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Grain growth stagnation in the dense nanocrystalline yttria prepared by combustion reaction and quick pressing with an ultra-high heating rate

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Abstract

Combustion reaction plus quick pressing was a developing technique that used the Joule heating effect of combustion reaction to sinter ceramics, and allows very high heating rate, short soaking duration and high pressure for densification of ceramics. By taking advantages of the particular conditions of this method, pure yttria ceramics with a relative density of 98.5% and an average grain size of 50 nm were obtained at 1620 K and 170 MPa. Moreover, the investigation on the grain growth of sintered yttria was carried out by analyzing the microstructure evolutions and responsible mechanisms. The combined effect of the ultra-high heating rate and the high pressure applied on compact at the peak temperature was effective in suppressing particle coarsening and enhancing densification. Besides, under the decreased sintering temperature and soaking duration, the retained nanostructure assisted to inhibit final-stage grain growth while without impeding the further densification of nanocrystalline ceramics.

Keywords: Grain growth; Densification; Yttria; Heating rate; Nanocrystalline ceramics

1. Introduction

Recently, nanocrystalline ceramics have attracted considerable attentions due to their excellent performances originated from the nanometer size effect, and high-density ceramics with nano-scale grain size are widely demanded for scientific research. To prepare dense nanocrystalline ceramics, one of the most common methods is to densify the compact of nano-particles at a sufficiently high temperature. However, the normally accompanied grain growth is not only inclined to impede densification by diminishing the driving force of sintering, but also adverse to the preservation of nanostructure.[1] Consequently, the control of grain growth is critical in optimizing the microstructure and performance of sintered nanocrystalline ceramics.

Nevertheless, for the conventional sintering methods including conventional (single-step) pressureless sintering and hot pressing, the preparation of dense nanocrystalline ceramics is still a challenging task, which is mainly due to the seemingly inevitable grain growth. Specifically, at early stages of sintering, the low heating rates (10–30 K/min) enable the effect of the grain growth (or particle coarsening) based on time-dependent surface diffusion to be significant. While at final stage of sintering, the high sintering temperatures and long soaking durations required for obtaining highly dense compacts frequently cause exaggerated grain growth. Furthermore, although the goal of restraining the final-stage grain growth has been achieved by adopting multiple special strategies, as utilizing the drag effect of sintering additives, or exploiting the difference in kinetics between densification and grain growth mechanism,[2,3] the side effects of degraded performances (e.g. optical transparency) and reduced efficiency of production are produced, respectively.[4,5]

Therefore, great efforts have been paid on developing the sintering techniques for obtaining dense nanocrystalline ceramics at the less demanding conditions. In comparison with the

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conventional hot pressing method, the advanced pressure-assisted fast-sintering method as spark plasma sintering (SPS) provides a more convenient and efficient way of manufacturing high-density nanocrystalline ceramics.[6,7] In detail, on one hand, the high heating rate is effective in suppressing grain growth in heat-up period.[8] On the other hand, owing to the retained nanostructure, the resulting high volume fractions of grain boundary (GB) are capable of slowing down the rate of the final-stage grain growth based on GB migration via decreasing the GB mobility.[8,9] Besides, the pressure could activate the densification mechanisms of plastic flow with faster kinetics, which also supports to suppress grain growth by decreasing the sintering temperature and soaking duration required for the densification of nano-compact, so as to reduce both the rate and action time of grain growth.

In recent years, the developing pressure-assisted fast-sintering method of combustion reaction plus quick pressing (termed as CR-QP) has been applied to prepare fine-grained ceramics.[10,11] By taking advantages of the sintering conditions typically characterized with the ultra-high heating rate (>1600 K/min), high pressure (>100 MPa) and short soaking duration (limited to several minutes), the grain growth in high-density submicron ceramics could be almost completely suppressed.[10] However, the applicability of CR-QP in manufacturing dense nanocrystalline ceramics has not been verified, and the correlation between the phenomenon of grain growth stagnation in sintered ceramics and the particular sintering conditions of CR-QP deserves further analysis.

Cubic yttria (Y_2O_3) is an appropriate model for researching the grain growth behavior at high temperature.[12] In this work, it is endeavored to prepare dense nanocrystalline yttria ceramics by CR-QP method. For this purpose, the pre-treated Y_2O_3 powders with nano-scale particle size and good sinterability are adopted as the raw materials. Furthermore, the impact of CR-QP conditions on the sintering effects of Y_2O_3 compact is investigated by analyzing the microstructural evolution and the operating conditions of potential mass-transfer mechanisms, so as to shed light on how to stagnate grain growth in sintered nanocrystalline ceramics.

2. Experiment

2.1. Raw materials

Polycrystalline Y_2O_3 powders (purity of 99.99%, Alfa Aesar Co., USA) with an average particle size of 50 nm were used as starting materials. The raw powders were firstly ball-milled in ethanol for 12 h and dried in vacuum at 333 K, then calcinated at 473 K for 6 h in air to eliminate the residual organic impurities. After the treatment, the powders had the preserved particle size and a decreased degree of agglomeration. Subsequently, the powders were compacted by the uniaxial pressing of 10 MPa and the cold isostatic pressing (CIP) of 200 MPa in sequence, so as to be shaped into disk-like compact with a diameter of 20 mm and a height of 4 mm. The relative density of the as-obtained Y_2O_3 green compact was about 48%.

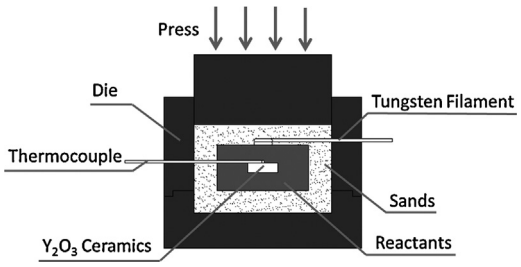

Fig. 1. Schematic of the Y_2O_3 prepared by combustion reaction plus quick pressing (CR-QP).

2.2. Method

In this work, the Y_2O_3 green compact was consolidated in air atmosphere, using a cylindrical CR-QP die (as shown in Fig. 1) with an inner diameter equal to 90 mm and a height of 60 mm. The combustion reaction or chemical reaction between nickel (Ni) and aluminum (Al) metal powders (Eq. (1)) was utilized as thermal source to provide Y_2O_3 compact with a heating effect.

$$Ni + Al \rightarrow NiAl \quad (1)$$

The reactants of combustion reaction was consisted of Ni (particle size of 74 μm, purity of 99.8%) and Al (29 μm, 98.7%) powders in a molar ratio of 1:1, with 0–30 mol% diluents (TiC, 2 μm, 99.5%) added in to adjust the external temperature of Y_2O_3 compact.

In each experiment, a batch of the reactants (150 g) were ball-milled for uniform mixing and then compacted into a cylinder of 60 mm in diameter and 35 mm in height, with a Y_2O_3 green compact in the center. Since that the combustion reaction was predicted to supply the compact with heating effect rather than any effect of mass transfer, a thin layer of graphite foil with excellent thermal conductivity was adopted to coat the compact beforehand, thereby preventing it from being contaminated by the reactants and facilitating thermal conduction.

The as-obtained cylinder of reactants was positioned in the center of die (Fig. 1) by fine sands, which had another important role in conducting the external uniaxial pressure to Y_2O_3 compact in a pseudo-isostatic manner. The combustion reaction of Ni/Al reactants was to be triggered by an energized tungsten filament, which was contacting with the upper surface of the reactants, and the actual temperature of Y_2O_3 compact was monitored by a contact-type thermocouple (WRe5/26-type).

As current was switched on, the tungsten filament instantly flared up to ignite the combustion reaction on the top of the reactant cylinder, and the combustion wave self-propagated to the bottom at a very high velocity. In this procedure, the temperature of Y_2O_3 compact increased to the peak at the ultra-high heating rate derived from the violent exothermic reaction, then immediately started to decrease naturally to the ambient. The heat-up and cool-down period of the compact was completed in several minutes.

In CR-QP process, a commercial available hydraulic machine supply Y_2O_3 compact with an external pressure, which was adjusted to be applied on the compact at the moment that the peak temperature was reached. The designed pressure of the