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Hardystonite bioceramics from preceramic polymers

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ABSTRACT

In this work, we demonstrated that the hardystonite ($\text{Ca}_2\text{ZnSi}_2\text{O}_7$) bioceramics can be produced with high phase purity, starting from different preceramic polymers and suitable fillers (precursors for CaO and ZnO) after heating at 1200 °C in air. Open-celled hardystonite foams were easily prepared from a filler-containing silicone resin using hydrazine as foaming agent. The fabrication of cellular structures using a preceramic polymer and fillers was possible because the polymeric melt allowed for the entrapment of the gases generated by the decomposition of hydrazine, and the simultaneous cross-linking of the preceramic polymer enabled the retention of the foam structure. Samples with a well-developed hierarchical porous structure, with an open porosity ranging from ~65 to ~81 vol% and an average cell window size ranging from 150 to 500 μm were produced. The hardystonite components possessed a compressive strength ranging from ~1.4 to ~2.1 MPa.

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1. Introduction

Porous bioceramics have received significant attention concerning their use in biomedical application such as bone graft scaffolds [1], bone filler [2], orbital implant [3] and drug delivery carriers [4,5]. Bioactive ceramics are often produced in the form of three-dimensional (3-D) porous scaffolds, which act as templates supporting and directing tissue in-growth and regeneration [6–8]. The current challenge in bone tissue engineering is the design of porous bioceramics possessing simultaneously a highly porous structure, adequate mechanical properties and bioactivity to repair load-bearing bone defects [9].

There are several different approaches on how to produce highly porous bioceramic foams, which can be grouped into three main techniques: i.e., the replica method [10,5], the sacrificial template method [11] and the direct foaming of liquid suspensions or solutions [12–14]. In the replica method, a macro-porous structure with uniform distribution of pores can be obtained by immersing a polyurethane foam in a ceramic slurry. By sintering at high temperature, the organic components are burnt out, producing a ceramic foam scaffold with interconnected pores [15,16]. In the sacrificial

template technique, porosity is generated by adding fillers (porogens). For instance, PMMA microbeads [17] can be added, which decompose during sintering, or freeze casting can be used, which generates *in situ* some interconnected structures that are eliminated by sublimation leaving porosity behind [18,19]. Ceramic foams with high levels of interconnected porosity can also be obtained by direct foaming of a slurry or a solution, followed by setting of the foamed mixture (by gel casting or cross-linking). The incorporation of bubbles is achieved by injection of gases through the liquid suspensions, mechanical agitation, blowing agents, evaporation of compounds or by evolution of gas by an *in situ* chemical reaction. The direct foaming method is one of the most versatile among the methods to fabricate ceramic foams, and it offers a simple, low-cost and effective way to produce ceramic foams in which the pore structure can be effectively controlled [20–23].

Calcium silicate-based bioceramics have been intensively investigated as potential bioactive material for bone tissue regeneration and implant coating due to their excellent bioactivity and degradability. A common Ca–Si ceramic used in bone tissue regeneration is CaSiO_3 . However, the chemical instability with high ionic dissolution and degradation rates of CaSiO_3 ceramics limit their biological applications. One strategy to improve the biological performance of CaSiO_3 bone substitute materials is to modify their chemical composition, by incorporating different elements such as Zn, Mg, Ti, Sr, and Zr to develop silicate ceramics with different degradation rates, biological behavior and mechanical properties. This

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combination of suitable features makes these ceramics excellent potential candidates for fulfilling the very broad and stringent requirements typical of medical applications [24–27].

Hardystonite ($\text{Ca}_2\text{ZnSi}_2\text{O}_7$, HT) is a Ca-rich silicate bioceramic, developed by incorporating Zn into the Ca–Si oxide system to improve the chemical stability [28]. Sol–gel derived hardystonite ceramics have also been considered as promising materials for bone tissue engineering [29,30]. These materials have been shown to increase the proliferation rate of human bone marrow-derived mesenchymal stem cells (MSCs) and to induce osteogenic differentiation of MSCs [29]. Furthermore, $\text{Ca}_2\text{ZnSi}_2\text{O}_7$ ceramics have been shown to support cell attachment, and to increase cellular proliferation and differentiation compared to CaSiO_3 . In addition, $\text{Ca}_2\text{ZnSi}_2\text{O}_7$ showed stimulate expression of alkaline phosphatase (ALP), osteocalcin and collagen type I when in contact with human osteoblast-like (HOB) cells [30]. Moreover, hardystonite ceramics possess improved biocompatibility [31], bending strength and fracture toughness in comparison to hydroxyl-apatite (Hap) and CaSiO_3 [28,32]. In a previous work, it was shown that it was also possible to develop Zn-containing silicates, such as hardystonite ($\text{Ca}_2\text{ZnSi}_2\text{O}_7$) and willemite (Zn_2SiO_4), by simply adding ZnO powders to a bioglass powder (AP40 glass) which crystallizes in the apatite/wollastonite system, and/or by embedding them in the preceramic polymer, as additional fillers, followed by heating at 1000°C . The mechanical properties of the produced bioceramics depended on the crystalline phase assemblage in the material, and increased when hardystonite was present [27]. These results indicate that hardystonite ceramics are appropriate candidates for bone regeneration and hard tissue engineering.

Particularly, due to the difficulties with sintering, silicate-based ceramics are generally obtained by complex techniques, such as the hydrothermal method, devitrification of glass, solution combustion processes, spark plasma-sintering, sol–gel processing etc. For preparation of pure ternary-oxide and quaternary-oxide silicate powders, it is necessary to select the proper chemicals and synthesis method, and to control the reaction conditions. The sol–gel method is undoubtedly favorable for the synthesis of ternary-oxide and quaternary-oxide silicate powder, but the preceramic polymer approach, with fillers reacting with the ceramic residue left by the polymer upon heat treatment, originally applied to binary silicates

(e.g., wollastonite, CaSiO_3), is currently extended to much more complex systems (such as $\text{CaMgSi}_2\text{O}_6$). The preceramic polymer route offers several advantages in terms of easy shaping of the components, high reactivity of the precursors in contact with the fillers, simplicity, reduced cost of the precursors in comparison to sol–gel, and low processing temperatures compared to classical ceramic processing. Furthermore, the addition of fillers, reacting with the ceramic residue left by the polymer upon heat treatment, permits the synthesis of a variety of ceramic materials with different compositions and high phase purity [20,24,27].

This research aimed at developing hardystonite bioceramics from different preceramic polymers, with the addition of CaCO_3 and ZnO as active fillers, as well as at the fabrication of highly porous cellular structures. This approach takes advantage of the fact that the use of preceramic polymers allows for different shaping possibilities, such as the production of bulk components or highly porous ceramics obtained by direct foaming.

2. Experimental details

2.1. Preliminary synthesis studies

Commercially available silicones, Silres[®] MK (Wacker–Chemie GmbH, München, Germany), H62C (Wacker–Chemie GmbH, Munich, Germany), CSO-300 and CSO-310 (EEMS LLC, USA), were used as precursors for silica. The MK is silicone resin in powder form, while the other silicone resins were in liquid form. Thermogravimetric analysis (TGA, STA409, Netzsch GmbH, Selb, Germany), with heating rate of $10^\circ\text{C}/\text{min}$, was applied to assess the ceramic yield of the different silica precursors.

The silicone resins were dissolved in isopropyl alcohol (15 ml for 10 g of final ceramic) under magnetic stirring. After this step, active fillers consisting of ZnO nano-sized particles (30 nm, Inframat[®] Advanced Materials, Manchester, CT, USA) and CaCO_3 micro-sized particles ($<10\ \mu\text{m}$, Sigma–Aldrich, Gillingham, UK) were dispersed into silicone solutions, without the addition of any chemical dispersant, in the stoichiometric proportions to yield pure hardystonite (for each silicone, the polymer/fillers weight ratio were adjusted considering the previously determined ceramic yield of the silicones). All suspensions were mixed by magnetic stirring and

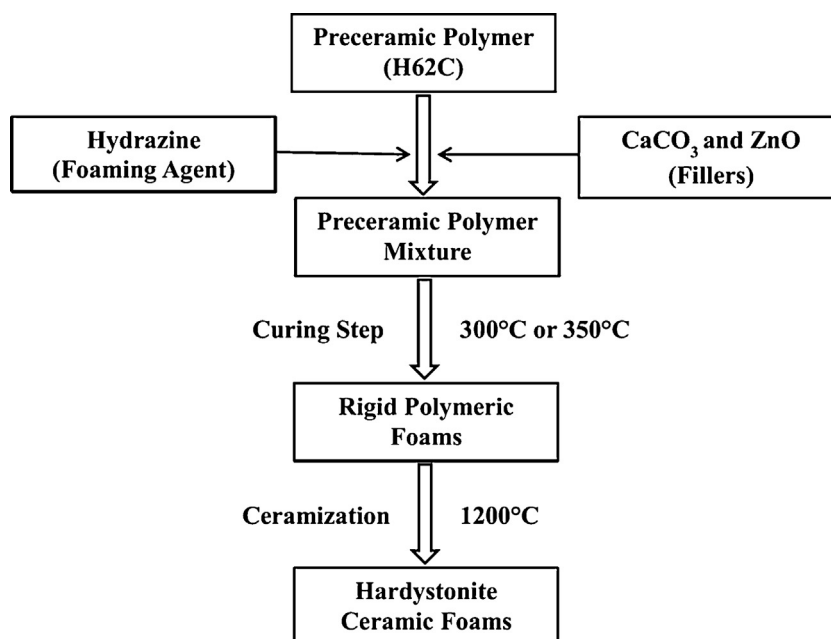


Fig. 1. Flow-chart of the process for the fabrication of porous hardystonite ceramic foams from a preceramic polymer and fillers.

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