Available online at www.sciencedirect.com

ScienceDirect

Ceramics International 41 (2015) 1317–1321

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

Enhanced mechanical properties of WC-reinforced Al_2O_3 ceramics via spark plasma sintering

Wei-Hsio Chen[a], Pramoda K. Nayak[a,b], Hao-Tung Lin[c], Alex C. Lee[a], Jow-Lay Huang[a,d,e,f,*]

[a]Department of Materials Science and Engineering, National Cheng Kung University, Tainan 701, Taiwan
[b]Department of Electrical Engineering, National Tsing Hua University, Hsinchu 30013, Taiwan
[c]Department of Materials Engineering, Kun Shan University, Tainan 710, Taiwan
[d]Center for Micro/Nano Science and Technology, National Cheng Kung University, Tainan 701, Taiwan
[e]Research Center for Energy Technology and Strategy, National Cheng Kung University, Tainan 701, Taiwan
[f]Department of Chemical and Materials Engineering, National University of Kaohsiung, Kaohsiung 811, Taiwan

Received 14 July 2014; received in revised form 13 August 2014; accepted 11 September 2014
Available online 19 September 2014

Abstract

We report the enhanced mechanical properties of WC-reinforced Al_2O_3 nano composites fabricated by spark plasma sintering (SPS). The addition of WC accelerates the sintering process and helps in improving the mechanical properties of Al_2O_3 matrix by hindering its grain growth. The resulting microstructure shows uniformity with a grain size of 300–500 nm. Due to the refined microstructure of composites, the hardness and fracture toughness values are found to increase with the decrease of the Al_2O_3 grain size. In addition, toughening mechanism of this composite also has been discussed in this study.

Keywords: B. Nanocomposite; Spark plasma sintering; Microstructure; Toughening mechanism

1. Introduction

The tungsten carbide (WC) is a good reinforcement material for Al_2O_3 due to its excellent properties including high melting point, good electrical and thermal conductivity, corrosion resistance and, a relatively high chemical and thermal stability [1–3]. Recently, WC–Al_2O_3 composite finds potential applications primarily in cutting-tool industries due to its superior hardness and strength [4]. Several ceramic nanocomposites have been constructed by dispersing second phase or third phase nanoparticles within the micrometer- or nanometer-sized ceramic matrix grains [5–10]. The inclusion of second phase nanoparticles produces a marked improvement in the microstructures and mechanical properties such as strength, hardness, toughness and wear resistance due to the thermal expansion mismatch between the matrix and second-phase particles.

The main controlling factors to prepare a nanosized composite include (i) avoiding agglomeration of initial powders, (ii) to prevent grain growth during sintering. A fine flowability of initial powders is much necessary during the processing of nanocomposites without aggregation [5–8,11]. An effective way to coat nanosized particles uniformly on the matrix is the combination of conventional fluidized bed or spouted bed technology with metal organic chemical vapor deposition (MOCVD) [12–14]. In this context, spark plasma sintering (SPS) is considered to be a suitable technique for the fabrication of ceramic nanocomposites. During SPS process, the initial powders get sintered rapidly by Joule heat and spark plasma generated by high pulsed electric current through the compact. The powder compact could be sintered at a lower temperature and in shorter time compared to conventional sintering to avoid grain growth [15,16].

In the present study, the initial powders of nanocrystalline WC–Al_2O_3 were fabricated by a MOCVD process combined

*Corresponding author at: Department of Materials Science and Engineering, National Cheng Kung University, Tainan 701, Taiwan.
Tel.: +886 6 2754410; fax: +886 6 2754410.
E-mail address: jlh888@mail.ncku.edu.tw (J.-L. Huang).

http://dx.doi.org/10.1016/j.ceramint.2014.09.063
0272-8842/

with spouted bed followed by carburization treatment under (CH_4:H_2=1:9) atmosphere. SPS process was adopted to prepare densified WC–Al_2O_3 nanocomposites. We have tried to investigate the effect of the SPS process on the densification and grain growth of WC–Al_2O_3 nanocomposites. The densification behavior and toughness mechanism of spark plasma sintered WC–Al_2O_3 nanocomposites have been discussed here particularly in detail.

2. Experimental

The α-alumina (α-Al_2O_3, 99.99%, Taimei Chemicals Co. Ltd., Japan) with a mean grain size of 150 nm was used as the matrix powder in this study. Tungsten hexacarbonyl ($W(CO)_6$, 99%, Acros Organics Co., USA) was used as the precursor of tungsten carbide in the MOCVD process. $W(CO)_6$ was initially heated at 90 °C for evaporation and He gas was inserted as the carrier gas for transporting these precursor vapors, into the reaction chamber in a spouted bed. The bed temperature was kept at 300 °C. After the deposition process, the mixture powder was carburized under (CH_4:H_2=1:9) atmosphere at 800 °C for 1 and 5 h to form WC–Al_2O_3 composites, with corresponding specimen designation as C1 and C5, respectively. During the above process, the raw $W(CO)_6$ precursor initially decomposed into an amorphous nanopowder consisting of carbon and WO_3, and finally there was a phase transformation from WO_3 powder to stable WC via intermediate phase W_2C [17]. All the gases such as methane, hydrogen used in this work were 99.9% purity. After carburized treatment, the composite powders were passed through a 200 mesh sieve.

Carburized powders of WC–Al_2O_3 (C1 and C5) were densified by using a SPS process (SPS-515S, Sumitomo, Japan). 2.1 g of powder was put into a graphite mold with a diameter of 15.5 mm. First, the sieved powders were uniaxially pressed at a pressure of 75 MPa for 1 min. A uniaxial pressure of 50 MPa was imposed on the powder and the vacuum was maintained at 6 Pa. The heating rates from room temperature to 600 °C and from 600 °C to 1300 °C were 200 °C/min and 100 °C/min, respectively. The specimens were kept at 1300 °C for10 min holding time followed by furnace cooling to room temperature. In order to understand the influence of nano-WC particles on the densification and microstructure evolution of composites, pure Al_2O_3 was also prepared under the same sintering conditions for comparison.

The phases of the sintered specimens were identified by X-ray diffraction (XRD; Rigaku MultiFlex 2 kW, Japan). The scanning rate was 2°/min and the scanning angles ranged from 20° to 80° with a sampling width of 0.02°. The microstructures and composition of the specimens were characterized by an ultrahigh-resolution scanning electron microscope (UHR-SEM, Carl Zeiss Microscopy, Germany) equipped with an energy-dispersive X-ray spectroscopy (EDS) detector.

All the specimens were smoothly polished using a diamond paste and ultrasonically cleaned. The density of the specimens was measured in water using Archimedes' principle. Hardness tests were carried out using a hardness tester (AVK-C21, Akashi Co., Yokohama, Japan) under an applied 10-kg load with an indentation time of 10 s. The fracture toughness was measured from the length of the cracks using an indentation method. The reported hardness and toughness values are the mean and standard deviation of five measurements.

3. Results and discussion

The pure alumina powder and WC–Al_2O_3 nanocomposite powders (C1 and C5) have been densified by spark plasma sintering at 1300 °C for 10 min. Density, porosity and mechanical properties of these Al_2O_3-based composites are analyzed and listed in Table 1. C1 and C5 composites exhibit higher apparent densities compared to that of monolithic Al_2O_3 due the higher density of tungsten carbide. It can be noted that the theoretical density of tungsten carbide is 15.63 g/cm^3, which is much higher than pure alumina (3.98 g/cm^3). However, the porosity increases a little bit from 0.5% to 0.7% with addition of WC and the reason is that the nanocomposite powders still not reach the full densification at 1300 °C.

The XRD patterns of pure Al_2O_3, C1 and C5 composites are shown in Fig. 1. The diffraction peaks from pure Al_2O_3 is assigned to rhombohedral α-Al_2O_3, and there is absence of any other peak, which shows the purity of the powder. However, besides Al_2O_3, XRD patterns of C1 and C5 contain peaks of other secondary phases including W_2C and W. The decomposition

Table 1
Density, porosity and mechanical properties for Al_2O_3-based composites with and without nano-WC.

	Al_2O_3	C1	C5
Apparent density (g/cm^3)	3.96	4.05	4.21
Apparent porosity (%)	0.55	0.75	0.76
Hardness (GPa)	21.5 ± 0.4	22.7 ± 1.3	22.0 ± 2.0
Fracture toughness (MPa*m$^{1/2}$)	3.39 ± 0.68	4.79 ± 0.42	5.58 ± 1.27

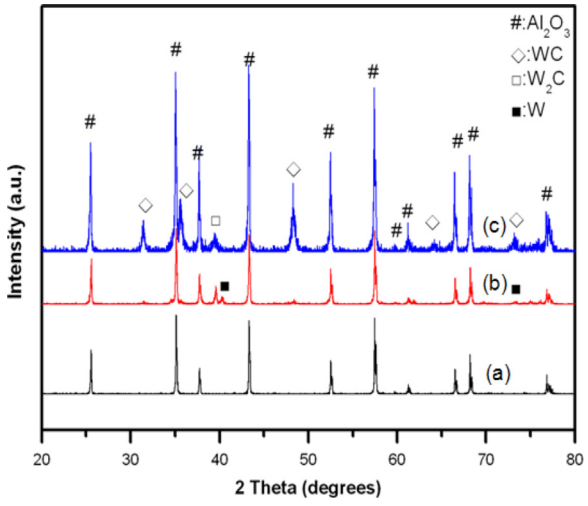

Fig. 1. The X-ray diffraction patterns of (a) Al_2O_3, (b) C1 and (c) C5 specimens sintered via SPS at 1300 °C.