

# Structural, mechanical and biological properties of hydroxyapatite-zirconia-lanthanum oxide composites



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## ABSTRACT

Novel hydroxyapatite-zirconia-lanthanum oxide composites for bioceramic applications were synthesized and their structural, mechanical and biological properties were studied. Pure HA was produced via precipitation method and the composites were obtained by several fabrication steps: powder milling, mixing, cold pressing and sintering at 1100 °C for 1 h. The experimental results indicated that the composites consisted of hydroxyapatite as the main phase with a trace amount of tricalcium phosphate. Calcium zirconate (CaZrO<sub>3</sub>) was also formed by the reaction between zirconia and calcium oxide (CaO) which is the thermal decomposition product of hydroxyapatite. Addition of zirconia and lanthanum oxide resulted a more loose and porous structure on the surface. The diametral tensile strength of the composites was higher with respect to pure hydroxyapatite. The microhardness of the composites, except the one with the composition of 90 wt% HA and 10 wt% Zr, was relatively lower than that of pure HA but these composites had higher machinability. Cell culture studies with osteoblast-like Saos-2 cell line showed that composites and pure hydroxyapatite were biocompatible. Based on these findings, hydroxyapatite-zirconia-lanthanum oxide composites hold potential to be used in hard tissue replacement and regeneration therapies.

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## 1. Introduction

The vast majority of implants used in hard tissue replacement or restoration therapies are comprised of metallic biomaterials, such as Ti and its alloys. These materials are generally treated with various surface modification techniques; especially, hydroxyapatite (HA) coating. Surface modifications can partially or totally prevent the localized corrosion of metallic biomaterials in the physical environment. However, the difference between the mechanical characteristics of human bone and the metallic implant would still remain as a major disadvantage. In addition, even if the metals or alloys can meet the biomechanical requirements, the bonding between the metallic surfaces and the surrounding bone tissue is poor which generally leads to the formation of a fibrous tissue around the implant as a sign of incomplete osseointegration. This is the reason why bioceramic coatings are widely applied to the metallic implants. Moreover, for many cases, a better solution is offered by bioceramics rather than metallic implants with surface coatings in terms of improved biocompatibility and bioactivity.

In general, bioceramics are more biocompatible than any other hard tissue implant material and have less effect on the immune system [1]. They can be produced in a broader range of biochemical, mechanical and functional properties. Their service time may be adapted to the specific applications by taking the advantage of biodegradability. Ceramics that are used as implant materials can be classified in two large groups: bioinert and bioactive. Bioinert ceramics, such as zirconia (ZrO<sub>2</sub>), show almost no interaction with the surrounding living tissue. On the contrary, bioactive ceramics, such as calcium phosphates (CaPs), are capable of bonding with living hard tissues. HA is one of the most interesting bioactive bioceramics since it is the main inorganic constituent of the bones and teeth of all vertebrates. It amounts to 65% of the total bone mass [2], and the remaining mass is composed of organic matter, mostly type-I collagen, and water.

The mineralized tissues are mostly exposed to mechanical load. Their function is to provide necessary stiffness and strength according to the correct balance between these and toughness. The mechanical properties of a hard tissue replacement implant must resemble their natural analogues to a high degree. HA can be used in its dense or porous forms, for example as a bone filler material, within the limits allowed by its intrinsic mechanical properties of low strength and high brittleness. However, it fails to reach the minimum required performance in the applications where local

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loading is high. For instance, the fracture toughness ( $K_{Ic}$ ) of pure HA is reported to be around 1 MPam<sup>1/2</sup>, while the reference value for human bone is up to 12 MPam<sup>1/2</sup> [3]. In order to achieve the necessary mechanical strength, biocompatibility and dissolution rate at the same time, HA can be used in combination with other materials in the form of composites.

There are mainly two alternative classes of components for obtaining HA-containing composites; (1) ceramics with small particles or as sintered dense bodies; (2) biocompatible polymers as matrices for HA particles. Similar to ceramics, phosphate-based glasses and bioactive silica glasses can also be combined with HA. Polymers, such as polyethylene, collagen and polylactide, can be used to obtain HA-polymer composites with the aim of enhancing strength and reducing brittleness of HA ceramics [4] or loading drugs against infection and various other diseases.

The combination of the properties of various materials with that of HA has been previously studied in order to optimize the properties of HA as an implant material. Partially stabilized zirconia (PSZ), which is bioinert and has the highest strength and fracture toughness values of oxide-based ceramics, was used in combination with HA to obtain a bioactive implant material with improved mechanical properties [5,6]. Similarly, other oxides, such as alumina (Al<sub>2</sub>O<sub>3</sub>) [7], titania (TiO<sub>2</sub>) [8], yttria (Y<sub>2</sub>O<sub>3</sub>) [9], were studied to prepare HA composites in different structures via several methods. Besides, lanthanum oxide (lanthana, La<sub>2</sub>O<sub>3</sub>) is one of the potentially interesting oxides that can be used as a reinforcement agent for HA. It is previously reported that the compression strength and microhardness of HA-La<sub>2</sub>O<sub>3</sub> composites increased with increase in the amount of La<sub>2</sub>O<sub>3</sub> and sintering temperature [10].

The aim of this study was to investigate the structural, mechanical and biological properties of HA-ZrO<sub>2</sub>-La<sub>2</sub>O<sub>3</sub> composites that were synthesized in different weight compositions. Since successful integration of the biomaterials with living tissues is related to the surface morphology and composition, attention was focused on the microstructural and morphological characterization together with the assessment of cytocompatibility as a basic requirement for biomaterials.

## 2. Materials and methods

### 2.1. Synthesis of pure HA and HA-ZrO<sub>2</sub>-La<sub>2</sub>O<sub>3</sub> composites

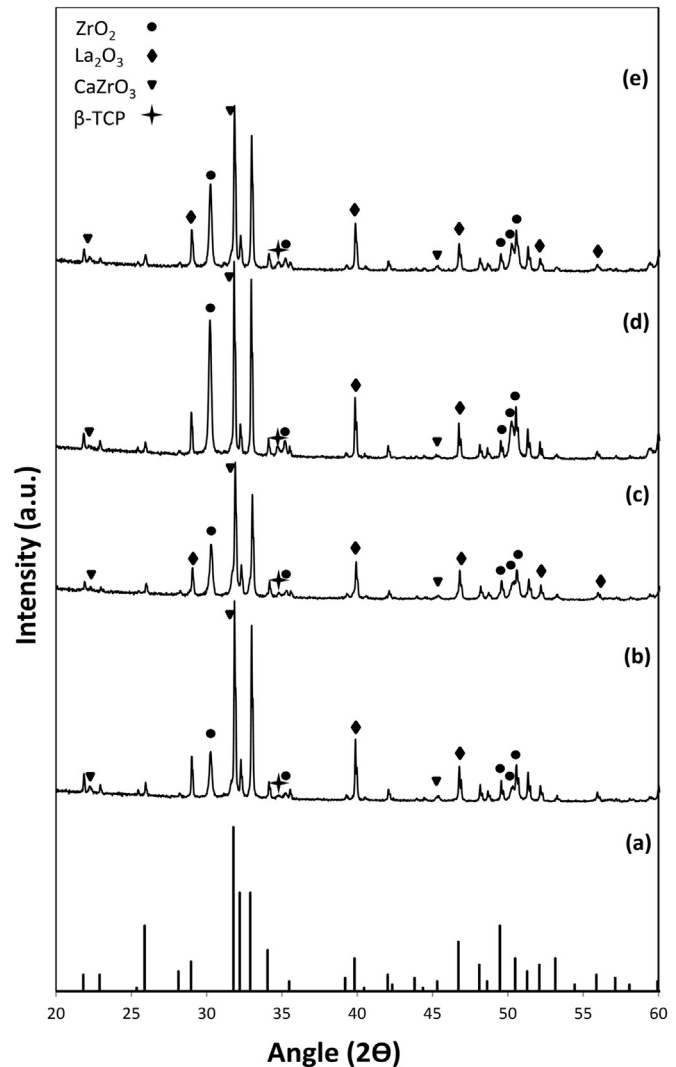
The solutions of 0.5 M calcium nitrate (Ca(NO<sub>3</sub>)<sub>2</sub> · 4H<sub>2</sub>O, Merck, Germany) and 0.3 M diammonium hydrogen phosphate ((NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub>, Merck, Germany) were prepared in distilled water, separately. The pH of the solutions was adjusted to 10 by using ammonia solution (NH<sub>4</sub>OH, Merck, Germany). Di-ammonium hydrogen phosphate solution was added dropwise into the calcium nitrate solution under constant stirring. The resulting solution was stirred for 2–3 h, and then it was heated at 90 °C for 1 h. The solution was further stirred for 1 day at room temperature. It was filtered through a fine filter paper and the filtrate was washed with distilled water several times. In order to remove the excess water and ammonia, the wet cakes were dried in an oven at 200 °C. The dried HA particles were ground to fine powder and calcined at 1100 °C for 1 h.

In order to synthesize HA-ZrO<sub>2</sub>-La<sub>2</sub>O<sub>3</sub> composites, commercially available tetragonal zirconia (*t*-ZrO<sub>2</sub>, Aldrich, Germany) and La<sub>2</sub>O<sub>3</sub> (Aldrich, Germany) were added to the initial precursors used for preparation of pure HA according to the defined ratios given in Table 1.

The powders were mixed in pure ethanol, stirred for 3–5 h and dried in an oven overnight to remove alcohol. The composite powder was milled and further mixed in a mortar and pestle. The

**Table 1**  
Compositions of HA and composites.

Sample ID	wt% HA	wt% <i>t</i> -ZrO <sub>2</sub>	wt% La <sub>2</sub> O <sub>3</sub>
HA	100	0	0
90HA10Zr	90	10	0
88HA10Zr2La	88	10	2
80HA20Zr	80	20	0
78HA20Zr2La	78	20	2



**Fig. 1.** XRD patterns of (a) standard HA (JCPDS #9-432); (b) 90HA10Zr; (c) 88HA10Zr2La; (d) 80HA20Zr; and (e) 78HA20Zr2La composites sintered at 1100 °C for 1 h.

composite powders were cold pressed in tool-steel die to form pellets with an automatic hydraulic press under 10 kN force for 1 min. The resulting pellets had a diameter of 1 cm and were about 2–3 mm thick. The pellets were sintered at 1100 °C for 1 h.

### 2.2. Characterization

X-ray diffraction (XRD, Bruker D8) analysis was performed to determine the presence of different phases by using Cu-K $\alpha$  radiation at 40 kV/40 mA. The scanning range was between 2 $\theta$  angles of 20° to 60°. The diffractograms were compared with the Joint Committee on Powder Diffraction Standards (JCPDS) cards. The surface morphology and grain sizes of the samples were

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