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Letter

Microstructure and mechanical properties of aluminum nitride co-doped with cerium oxide via hot-pressing sintering

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ABSTRACT

The effects of hot-pressing sintering of CeO_2 additive on the densification behaviors and microstructural development of AlN ceramics are investigated. CeO_2 additive can improve the physical and mechanical properties, thermal conductivity, and purification of the grain boundaries. The resulting AlN ceramic exhibits a flexural strength of 387.9 MPa, a fracture toughness of 3.34 MPa $m^{1/2}$, a Vickers' hardness of 12.68 GPa, a thermal conductivity of 168.3 Wm^{-1} K^{-1}, and a relative density of 99.59%.

1. Introduction

Aluminum nitride (AlN) has recently received considerable attention from the metallurgical industry because of its potential application for high power and temperature electronic devices, given its excellent thermal conductivity and chemical stability, low dielectric constant being close to that of silicon, and large band gap [1–3]. However, AlN ceramics are difficult to fabricate and densify because of the presence of highly covalent-bonded wurtzite crystal structures and an oxide (alumina) layer on AlN particle surface. Many methods for synthesizing AlN ceramics have been proposed to reduce sintering temperature and eliminate oxide impurities on AlN surface. AlN ceramics can be obtained through solid-state metathesis reaction [3], powder injection molding [4], pressureless sintering [5], gas pressure sintering [6], two-step sintering [1], and spark plasma sintering [7]. In the fabrication process of AlN ceramics, a sintering additive is typically introduced and liquid-phase sintering is employed to promote the consolidation of AlN ceramics [2]. At present, sintering additives mainly consist of three types, namely, rare-earth oxides, alkaline-earth oxides, and non-oxide additives. Among the additives, Li-salts [8], CaO [9], Y_2O_3 [7], fluorides [10], $CaCN_2$ [11], CaC_2 [12], CaF_2 [2], $Y(NO_3)_3$ [13] are used in AlN ceramics sintering. The concurrent addition of $MgO–Y_2O_3$ [5], and $CaZrO_3–Y_2O_3$ [1] are also used to fabricate AlN ceramics. All sintering additives play a role in decreasing sintering temperature and oxygen content in AlN lattices by forming grain-boundary phases [2].

Hot-pressing sintering technique is the most commonly used method for preparing ceramics. This technique provides an additional axial pressure that aids in decreasing the sintering temperature and promoting the densification of AlN ceramics. Hot-pressing sintering technique is considered a promising method for obtaining appropriate AlN products for industrial usage. AlN-BN ceramic composites are fabricated by hot-pressing sintering [14]. Previous studies have investigated the effects of the method of adding a sintering additive on the mechanical properties of AlN ceramics through an advanced hot-pressed method [13]. However, only a few studies have focused on the mechanical properties of AlN ceramics fabricated through hot-pressing sintering. In addition, sintering aids can be added to attain full densification. CeO_2 has been used successfully used as a sintering additive for Si_3N_4 [15], Ti/Al_2O_3 cermet composite [16], and α-SiC [17]. Thus, the present investigation aims to clarify the effects of CeO_2 additive on the densification behaviors and microstructural development during AlN sintering through hot-pressing sintering technique.

2. Experimental procedure

Commercially available AlN powder (99.99% pure, an average particle size of 1 μm), CeO_2 powder (99.999% pure, an average particle size of 20 μm) were used as starting material. AlN and 2 vol%CeO_2 were weighed in an appropriate proportion and ground together with an agate mortar and pestle. The mixtures were firstly mixed homogeneously in ethanol medium for 24 h using a planetary mill with

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Table 1
Mechanical and thermal properties of sintered samples.

Sample	Flexural strength (MPa)	Vickers' hardness (GPa)	Fracture toughness (MPa $m^{1/2}$)	Thermal conductivity ($Wm^{-1} K^{-1}$)	Relative density (%)
Pure AlN	246.0 ± 18	11.92 ± 0.17	2.67 ± 0.21	134.2	92.40
AlN + 2 vol%Y_2O_3	325.5 ± 16	12.32 ± 0.19	3.12 ± 0.13	146.7	98.72
AlN + 2 vol%CeO_2	387.9 ± 21	12.68 ± 0.11	3.34 ± 0.15	168.3	99.59

agate balls as grind media at the speed of 300 rpm. After milling, the slurry was dried in vacuum at 80 °C for 12 h. Powders obtained were crushed and passed through a 100-mesh sieve. Finally, the composite powders were poured into a graphite mold with a diameter of 45 mm and sintered at 1700 °C under 30 MPa uniaxial pressure for 60 min at a rate of 10 °C/min in a vacuum environment using a vacuum hot-pressing furnace (VVPgr-80-2300, China). For comparison, pure AlN and AlN with 2 vol%Y_2O_3 were fabricated under the same conditions.

Relative density of specimens was determined by the Archimedes' method. The microstructures of the samples were detected by a scanning electron microscopy (SEM) (FEI QUANTA FEG 250, United States). The phase formation of the samples was analyzed by X-ray diffraction (D8-ADVANCE, Germany). The flexural strength was measured by three-point bending method with the specimen size of 4 mm × 5 mm × 60 mm, and the cross-head speed of 0.5 mm/min and a span of 30 mm through electromechanical universal testing machine (CMT5504, MTS SYSTEMS, co., LTD, China). Fracture toughness was measured by the single edge notch beam (SENB) method with the specimen size of 4 mm × 5 mm × 60 mm, and the crosshead speed of 0.05 mm/min and outer support span of 30 mm. Vickers hardness were measured by means of the Vickers indentation method (Model HV-1000IS, Shanghai Jvjing Precision Instrument Manufacturing Co., Ltd.) with five samples each, using a load of 5 kg and a dwell time of 10 s. The thermal conductivity was measured by TC-7000 Laser Flash Thermal Constant Analyzer (ULVAC SINKU-RIKO, Inc.).

3. Results and discussion

Physical and mechanical properties of AlN ceramics with different additives are summarized in Table 1. Density, flexural strength,

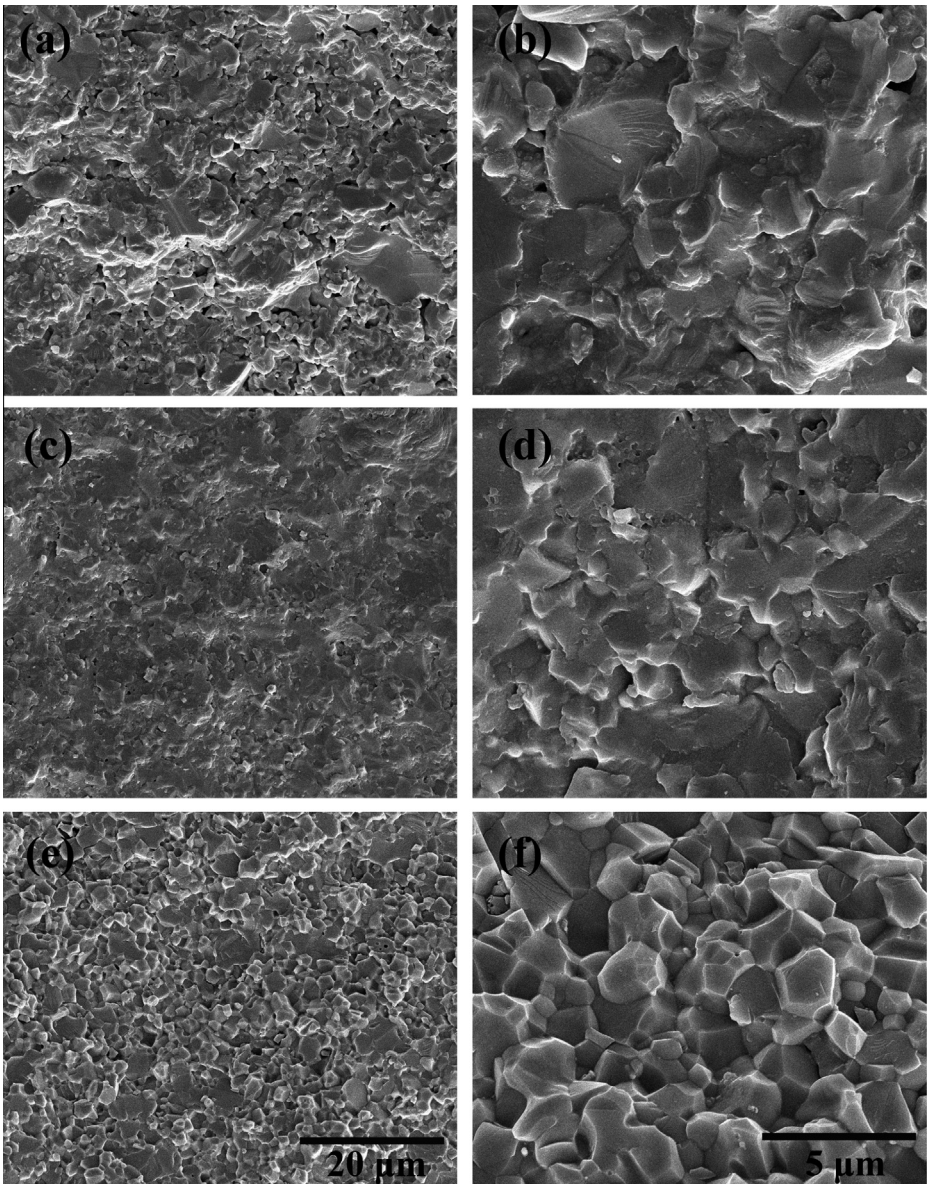

Fig. 1. SEM images of the AlN ceramics with (a, b) none additive and (c, d) 2 vol%Y_2O_3 additive and (e, f) 2 vol%CeO_2 additive, respectively.