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Synthesis and consolidation of $(Zr_{0.94}Y_{0.06})O_{1.88}$ nanopowders

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Abstract

In this study, a crystalline nanopowder of $(Zr_{0.94}Y_{0.06})O_{1.88}$ has been synthesized by means of combined method of coprecipitation and hydrothermal decomposition using $(ZrO(NO_3)_2 \times 1H_2O)$, $(Y(CH_3COO)_3 \times 1H_2O)$, NH_4OH as a precipitator, and hydroxyethylated nonylphenol as SAS. According to the dilatometry data optimal parameters of hot pressing have been determined. Powders were consolidated by means of the method of hot pressing with direct passage of current at 1100–1200 °C. The microstructures and properties of $(Zr_{0.94}Y_{0.06})O_{1.88}$ powder and ceramics were examined with the use of scanning and transmission electron microscopy, scanning probe microscopy, X-ray phase analysis, Vicker's hardness tester at a test load of 1 kg, and uni-axial compressive tests. It was found that as a result of compaction $(Zr_{0.94}Y_{0.06})O_{1.88}$ ceramics have uniform density distribution (grain sizes of 50–200 nm) with the following optimal mode consolidation values $T=1200$ °C and $P=30$ MPa. The fine-grain materials produced show high microhardness of 1408 ± 30 HV_1 and compressive strength of 2586 MPa.

Keywords: A. Hot pressing; C. Mechanical properties; D. ZrO_2; Hydrothermal synthesis

1. Introduction

Since the discovery of the transformation-toughening phenomenon in zirconia based ceramics [1], these materials have gained extensive interests of research. The unique combination of their excellent qualities such as high mechanical strength, crack resistance, refractoriness, chemical stability, biological inertness [2,3] makes it an essential material in a number of modern technological processes and productions starting with metallurgy and machine building and ending with dentistry [1,4]. However, products from ceramics at high hardness and wear resistance have several disadvantages, primarily, such as brittleness and instability to thermal shocks that often limits the use of ceramics as construction materials [5].

To attain these requirements in ceramic materials, nanocrystallization seems to be one of the promising ways [6]. In the past decade the number of papers devoted to the preparation of substances in nanostate and study of their properties have constantly grown [7–9]. It is known that during the transition from conventional ceramic materials with a grain size of several microns to actually nanometric (20–100 nm), the optical, electrical, magnetic and mechanical properties of the materials change significantly [10]. The introduction of only a few percent of the nanosized particles makes it possible to significantly change properties, and to reach previously unattainable combination of properties [11].

The production of durable nanostructural ceramics on the base of ZrO_2 with a significant increase in mechanical quantities can be realized while creating the material with a thin homogeneous structure [12]. Consequently, nano-crystalline ceramic powders with specified morphology, phase composition, qualities of volumetric (bulk) phase and surface are required to produce different ceramic materials and devices of a new generation such as sensors, storage batteries, high-density ceramic ware, efficient heterogeneous catalysts and sorbents.

Nowadays to obtain nano-crystalline zirconium oxide, a number of methods are used [13–16]. The use of various

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methods of synthesis leads to the formation of nano-crystals ZrO_2 of different crystalline structure, morphology, with different, and, as a rule, rather broad distribution of particles according to their sizes. Besides, technological parameters of nano-crystalline zirconium oxide formation differ greatly not only from one method to another but also within the frames of usage of one method [14–17]. The method of hydrothermal synthesis [14] has become widespread owing to the possibility of obtaining practically isolated nano-crystals ZrO_2 with rather narrow distribution of particles according to their sizes. In spite of the variety of data presented in literature works dedicated to ZrO_2 nano-crystal synthesis in hydrothermal conditions, there is no common opinion as to the temperature and mechanism of zirconium oxide nano-crystals formation in hydrothermal conditions. The production of metal oxide nano-powders from aqueous solution of salts by the hydrothermal method is connected with technological difficulties and requires high temperatures, very high pressure and special devices, that is why the application of the combined method [18], which requires the preliminary production of precipitated mixture of oxides and their further high temperature decomposition is more preferable. The method of mutual precipitation allows reaching high degree of homogeneity of initial stock molecularly and high-temperature hydrolysis contributes to the formation of poorly agglomerated initial nano-crystalline ZrO_2 particles.

The densification processes for conventional powders are investigated, both in theory and practically. However, compaction of nanopowders creates considerable additional problems. On the other hand, general peculiarity of nano-powders, obtained by any method, is their ability to unite into aggregates that leads to some difficulties in compaction (inhibition of intensive growth of grains, provision of even density, preservation of initial fine dispersed structure). To take advantage of the unique properties of bulk nanocrystalline materials, the nanometer range powders have to be compacted into parts of certain properties, geometry, and size. The key to the nanopowder consolidation process is to achieve densification with minimal microstructural coarsening and/or undesirable microstructural transformations. In addition, the fully dense specimen must be of sufficient size for reliable testing of final properties or a useful final product [19].

A number of reviews specifically on nanopowder processing [19–21] and general reviews addressing sintering issues have been published [22,23]. At present there are various efficient methods to consolidate nanopowders that allow the production of nanostructural materials. But analysis of the published information shows that it is difficult to obtain the compacts at the level of density being 0.4–0.6 of the theoretical YSZ density using the traditional ceramic technologies. The sintering temperature heavily depends on the original dispersivity of the powder. The smaller the particles, the higher is their surface energy which provides the reduction of the sintering temperature [24]. At present the synthesis of the powders with particles of micrometer size does not present a significant concern and it is beyond the scope of laboratory investigations. The sintering temperature for YSZ powders with particles of size several nanometers compacted by special methods is 1100 °C [25].

One of the special methods of consolidation of nanopowders is the production of nanostructural materials by uniaxial pressing with simultaneous powerful ultrasonic action (PUA) [26]. The authors in [27] applied the method of dry NP compacting under PUA to sinter high dense nanostructured zirconia ceramics. High dense ceramics with average grain size of $D50 \leq 200$ nm has been sintered at low sintering temperature (1325 °C) using initial zirconia nanopowder having an average particle size of 40 nm.

The most effective methods of activation of sintering approaches are based on the use of electro physical effects on particulate material [28,29]. A wide range of opportunities under electro physics effects on the powder material causes a variety of these methods. The difficulties of nano-powder compaction promoted the development of new high-energy methods of compaction particularly under the influence of electric current, known as field assisted sintering technique (FAST-methods) [30]. A particular case of such technologies is the investigated technology of hot pressing with direct transmission of high amperage current [31].

The methods like hot isostatic pressing (HIP), sintering by high frequency induction heating (HFIHS), rapid compaction (ROC), pulse plasma sintering (PPS), and ultrahigh speed hot consolidation (UPRC) are sufficiently well and completely described in [32–34]. Each of these methods has its own advantages and disadvantages in sintering mono- and polydispersed electroconductive and electrical insulating nanopowders. Thus, the SPS (Spark Plasma Sintering) method, which is widely used at present, allows nanostructural materials to be produced from refractory compounds, e.g., Al_2O_3, SiC, B_4C, $MoSi_2$ [35]. A distinctive feature of the method is the use of a short time pulse current in the process of hot pressing. In the device for pressing by SPS it is possible to avoid rapid growth of nanograins in the material by high heating rate. The process of plasma sintering starts with compression of a powder material under pressure. At first, the mold is rapidly heated by a current of 5000–10000 A, and then an electric pulse of a high power is supplied with the help of a direct current generator. Under the pulse action the energy of a high density is concentrated at the regions of the formation of contact necks between particles of sintered materials, thus initiating the sintering. The method ensures a spatial accuracy of the compact and a high uniformity of the resultant material. It should be noted that up to now the SPS method is the only one of the listed above, which has found a wide industrial application. All the other methods are at the stage of experimental developments [36].

In this study to consolidate nanopowders at hot pressing, it has been proposed to use alternating currents of 1500–2000 A at a voltage of 5–10 V. The device developed is simpler in design for the industrial application, as it does not include a special expensive pulse generator (like in the SPS method) [36,37].

Also, this work aims at studying the peculiarities of the synthesis of the nanocrystalline $(Zr_{0.94}Y_{0.06})O_{1.88}$ powders by means of the combined method and consolidation of the