Materials Science & Engineering A 606 (2014) 139–143

Contents lists available at ScienceDirect

Materials Science & Engineering A

journal homepage: www.elsevier.com/locate/msea

Rapid communication

Enhanced mechanical properties of a nanostructured $Mg_2Al_4Si_5O_{18}$–$MgAl_2O_4$ composite

In-Jin Shon [a,*], Hyun-Su Kang [a], Jung-Mann Doh [b], Jin-Kook Yoon [b]

[a] Division of Advanced Materials Engineering and the Research Center of Advanced Materials Development, Engineering College, Chonbuk National University, 561-756, Republic of Korea

[b] Interface Control Research Center, Korea Institute of Science and Technology, PO Box 131, Cheongryang, Seoul 130-650, Republic of Korea

ARTICLE INFO

Article history:
Received 14 February 2014
Received in revised form
24 March 2014
Accepted 25 March 2014
Available online 1 April 2014

Keywords:
Compound
Nanomaterials
Mechanical properties
Microstructure

ABSTRACT

Single-step synthesis and consolidation of nanostructured $Mg_2Al_4Si_5O_{18}$–$MgAl_2O_4$ composite were achieved via pulsed-current-activated heating using a mixture of 3MgO, $3Al_2O_3$ and $5SiO_2$ powders. Before sintering, the powder mixture was high-energy ball milled for 10 h. From the milled powder mixture, a highly dense nanostructured $Mg_2Al_4Si_5O_{18}$–$MgAl_2O_4$ composite could be obtained within one minute by simultaneously applying 80 MPa of pressure and a pulsed current. The advantage of this process is that it allows simultaneous synthesis and densification to near theoretical density while sustaining the nanosized microstructure of raw powders. The mechanical properties (hardness and fracture toughness) of $Mg_2Al_4Si_5O_{18}$ were improved by the addition of $MgAl_2O_4$.

1. Introduction

A magnesium aluminosilicate, with composition $Mg_2Al_4Si_5O_{18}$, offers high thermal stability, low dielectric constant, and good electrical insulating properties, which makes it an attractive material for lining induction furnaces, providing a substrate for electronic devices, and fabricating heat resistant parts of resistance furnaces for the engineering industry [1,2]. The drawbacks that limit the use of this material include its insufficient mechanical properties and density and, accordingly, increased porosity [3,4]. To improve the mechanical properties of these materials, the fabrication of a nanostructured material and composite material [5–8] has been found to be effective. One example of this is the addition of $MgAl_2O_4$ to $Mg_2Al_4Si_5O_{18}$ to improve the latter's properties. The desirable properties of $MgAl_2O_4$ are its high hardness (16 GPa), low density (3.58 g/cm^3), high melting point (2135 °C), high chemical inertness, and high thermal shock resistance [9–11]. Due to its excellent properties, $MgAl_2O_4$ ceramic has been employed mainly in the glass industries, steel industries, etc.

Nanostructured materials have been widely investigated because they display a wide functional diversity of enhanced or different properties compared to bulk materials. Particularly, in the case of nanostructured ceramics, the presence of a large fraction of grain boundaries can lead to unusual or better mechanical, electrical, optical, sensing, magnetic, or biomedical properties [12–17]. Recently, nanocrystalline powders have been produced via high-energy milling [18,19]. The sintering temperature of high-energy, mechanically milled powder is lower than that of unmilled powder due to the increased reactivity, internal and surface energies, and surface area of the milled powder, all of which contribute to its so-called mechanical activation [20–22]. The grain size in sintered materials becomes much larger than that of pre-sintered powders due to rapid grain growth during a conventional sintering process. Therefore, controlling grain growth during sintering is one of the keys to the commercial success of nanostructured materials. In this regard, the pulse current activated sintering method (PCASM), which can make dense materials within 2 min, has been shown to be effective in achieving not only rapid densification to near theoretical density, but also the prohibition of grain growth in nanostructured materials [23–26].

This paper reports on the rapid synthesis and consolidation of dense nanostructured $Mg_2Al_4Si_5O_{18}$–$MgAl_2O_4$ composite starting with high-energy ball-milled nanopowders. The mechanical properties and grain sizes of the resulting nanostructured $Mg_2Al_4Si_5O_{18}$–$MgAl_2O_4$ composites were also evaluated.

2. Experimental procedure

All raw powders were purchased from Alfa, Inc. The average particle sizes and purities of MgO, Al_2O_3, and SiO_2 powders were <45 μm, <2.2 μm, <45 μm and 99%, 99.99%, and 99.8%,

* Corresponding author. Tel.: +82 63 270 2381; fax: +82 63 270 2386.
E-mail address: ijshon@chonbuk.ac.kr (I.-J. Shon).

http://dx.doi.org/10.1016/j.msea.2014.03.095
0921-5093/

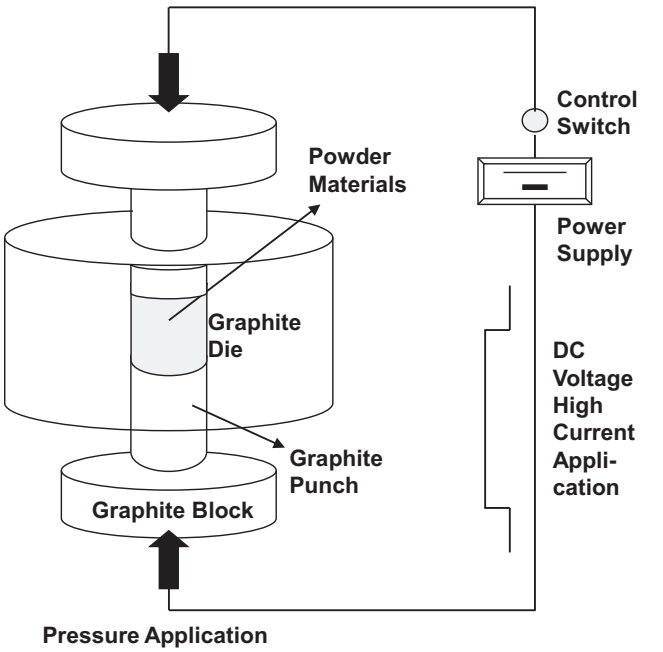

Fig. 1. Schematic diagram of the apparatus for pulsed-current-activated sintering.

respectively. The raw powders ($3Al_2O_3$–$3MgO$–$5SiO_2$) were first milled in a high-energy ball mill (Pulverisette-5 planetary mill) at 250 rpm for 10 h. Tungsten carbide balls (9 mm in diameter) were used in a sealed cylindrical stainless-steel vial under argon atmosphere with a ball-to-powder weight ratio of 30:1.

The powders were placed in a graphite die (outer diameter: 35 mm; inner diameter: 10 mm; height: 40 mm) and then introduced into the pulsed-current-activated sintering (PCAS) apparatus shown schematically in Fig. 1. The four major stages of the synthesis are as follows: evacuation of the system to 40 mtorr (stage 1), application of a uniaxial pressure of 80 MPa (stage 2), activation of a pulsed current (on time, 20 μs; off time, 10 μs), which was maintained until densification was attained as indicated by a linear gauge measuring the shrinkage of the sample (stage 3), and cooling the sample to room temperature (stage 4). Temperatures were measured with a pyrometer focused on the surface of the graphite die. The process was carried out under a vacuum of 40 mtorr (5.3 Pa).

The relative density of the sintered sample was measured using the Archimedes method. Microstructural features were examined after polishing and etching thermally for 1 h at 1000 °C. Compositional and microstructural analyses of the products were conducted via X-ray diffraction (XRD) and field emission scanning electron microscopy (FE-SEM) equipped with energy dispersive spectroscopy (EDS). Vickers hardness measurements were performed on polished sections of the $Mg_2Al_4Si_5O_{18}$–$MgAl_2O_4$ composite using a 5-kg load and a 15-s dwell time.

The grain sizes of the powders and sintered product were calculated from the full width at half-maximum (FWHM) of the diffraction peak using Suryanarayana and Norton's formula [27]:

$$B_r(B_{crystalline}+B_{strain})\cos\theta = k\lambda/L+\eta\sin\theta \quad (1)$$

where B_r is the full width at half-maximum (FWHM) of the diffraction peak after instrumental correction; $B_{crystalline}$ and B_{strain} are the FWHMs caused by small grain size and internal stress, respectively; k is a constant (with a value of 0.9); λ is the wavelength of the X-ray radiation; L and η are the grain size and internal strain, respectively; and θ is the Bragg angle. The parameters B and B_r follow Cauchy's form with the relationship: $B=B_r+B_s$, where B and B_s are the FWHMs of the broadened Bragg peaks and the standard sample's Bragg peaks, respectively.

3. Results and discussion

Fig. 2 shows the X-ray diffraction pattern of the $3MgO$–$5SiO_2$–$3Al_2O_3$ powders after high-energy ball milling for 10 h. Only MgO, SiO_2 and Al_2O_3 peaks were observed, as marked in Fig. 2. Therefore, it is obvious that no chemical reaction occurred between the component powders during milling. Nevertheless, the peaks of the powders are significantly wide suggesting that their crystallized sizes became very fine by milling. The average grain sizes of MgO, SiO_2, and Al_2O_3 measured with Suryanarayana and Grant Norton's formula [27] were about 9, 27, and 49 nm, respectively. The FE-SEM image of $3MgO$–$5SiO_2$–$3Al_2O_3$ powders after milling are shown in Fig. 3. It shows that the mixture powders have round-shaped nanosize-grains with some agglomerations.

The variations in shrinkage displacement and temperature with heating time during the sintering of the high-energy ball-milled $3MgO$–$5SiO_2$–$3Al_2O_3$ powders are shown in Fig. 4. The application of pulsed current resulted in the shrinkage of the compact. As the pulsed current was applied, the shrinkage displacement was nearly constant up to 900 °C, and then abruptly increased. The synthesis and consolidation of $3MgO$–$5SiO_2$–$3Al_2O_3$ mixture were

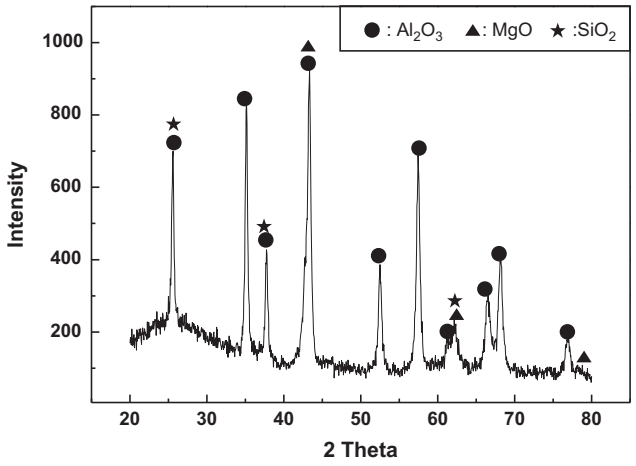

Fig. 2. X-ray diffraction pattern of the powders of Al_2O_3, MgO, and SiO_2 milled for 10 h.

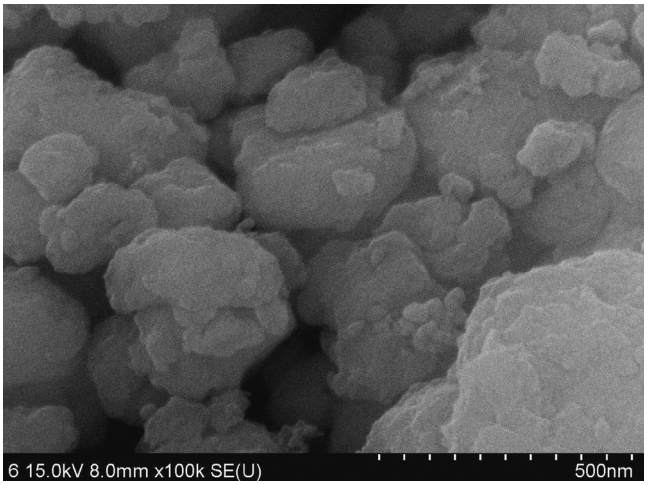

Fig. 3. FE-SEM image of the powders of Al_2O_3, MgO, and SiO_2 milled for 10 h.