BIOCYBERNETICS AND BIOMEDICAL ENGINEERING XXX (2018) XXX–XXX

ELSEVIER

Available online at www.sciencedirect.com

ScienceDirect

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

Original Research Article

Experimental investigation of particle size distribution and morphology of alumina-yttria-ceria-zirconia powders obtained via sol–gel route

Q1 D.S. Nakonieczny [a,*], M. Antonowicz [b], Z.K. Paszenda [a], T. Radko [b], S. Drewniak [c], W. Bogacz [d], C. Krawczyk [e]

[a] Department of Biomaterials and Medical Devices Engineering, Faculty of Biomedical Engineering, Silesian University of Technology, Roosevelt 40 st., 41-800 Zabrze, Poland

[b] Institute for Chemical Processing of Coal, Zamkowa Str. 1, 41-803 Zabrze, Poland

[c] Department of Optoelectronics, Silesian University of Technology, B. Krzywoustego st., 2, 44-100 Gliwice, Poland

[d] Department of Chemical Engineering and Process Design, Faculty of Chemistry, Silesian University of Technology, M. Strzody 7 st., 44-100 Gliwice, Poland

[e] Department of Dental Technology, Medical College of Zabrze, 3 maja 63 st., 41-800 Zabrze, Poland

ARTICLE INFO

Article history:
Received 2 December 2016
Received in revised form
20 February 2018
Accepted 25 February 2018
Available online xxx

Keywords:
Zirconia
Prosthetic dentistry
Sol–gel
Thermal analysis
Morphology

ABSTRACT

Background: Oxide-doped zirconia is currently commonly used ceramics in dental prosthetics. However, its use raises a lot of controversy. This is related to the stability of the zirconia metastable phases in the human mouth environment and it sensitivity for the so-called *low-temperature degradation*. A key way to avoid this type of negative phenomena is doping ZrO_2 with selected metal oxides and choosing appropriate methods for the synthesis of ceramic powders.

Objective: The aim of this paper is to present investigations of modification and to analyse the influence of chemical composition and volume of parent-solvent for the morphology and thermal properties of ceramic powders prepared in a ZrO_2-CeO_2-Y_2O_3-Al_2O_3 system.

Methods: The powders were obtained by using the sol–gel method in an inert gas atmosphere and ambient temperature using zirconium n-propoxide for this purpose. Morphology was examined by using scanning electron microscopy (SEM) and particle size distribution (PSD); thermal properties was evaluated using thermogravimetric analysis (TGA/DTA/DTG), and chemical composition was confirmed by using electron probe microanalysis (EPMA)

Results: Depending from the volume of the CeO_2 precursor solution of and regardless of the Q2 volume of the second oxide precursor, was observed difference in morphology of the obtained powders. Overall trend is related to reduce the size of agglomerates with an increase in the volume of the precursor of CeO_2.

Conclusions: The influence of various chemical compositions for morphology and thermal properties is negligible. In contrast, a clear correlation is observed between the volume of

* *Corresponding author at*: Charles de Gaulle'a 40 st, 41-800 Zabrze, Poland.
E-mail address: damian.nakonieczny@polsl.pl (D.S. Nakonieczny).

https://doi.org/10.1016/j.bbe.2018.02.010

Please cite this article in press as: Nakonieczny DS, et al. Experimental investigation of particle size distribution and morphology of alumina-yttria-ceria-zirconia powders obtained via sol–gel route. Biocybern Biomed Eng (2018), https://doi.org/10.1016/j.bbe.2018.02.010

parent alcohol for both morphology and thermal properties. Use of sol–gel method to further research in view of these results appears to be appropriate.

1. Introduction

Zirconium oxide is the most widely used ceramic material for the fixed-prosthetic dentures (FPD) [1,2]. Its use in dental technology is associated with very good mechanical properties and the ability to achieve very good aesthetic effect of designed prostheses [1,2]. Zirconia is used for preparing all types of restorations from single crowns through complicated bridges, abutments and implants [1–3]. The most popular material used in the medical applications is a type of zirconia i.e., 3Y-TZP, Y_2O_3-stabilized tetragonal phase β ZrO_2 [4–6]. The use of ZrO_2 in biomedical engineering is quite controversial, which is due to the instability of zirconia metastable phase in an environment of body fluids. This is a result of uncontrolled martensite type phase transformation from tetragonal phase to the monoclinic $\beta ZrO_2 \rightarrow \alpha ZrO_2$. The transformation is accompanied by a volume increase of the ZrO_2 grains from 4 to 6% and an increase in hardness and in consequence possibility of ceramic fracture [4–7]. Another negative phenomenon affecting exploitation of zirconia ceramics in the human body fluids environment is the so-called low thermal degradation (LTD) [8,9]. Both phenomena are widely discussed and mutually interrelated. Numerous investigations about LTD and the martensitic transformation can be found in the work of Chevalier and Gremillard and Guo [10–13]. LTD becomes a gradual and systematic destruction from the surface contacting with the electrolyte. The exact mechanism of this phenomenon is not clearly described and understood. Most information on this topic can be found in the cited work containing a comprehensive description of the thermodynamic and kinetic models and data from computer simulations verified experimentally [14–16]. Degradation changes occurring most rapidly in an environment of varying pH, particularly in the acid range and an elevated constant temperature, generally from body temperature to about 400 °C [9,10,17]. The ways to reduce these phenomena are doping metastable phases with metal oxides, and applying a strict control of the ceramics grain size [5,18]. In general, the observed trends make use of the reduction of grain which allows to increase the resistance for the LTD [10,19]. Regarding the doping and doping method there are some particularly desirable additives, in which the central atom has a larger or equivalent radius than the Zr^{4+} ion [18,20]. Additionally, it should also be taken into account the following factors: the valence of the cation, (depending on the different stabilization achieved); the influence dopant type to the formation of liquid solution during sintering (the occurrence of the liquid phase during sintering results in an inhomogeneous distribution of dopant species in zirconia lattice); value of the diffusion coefficient (D_{Me}) (this factor mainly influences the time of sintering, distribution of O^{2-} vacancies and zirconia phase composition); amorphousness or crystallinity of the oxide dopant (the mechanism of stabilization is different depending on the dopant nature). Analysing some research results, it can be stated that CeO_2, as a dopant, provides proper stability of the β phase and uniform grain size [17,18,23,24]. Stabilization of β-ZrO_2 structure of cerium oxide is proven by oxygen vacancies, introduced with CeO_2, which further stiffens the lattice network, preventing the unfavourable $\beta ZrO_2 \rightarrow \alpha ZrO_2$ transformation [18,24,25,27,28]. The next factor that predisposes CeO_2 or other tetravalent dopants for stabilizing β phase is that Ce^{4+} ion has a similar atomic radius to that of the Zr^{4+} ion, which stiffens the zirconia crystal lattice [9,18,20].

It is generally known that the sol–gel method allows obtaining a wide range of different types of materials including powders with strictly controlled chemical and phase composition, along with a good morphology. Changing the variables, such as solvent type, pH, reaction temperature, molar ratios of reactants or type of the oxide precursors which can affect final properties of ceramic can be strictly controlled [29,31]. There are some valuable investigations, for example, the one found by Caruso et al. [29], which deals with the correlations between synthesis parameters and the morphology of ceramic powders, and their technological properties during the preparation , for example, during sintering. From this point of view, the main aim of present study is to investigate the mechanisms of sol–gel powder preparation and its influence for the thermal properties and morphology of as-obtained cerium-yttria-alumina-doped zirconia powders. For these reasons, the main subject of this paper was to investigate influence of various chemical composition and molar ratios of parent-alcohol for as-mentioned properties.

2. Materials and methods

2.1. Material

Samples preparation procedure followed in this study was similar to the one previously applied [21,22]. On previous investigations, some base subsequent procedures for the following tests have been developed. As a zirconia precursor we used zirconium n-propoxide (ZNP, 70 wt.% in propanol, Sigma–Aldrich). 2-Propanol was used as a solvent (PrOH, Avantor). As a ceria precursor, cerium nitratehexahydrate (CNH, 1 M solution in 2-propanol, Sigma–Aldrich) was used. As yttria precursor ytrria nitrate hexahydrate was used (YNH, 1 M solution in 2-propanol, Sigma–Aldrich), and as an alumina precursor aluminium isopropoxide (AlP, 0.5 M solution in 2-propanol, Sigma–Aldrich) was used. To control the hydrolysis rate as a chelating agent acetylacetone (AcAc, Avantor) was used. As a pH-agent ammonia (NH_4OH, Avantor) was used.

Please cite this article in press as: Nakonieczny DS, et al. Experimental investigation of particle size distribution and morphology of alumina-yttria-ceria-zirconia powders obtained via sol–gel route. Biocybern Biomed Eng (2018), https://doi.org/10.1016/j.bbe.2018.02.010