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Research Paper

Preparation and evaluation of cerium oxide-bovine hydroxyapatite composites for biomedical engineering applications

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ABSTRACT

The fabrication and characterization of bovine hydroxyapatite (BHA) and cerium oxide (CeO_2) composites are presented. CeO_2 (at varying concentrations 1, 5 and 10 wt%) were added to calcinated BHA powder. The resulting mixtures were shaped into green cylindrical samples by powder pressing (350 MPa) followed by sintering in air (1000–1300 °C for 4 h). Density, Vickers microhardness (HV), compression strength, scanning electron microscopy (SEM) and X-ray diffraction (XRD) studies were performed on the products. The sintering behavior, microstructural characteristics and mechanical properties were evaluated. Differences in the sintering temperature (for 1 wt% CeO_2 composites) between 1200 and 1300 °C, show a 3.3% increase in the microhardness (564 and 582.75 HV, respectively). Composites prepared at 1300 °C demonstrate the greatest compression strength with comparable results for 5 and 10 wt% CeO_2 content (106 and 107 MPa) which

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are significantly better than those for 1 wt% and those that do not include any CeO_2 (90 and below 60 MPa, respectively). The results obtained suggest optimal parameters to be used in preparation of BHA and CeO_2 composites, while also highlighting the potential of such materials in several biomedical engineering applications.

1. Introduction

It is estimated that 280,000 hip, 700,000 vertebral and 250,000 wrist fractures cost US $10 billion per year. This has led to a significant increase in demand within the surgical and biomaterial market. Globally, procedures involving bone grafts or bone substitutes, fluctuate around 4,000,000 per annum (Brydone et al., 2010). One of the main reasons for this is due to the increase in life expectancy, which comes with the natural aging process. For example, the life expectancy of 27 EU countries continues to project an increase in this, with the share of the population aged around 65 years and over rising from 17% in 2010 to 30% in 2060, and those aged 80 and over rising from 5% to 12% over the same period (Eurostat, 2011; Kalache et al., 2007). With such predictions in mind, an increase in the biomaterial and surgical markets can also be expected, as the number of people prone to bone fracture will also increase.

Surgically, autografts and allografts (regarded as the golden standard) are accepted as standard treatments; they are sometimes accompanied by problems of donor site scarcity, rejection by the immune system, resorption and transfer of pathogens (Gao et al., 2006). Due to the scarcity of golden standards, biomaterials research has led to the development of new materials, which offer properties similar to those of bone. One synthetic material, hydroxyapatite (HA) [Ca_{10} $(PO_4)_6$ $(OH)_2$], is perhaps one of the most utilized biomaterial in this regard (Gao et al., 2006). The approximate composition of this synthetic material is comparable to that of bone (Silva and Lameiras, 2000; Valerio et al., 2004; Oktar et al., 2001) and teeth (Silva and Lameiras, 2000; Oktar and Altintas, 1998) (naturally occurring HA). However, it exhibits low fracture toughness due to its lack of strength and high brittleness (Silva and Lameiras, 2000; Sampaio et al., 2005; Valerio et al., 2004; Gunduz et al., 2008), thereby providing an obstacle to its application as a standalone biomedical device that must withstand high loads (Silva and Lameiras, 2000). To improve such mechanical properties of HA materials (i.e., to increase their fracture toughness), incorporation of metallic materials, ceramic oxides, whiskers or even fibers have been suggested (Salman et al., 2009; Erkmen et al., 2007).

In this study, CeO_2 – a rare earth oxide was used. CeO_2 is generally used for stabilizing the tetragonal polycrystalline structure of zirconia (Yousefpour et al., 2011). The potential use of rare-earth elements in the development of bone tissue has been unclear up to now (Ivanchenko et al., 2009). There are also some studies with lanthanum oxide and its potential use as a reinforcement material for HA (Pazarlioglu et al., 2011; Oktar et al., 2006). Some groups are also addressing other rare-earth elements such as cerium and what potential this might have in reduction of dental enamel demineralization (Feng and Liao, 2005) and its antibacterial abilities in preventing caries (Yingguang et al., 2007). It is also known that CeO_2 is used for improving the sintering of glass ceramics and strength and thermal stability (Ivanchenko et al., 2009). Cerium has also been used as a luminophor agent in the composition of ceramic powders, which may also be beneficial in dental applications. When a dental ceramic is not fluorescent, it tends to have an appearance of reduced vitality, presenting a grayish appearance (Volpato and Fredel, 2010).

The aim of this study is to improve mechanical properties of bovine derived hydroxyapatite (BHA)–CeO_2 composites by using different sintering temperatures in order to assess their microstructural and mechanical properties for high load-bearing biomedical engineering applications.

2. Materials and methods

2.1. Materials

Naturally occurring apatite (approximate composition of synthetic apatite) was obtained from fresh bovine bones. The shaft segment of femoral bovine bones was obtained (epiphyseal components were excluded due to excessive organic residues) from an international abattoir (CarrefourSA Erenkoy, Istanbul, Turkey). CeO_2 powder (99.5% purity, REO) was obtained from Alfa Aesar, USA. This displayed an average particle size range between ~0.07–0.1 μm and a surface area of ~12 m^2/g. Sieves were purchased from Horizontal Sieve Shaker AS 400 Retsch, Haan, Germany.

2.2. Preparation of CeO_2–BHA composites

The specimens were dissected and the medulla osseo components of the samples were removed by manual abrasion. The bone samples were irrigated with copious amounts of tap water and were then deproteinized using an alkali solution (NaOH, 1%) (Goller and Oktar, 2002). Following deproteinization, BHA samples were washed thoroughly again (using copious tap water), dried and calcined at 850 °C (Goller et al., 2006; Oktar et al., 2006) to remove any prions (i.e., Creutzfeldt–Jakob disease (CJD), bovine spongiform encephalopathy and BSE) (Goller et al., 2004; Ozyegin et al., 2004). The samples were then crushed first in a ceramic mortar and then with a ball grinder for 4 h (Planetary Ball Mill PM 200 Retsch, Haan, Germany). The resulting BHA powder was sieved (100 μm) and was subsequently mixed with 1, 5 and 10 wt% of CeO_2 content separately. Each mixture was then ball grinded for further 4 h. According to British Standard 7253 (Salman et al., 2009), the prepared powder mixtures