homogenized	97.63 ± 0.05	99.53 ± 0.06

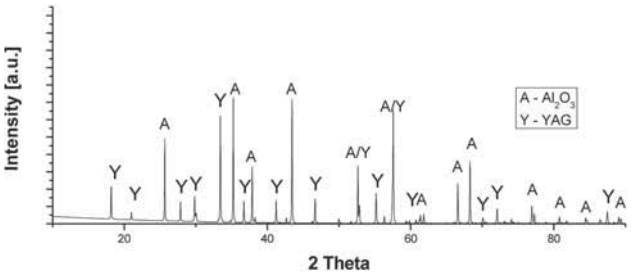

Fig. 6. X-ray diffraction pattern of the sample derived from the powder homogenized at pH = 8 and sintered at 1500ºC.

Table 2. Vickers hardness (HV) and fracture toughness (K_{Ic}) of Al_2O_3-YAG composites and Al_2O_3 materials manufactred at the sintering temperature (T_s) of 1500ºC and 1600ºC; the attrition homogenized Al_2O_3-YAG composite powder was used.

Material	T_s [ºC]	HV [GPa]	K_{Ic} [MPa·m$^{0.5}$]
Al_2O_3	1500	16.96 ± 1.02	4.50 ± 0.27
Al_2O_3-YAG		20.43 ± 1.72	5.97 ± 0.28
Al_2O_3	1600	16.33 ± 0.90	4.37 ± 0.32
Al_2O_3-YAG		20.83 ± 1.51	6.19 ± 0.44

4. Conclusions

The well homogenized alumina-yttria powder can be prepared by precipitation of the yttria precursor within alumina suspension, using ammonium carbonate as the precipitation agent, and calcination at 600ºC followed by attrition milling. The powder results in the compacts characterized by a mono-modal pore size distribution. Such compacts densify better during sintering in comparison to the compacts derived from the powder not subjected to the operation of attrition milling. During heat treatment the reaction between alumina and yttria leads to the synthesis of YAG inclusions evenly distributed in the alumina matrix. The material prepared by this method shows fracture toughness comparable to the 3Y-TZP polycrystals.

Acknowledgments

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