Available online at www.sciencedirect.com

ScienceDirect

Journal of the European Ceramic Society 34 (2014) 3095–3102

www.elsevier.com/locate/jeurceramsoc

Ultra-high heating rate densification of nanocrystalline magnesia at high pressure and investigation on densification mechanisms

Jianghao Liu [a], Zhengyi Fu [a,*], Weimin Wang [a], Jinyong Zhang [a], Hao Wang [a], Yucheng Wang [a], Soowohn Lee [b], Koichi Niihara [c]

[a] State Key Lab of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Wuhan 430070, China
[b] Department of Environmental Engineering, Sun Moon University, Chungnam 336-708, Republic of Korea
[c] Extreme Energy-Density Research Institute, Nagaoka University of Technology, 1603-1 Kamitomioka, Nagaoka, Niigata 940-2188, Japan

Received 25 August 2013; received in revised form 31 March 2014; accepted 2 April 2014
Available online 2 May 2014

Abstract

The pressure-assisted densification method based on combustion reaction heating was applied to prepare dense nanocrystalline ceramics. The densification process of magnesia compact with a particle size of 50 nm was investigated, under the pressure range of 0–170 MPa, and the temperature range of 1620–1880 K with ultra-high heating rate (above 1600 K/min). The pressure was found to have an effect on enhancing densification while suppressing grain growth, and the higher sintering temperature lead to the larger grain size and lower density of the compact. Pure magnesia nanocrystalline ceramics with a relative density of 99.1% was obtained at 1620 K and 170 MPa, and the concurrent grain growth was almost completely restrained. Furthermore, the investigation on the pressure-dependent densification mechanisms including plastic flow, diffusion and power-law creep was also carried out. The result indicated the rate-controlling mechanism was the plastic flow accommodated by grain-boundary diffusion creep.

Keywords: Heating rate; Densification; Nanocrystalline ceramics; Plastic deformation; Grain growth

1. Introduction

Nanocrystalline ceramics have attracted considerable attentions due to their outstanding properties originated from the nanometer size effect.[1–3] So far, to prepare dense nanocrystalline ceramics, one of the most common methods is to densify the compact of nano-particles at sufficiently high temperature while retain its grain size to nanometer degree.

However, it is still a challenging task to prepare nanocrystalline ceramics by the conventional sintering methods, including conventional pressureless sintering and hot pressing, which is mainly due to the seemingly inevitable grain growth. Therefore, for the purpose of densifying nanocrystalline ceramics while minimizing grain growth, great efforts have been devoted to advancing the sintering techniques. For instance, Chen et al. adopted the special strategy of two-step pressureless sintering to prepare dense nanocrystalline yttria ceramics. However, although the final-stage grain growth was inhibited by exploiting the difference in kinetics between densification and non-densification mechanism, the particle coarsening was strengthened by the rather low heating rate (10–20 K/min).[1,4] Compared with the conventional sintering techniques, the fast sintering method had the advantage of restraining grain growth during the sintering process of ceramics. Particularly, the fast sintering method of spark plasma sintering (SPS) provides a more convenient and efficient way of fabricating high-density nanocrystalline ceramics.[5,6] To be specific, the particle coarsening at early stage of sintering could be effectively suppressed by the high heating rate (up to 600 K/min), and the grain growth at final stage of sintering is limited by the decreased sintering temperature and shortened soaking duration.[7–9]

Recently, a developing fast sintering method based on the combustion reaction heating and quick pressing (termed as CR-QP) has been applied to prepare fine-grained ceramics.[10,11]

* Corresponding author. Tel.: +86 027 87662983; fax: +86 027 87879468.
E-mail address: zyfu@whut.edu.cn (Z. Fu).

http://dx.doi.org/10.1016/j.jeurceramsoc.2014.04.004
0955-2219/

By taking advantages of the particular sintering conditions characterized with the ultra-high heating rate (>1600 K/min), high external pressure (up to 200 MPa) and short effective soaking duration (not exceeding several minutes), the grain growth of submicron ceramics is almost completely restrained.[10] However, the applicability of CR-QP for preparing dense nanocrystalline ceramics has not been verified, and the effects of the sintering conditions on densification and grain growth behavior deserve further analysis. Furthermore, to elucidate the densification process and mechanisms of the nanocrystalline ceramics prepared under the sintering conditions of CR-QP, it is required to enhance densification of nanocrystalline ceramics and suppress grain growth.

Cubic magnesia (MgO) is an appropriate model for studying the densification mechanisms under high temperature and pressure.[12] In this work, the CR-QP method was employed to fabricate high-density nanocrystalline MgO ceramics, and the effects of relevant densification mechanisms were also investigated.

2. Experimental

2.1. Raw materials

Polycrystalline MgO powders (99.99%, Alfa Aesar Co., USA) with an average crystallite size of 50 nm were used as the sintering materials. The powders were subjected to uniaxial pressing of 8 MPa and cold isostatic pressing (CIP) of 200 MPa in sequence, so as to be formed into disk-shaped compact with a diameter of 20 mm and a height of 5 mm. The relative density of the as-obtained green compact was about 45% (the theoretical density of MgO is 3.58 g/cm^3).

2.2. Method

In this work, the combustion reaction between Ni and Al (as Eq. (1)) was utilized as thermal source to supply the MgO compact with a heating effect.[13]

$$\mathrm{Ni} + \mathrm{Al} \rightarrow \mathrm{NiAl} \quad (1)$$

The reactants of combustion reaction consisted of Ni (74 μm, 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 temperature profile of MgO. In each experiment, a batch of the reactants (150 g) was ball-milled for uniformly mixing and then compacted into a cylinder with a green compact of MgO in the center. The green compact was coated by a layer of graphite foil in advance, thus preventing the MgO from being contaminated by the reactants while without hindering the heat transfer from the external combustion reaction to the compact.

The cylinder of reactants was positioned in a self-made steel die (as illustrated in Fig. 1) by sifted sand (95%, quartz, sifted through 100-mesh sieve). In CR-QP experiment, the sand has the function of conducting the external pressure to MgO compact in a pseudo-isostatic manner, and facilitating the release of the gas produced by combustion reaction. The combustion reaction of the reactants was ignited by an energized tungsten filament. As current was switched on, the filament immediately flared up to initiate combustion reaction, and the temperature of MgO started to increase after several seconds. The temperature of sample (MgO compact) was monitored by the WRe5/26-type thermocouple. In CR-QP process, the external pressure was adjusted to apply to sample at the moment that the peak temperature of sample was reached, and the designed stress of sample was attained within one second after the application of external pressure to the CR-QP die. In this pressing mode, the pressure value displayed on the pressure gage represented the pressure between the squeeze head and the upper surface of sand, and there was a certain percentage of loss in pressure during the conduction process of external pressure from its loading surface to sample. For instance, as the external pressure was 200 MPa, the actual stress of sample was measured by a strain gauge to be 170 MPa. To be concise, the stress data mentioned in the following context had been adjusted to the actual stress. Subsequently, the designed stress of sample was maintained constant during the hold duration until the release of external pressure, and the temperature of sample was allowed to decrease naturally to the ambient.

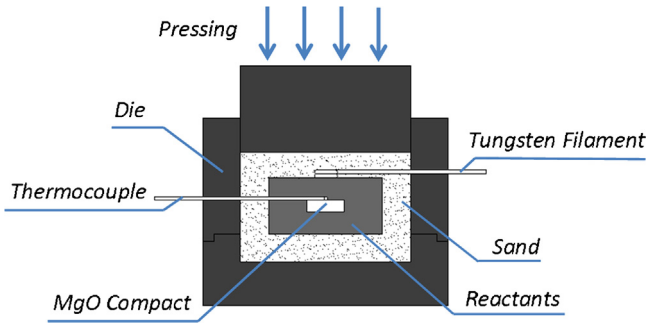

Fig. 1. Schematic of the MgO prepared by combustion reaction and quick pressing (CR-QP).

After the process of combustion reaction and CR-QP, the samples were taken out from the cooled reactants and then mechanically polished to remove the adhering graphite foil, so as to serve for the following tests.

2.3. Test

The relative density of as-preserved MgO was measured by the Archimedes method. The particle or grain size of raw powders and sintered ceramics were measured by averaging the intercepts of 150 grains depicted in the images of field-emission scanning electronic microscopy (FESEM, S-4800, Hitachi). The microstructure of sample was characterized using transmission electron microscopy (TEM, Tecnai G^2 20, FEI). The Vickers hardness of sample was tested by the indentation method (Wolpert-430SVD), which was carried out on the polished surface of sample, with a load of 98 N held for 20 s.

3. Results

In this work, the combustion reaction was adopted to provide the MgO with four different temperature profiles (as