Ceramics International xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Ceramics International

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

Synthesis and two-step sintering behavior of solution derived corundum abrasives with plate-like grains

Jiali You, Tao Yang, Dandan Feng, Zhihong Li*

Key Laboratory for Advanced Ceramics and Machining Technology of Ministry of Education, School of Materials Science and Engineering, Tianjin University, Tianjin 300350, China

ARTICLE INFO

Keywords:
A. Corundum
B. Platelets
C. Strength
Abrasives

ABSTRACT

Corundum abrasives with plate-like grains were fabricated by a two-step sintering technique using the solution-based process with the addition of the ternary compound additive Na_3AlF_6-CaO-SiO_2. The two-step sintering method showed obvious advantages over conventional sintering methods in promoting sample densification, suppressing grain growth, and homogenizing the microstructure of the corundum abrasives. The sample doped with 2.5 wt% Na_3AlF_6 and 4 wt% CaO + SiO_2 in the molar ratio of 1:1 possessed a relative density of 99.3%, average grain size of 0.54 μm, and single-particle compressive strength of 49 N. The introduction of seeds reduced the temperature of θ- to α-Al_2O_3 phase transformation. The relationship between the microstructure and the mechanical properties of the abrasives was also discussed.

1. Introduction

Corundum abrasives derived from the solution-based method exhibit superior performance, such as high hardness, high strength, excellent wear resistance, good processing efficiency, and long service life [1–4], during grinding processes. Since the mechanical properties of these abrasives strongly depend on their microstructure, obtaining homogeneous fine-grained corundum abrasives with full densification is of great interest. In fact, a reduced grain size and narrow grain size distribution have been extensively reported to enhance the wear resistance of alumina abrasives [5–9].

However, it is difficult to obtain such refined microstructures through conventional sintering because of the low sinterability of corundum. Two-step sintering (TS) [10] stands out among various densification routes such as pressure-assisted sintering [11,12], microwave-assisted sintering [13], spark plasma sintering [14], and pulsed electric current sintering [15] owing to its simplicity and cost efficiency. In this technique, samples are first heated to a high temperature without holding time at which a sufficiently high relative density (75% or higher) [10] is achieved, and then they are cooled to a lower temperature for a certain period of time. The application of the TS regime can suppress grain growth in the final sintering stage and promote densification, thus successfully fabricating desirable corundum abrasives with improved properties.

Cracks are more likely to be trapped or deflected along randomly oriented platelets, which is greatly beneficial for increasing fracture toughness [6]. Thus, considerable efforts have been made to synthesize plate-like grains. Wu et al. [16] prepared plate-like α-alumina particles with an average size of 40 nm by introducing fine α-alumina seeds and ZnF_2. Zhu and Huang [17] obtained α-alumina platelets by molten salt synthesis and controlled the morphology of α-alumina by adjusting the molten salt species, the salt-to-powder weight ratio, additives, and so on. However, few studies have focused on the synthesis of corundum abrasives with plate-like grains, although such grains increase the fracture toughness, improving the wear resistance of the abrasives [18].

Incorporating additives into alumina is generally believed to promote sintering by generating solid solutions, retarding grain growth, or forming a liquid phase [19–23]. CaO and SiO_2 are typical additives forming a liquid phase during sintering, thus contributing to a decrease in the sintering temperature. In some cases, co-doping with CaO and SiO_2 is considered to trigger abnormal grain growth as a result of the uneven distribution of the liquid phase and the subsequent interfacial energy difference between the basal and non-basal planes [24,25]. In addition, fluorides have proven to be effective in promoting the formation of plate-like α-Al_2O_3 and reducing the transformation temperature of Al_2O_3 from a metastable phase to a thermodynamically stable phase [26–29]. Thus, incorporation of additives is a practical approach to control the morphology of Al_2O_3.

Abbreviations: TS, two-step sintering; CS, conventional sintering; TG-DSC, thermogravimetric differential scanning calorimetry; PEG, polyethylene glycol; SEM, scanning electron microscopy

* Corresponding author.
E-mail address: lizhihongtg@163.com (Z. Li).

https://doi.org/10.1016/j.ceramint.2018.04.059
Received 16 March 2018; Received in revised form 8 April 2018; Accepted 8 April 2018

The aim of this study was to obtain homogenous fully-dense corundum abrasives with plate-like grains by TS sintering at a relatively low temperature. Na_3AlF_6 was utilized for the formation of plate-like grains. The abrasives were co-doped with various amounts of CaO and SiO_2 (1–5 wt%) to obtain a liquid phase during sintering. The combined effect of seeds, the ternary compound additive Na_3AlF_6-CaO-SiO_2, and the TS technique on the sintering behavior and grain morphology of the corundum abrasives was investigated. The correlation between the microstructure and mechanical properties of the abrasives was also discussed.

2. Experimental procedure

2.1. Sample preparation

The corundum abrasives were synthesized by solution-based process using aluminum sulfate octadecahydrate ($Al_2(SO_4)_3{\cdot}18H_2O$) as raw materials. Firstly, an aqueous solution containing 0.4 mol/L Al^{3+} was achieved by dissolving the raw materials into distilled water. PEG (molecular weight 1000) was added as dispersant to prevent the powder from agglomerating. The PH value of the solution was adjusted to 9 by adding diluted $NH_3{\cdot}H_2O$ (1 mol/L) at a rate of 5 ml/min with constant stirring for 2 h. Then the mixture was filtrated from the mother solution and washed for three times with deionized water to remove the residual ions. The obtained precursor was milled with deionized water by adding seeds (3 wt% of α-Al_2O_3 abrasive (grain size < 0.2 μm)) and sintering additives (Na_3AlF_6, CaO and SiO_2) for 10 h. The sintering additives were introduced in the form of trisodium hexafluoroaluminate (Na_3AlF_6) (AnalaR grade, Aladdin Ltd., Shanghai, China), calcium nitrate tetrahydrate ($Ca(NO_3)_2{\cdot}4H_2O$) (AnalaR grade, Kewei Ltd., Tianjin, China) and tetraethylorthosilicate (TEOS) (AnalaR grade, Kewei Ltd., Tianjin, China). The amount of Na_3AlF_6 was fixed at 2.5 wt%. The amount of CaO and SiO_2 ranged from1 to 5 wt% with CaO: SiO_2 molar ratio of 1:1. These additive mentioned above are listed in Table 1. After being dried at 80 °C for 24 h, the precursor was crushed into particles with certain size. The resulting precursor particles were sintered by two different sintering methods as shown in Table 1. In the conventional sintering method (named as CS), samples were sintered at 1400 °C with a holding time of 1 h. In the two-step sintering method (named as TS), samples were first heated to 1400 °C without holding time, and then they were cooled to 1300 °C with a 2-h holding time. After sintering, the abrasive particles were sieved by 60/80 mesh screen. As a comparison, abrasives singly doped with 2.5 wt% Na_3AlF_6 were also synthesized by the same process.

Table 1
Description, relative density and average grain size of different samples.

Designation of sintered sample	Additive composition and content (CaO:SiO_2 = 1:1)		Sintering temperature (°C)	Relative density (%)	Average grain size (μm)
	Na_3AlF_6 (wt%)	CaO + SiO_2 (wt%)			
CS0	2.5	0	1400	95.1	0.78
CS1	2.5	1	1400	96.2	0.66
TS1	2.5	1	1400–1300	96.8	0.61
CS2	2.5	2	1400	97.3	0.66
TS2	2.5	2	1400–1300	97.9	0.63
CS3	2.5	3	1400	98.2	0.64
TS3	2.5	3	1400–1300	98.8	0.61
CS4	2.5	4	1400	98.7	0.62
TS4	2.5	4	1400–1300	99.3	0.54
CS5	2.5	5	1400	98.5	0.73
TS5	2.5	5	1400–1300	99.0	0.68

2.2. Sample characterization

The relative density of abrasive particles was determined by Archimedes method. First, a certain amount of abrasive particles were baked in an oven at 105–110 °C for 12 h to constant weight. After that, the particles were placed in a desiccator and cooled to room temperature and their dry weight was measured and recorded as *m*. Then, the abrasive samples were vacuumized in a dry container for 15 min to remove the air in the open pores. The abrasive samples were submerged by injecting distilled water, and the vacuum was continued for 5 min to make the samples saturated. The samples were placed in a pycnometer, and then the pycnometer was filled up with distilled water. When the pycnometer was plugged with a plug with a capillary in the middle, excess water overflowed from the capillary so that the volume of distilled water contained in the bottle was fixed. The weight of the pycnometer filled up with water was measured and recorded as m_a, and the weight of abrasive particles and pycnometer filled up with water was measured and recorded as m_b. The density of abrasive particles was calculated by using the following equation:

$$\rho = m\rho_0/(m + m_a - m_b) \quad (1)$$

where ρ and ρ_0 (0.997 g cm^{-3}) are the density of abrasive particles and distilled water, respectively. The relative density exhibited in Table 1 was expressed in percent of the theoretical density of α-Al_2O_3 (3.98 g cm^{-3}). The microstructure of sintered samples was observed by field emission scanning electron microscopy (FESEM, S4800, Hitachi, Japan). The average grain size and its distribution were calculated and analyzed by using Nano-measurer software based on SEM images. The strength of abrasive particles was measured by using a single particle compressive strength tester (ZMC-II, China). An average of 40 particles were tested for each sample. The temperature of θ- to α-Al_2O_3 phase transformation of the precursor was examined by thermogravimetric differential scanning calorimetry (TGA/DSC, METTLER TOLEDO, Germany) heating 10 mg samples at a rate of 10 °C min^{-1}.

3. Results and discussion

As shown in Fig. 1, the TG-DSC curves of precursor singly doped with 2.5 wt% Na_3AlF_6 were obtained by heating 10 mg of the samples at a rate of 10 °C min^{-1} in air. The endothermic peak at about 100 °C responded to the vaporization of physically absorbed water. The degree of weight loss at this stage depended on the amount of adsorbed water remaining in the sample after drying at 80 °C. Broad exothermic peaks at 329.2 °C were associated with the decomposition and combustion of PEG. The exothermic peak at 1093.9 °C was attributed to the θ- to α-Al_2O_3 phase transformation and was 106 °C lower than the conventional transformation temperature [30]. Different from the transition between metastable polymorphs, the θ- to α-Al_2O_3 phase

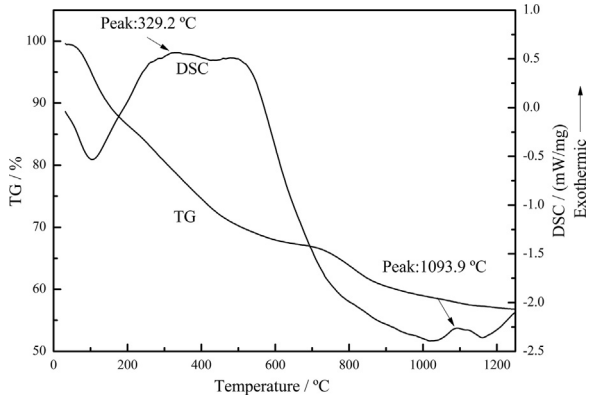

Fig. 1. TG-DSC curves of precursor with 2.5 wt% Na_3AlF_6 and seeds (heating rate = 10 °C min^{-1}).