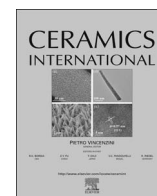




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Preparation and characterization of iron/ β -tricalcium phosphate biocermet for load-bearing bone substitutes

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ABSTRACT

Ceramic-metal composite materials, namely cermet, are provided with characteristics of both ceramic and metal. Herein, for the first time bio-cermet based on β -tricalcium phosphate (β -TCP) bioceramic with biodegradable iron being reinforcement phase, were fabricated using the powder metallurgical method. The phase composition, microstructure, mechanical properties and in vitro cell behaviors of bio-cermet were investigated. The results revealed that atomic diffusion occurred between the iron and β -TCP matrix during the sintering process. The bio-cermet attained remarkable increase in fracture toughness ($1.16\text{--}1.55\text{ MPa m}^{1/2}$) compared to the β -TCP bioceramic ($0.54\text{ MPa m}^{1/2}$). The bio-cermet with 10 vol% iron showed the highest compressive strength (640 MPa), significantly higher than that of plain β -TCP bioceramic (285 MPa). The in vitro cell behaviors test indicated that the bio-cermet did not show any sign of toxicity; the iron ions released from bio-cermet up-regulated bone-related gene expression of bone mesenchymal stem cells. The bio-cermet developed in this study represent potential bone substitutes for application in the load-bearing bone defects.

1. Introduction

Calcium phosphate (CaP) bioceramics are attractive synthetic bone grafts for critical-size bone defect. CaP materials possess excellent biocompatibility and osteoconductivity, owing to their chemical similarity to mineral phase of living bone [1,2]. Among CaP materials, hydroxyapatite (HA), β -tricalcium phosphate (β -TCP), and biphasic calcium phosphate (BCP, a mixture of HA and β -TCP) are the most widely studied and used in clinic [3,4]. HA is structurally stronger but poorly resorbed [5]. β -TCP features more considerable resorption rate due to its higher solubility and cell mediation in vivo. The calcium and phosphate ions released from β -TCP are involved in the bone remodeling process [6,7]. Nevertheless, the inherent brittleness and low mechanical strength of β -TCP bioceramics restrict their application in the load-bearing bone defects.

Treatment of load-bearing bone defects usually selects biocompatible metals (titanium and its alloy, stainless steel, cobalt-based alloys, etc.), because of their high strength and ductility [8]. Unfortunately, a severe stress shielding often occurs to metal implants on account of their non-degradable characteristic, and significantly higher elastic modulus compared to that of natural bone [9]. The stress shielding

leads to a decrease of bone mass, because of which the metallic implants are removed by a second surgery [10]. The advent of degradable metals, especially magnesium and iron (Fe) and their alloys, holds a promise for better treatment of bone defect in the load-bearing region [11]. Plentiful investigations have been focused on magnesium alloy due to its non-toxicity and mechanical similarity to human bone [12]. However, magnesium alloy is characterized by high degradation rate in the body fluid environment and formation of hydrogen during the magnesium corrosion process; this may cause tissue necrosis, cell apoptosis and formation of cavity [13]. In contrast to magnesium and its alloy, pure iron and its alloy possess much lower degradation rate, higher strength and fracture toughness [14,15]. Furthermore, pure iron and its alloy are free from hydrogen evolution and solution alkalization during the degradation process. Iron is an important element in the human body. A majority of investigations have confirmed that pure iron and its alloy did not have adverse effect on the cell reaction and tissue response [16,17]. Pure iron and its alloy have been proposed as potential load-bearing implants, such as bone pins, screws, plates and substitutes [18]. However, the degradation rate of pure iron is $< 300\text{ }\mu\text{m/year}$ [19]. By alloying with other elements (Mn, C, Si, Pt), the degradation behavior of iron can be accelerated;

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nevertheless, the efficiency in expediting degradation was limited [11,17]. The nature of slow degradation is the hindrance of advancing iron-based implants. Furthermore, pure iron and its alloy are devoid of bioactivity and osteoconductivity, which are the essential criterions for bone substitutes [19].

Mixtures of metal and ceramic, namely cermets, are endowed with the characteristic of both ceramic and metal [20]. Traditional cermets are mainly used as cutting tool, structural part of aircraft, and chemical machinery, etc. Cermets are accepted in the biomaterial field as well. Most bio-cermets are based on ductile metal, supplemented with bioceramic coating or particulates which are contributive to bioactivity [21]. For iron-based bio-cermets, the presence of bioceramic particles (calcium phosphate and calcium silicate, etc) not only contributes to bioactivity but also accelerates degradation of iron [19,22]. To date, the bio-cermets based on bioceramic are not extensively reported. Chu et al. and Zhang et al. prepared HA-based bio-cermets using silver and titanium as second phases, respectively; they found that the metal phases were considerably contributive to the toughness of HA-based bio-cermets [23,24]. Due to non-degradation of metal phases and slow degradation of HA, those HA-based bio-cermets were hardly resorbable, which restricted their wide clinical application. Considering the faster degradation of β -TCP, excellent mechanical properties and degradability of iron, in this study we aimed to fabricate Fe/ β -TCP bio-cermets based on β -TCP, with iron particles being reinforcement phases. The phase composition, microstructure, porosity, and mechanical properties and in vitro cell behaviors of Fe/ β -TCP bio-cermets were investigated.

2. Materials and methods

2.1. Materials

Iron powders (99.5% purity, $d_{50}=35\ \mu\text{m}$) and paraffin were purchased from Aladdin Bio-Chem Technology Co., Ltd (Shanghai, China). $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$, $\text{NH}_3 \cdot \text{H}_2\text{O}$, $(\text{NH}_4)_2\text{HPO}_4$, and heptane were purchased from Tianjin Fuchen Chemical Reagent Factory, China. All the reagents were analytically pure except specialized. The cell-culture related reagents were obtained from Gibco except specialized.

2.2. Synthesis of β -TCP powders and preparation of Fe/ β -TCP bio-cermets

β -TCP powders were synthesized by chemical reaction between $\text{Ca}(\text{NO}_3)_2$ and $(\text{NH}_4)_2\text{HPO}_4$. In brief, 500 mL $\text{Ca}(\text{NO}_3)_2$ solution (0.6 M) was added to 500 mL $(\text{NH}_4)_2\text{HPO}_4$ solution (0.4 M). During the reaction process, the pH value of suspension was kept at around 7 by the addition of $\text{NH}_3 \cdot \text{H}_2\text{O}$ dropwise under the stirring condition. The obtained precipitates were washed with deionized water three times, then freeze-dried. The β -TCP powders ($d_{50}=2.1\ \mu\text{m}$) were obtained. Iron/ β -tricalcium phosphate (Fe/ β -TCP) bio-cermets were prepared using the powder metallurgy method. Paraffin was used as a binder. 12% (w/w) paraffin solution was prepared by dissolving paraffin in the heptane. The iron and β -TCP powders were dispersed in the paraffin solution, then grinded in a miller (QM-2SP20, Nanjing University Instrument Factory, China) for 2 h, air-dried at $60\ ^\circ\text{C}$ for 6 h to eliminate the heptane. The mass fraction of paraffin in the dried mixtures was 2.5%. The volume fraction of iron in the mixtures of iron and β -TCP was 0%, 5%, 10%, 20%, 30% and 40%, respectively. The dried mixtures of β -TCP, iron, and paraffin were poured into a mold, then prepressed using a powder pressing machine (FLS, Taizhou Rongmei, China) under a pressure of 5 MPa. The obtained pellets (50 mm $\times$ 50 mm $\times$ 8 mm) were subjected to isostatic pressing with a cold isostatic pressing machine (LDJ100/320-300, Western Sichuan Machinery Co., Ltd, China) under a pressure of 200 MPa at room temperature for 2 min. In order to eliminate the paraffin binder, the green bodies were heated in a tube furnace under the argon atmo-

Table 1

RT-PCR primer sequences used in this study.

Gene	Primer sequences
ALP	Forward: 5'-TGCCTACTTGTGTGGCGTGAA-3' Reverse: 5'-TCACCCGAGTGGTAGTCACAATG-3'
Col I	Forward: 5'-ATGCCGCGACCTCAAGATG-3' Reverse: 5'-TGAGGCACAGACGGCTGAGTA-3'
OCN	Forward: 5'-AGCAGCTTGGCCAGACCTA-3' Reverse: 5'-TAGCGCCGAGTCTGTTCACCTAC-3'
OPN	Forward: 5'-TGCAAACACCGTTGTAACCAAAAGC-3' Reverse: 5'-TGCACTGGCCGTTTGCATTTCT-3'
GAPDH	Forward: 5'-TGTGTCCGTCGTGGATCTGA-3' Reverse: 5'-TTGTCTTTGAAGTCGACAGGAG-3'

sphere, with a heating speed of $2\ ^\circ\text{C min}^{-1}$ up to $420\ ^\circ\text{C}$. After being held at $420\ ^\circ\text{C}$ for 2 h, the samples were cooled down in the furnace naturally. Subsequently, the debound samples were heated in the argon atmosphere with a heating speed of $2\ ^\circ\text{C min}^{-1}$, and sintered at $1120\ ^\circ\text{C}$ for 2 h, then cooled down in the furnace naturally. Finally, the Fe/ β -TCP bio-cermets were obtained. The Fe/ β -TCP bio-cermets with 5, 10, 20, 30 and 40 vol% iron were designated as 5Fe/ β -TCP, 10Fe/ β -TCP, 20Fe/ β -TCP, 30Fe/ β -TCP, and 40Fe/ β -TCP, respectively.

2.3. Materials characterization

2.3.1. Phase composition and microstructure observation

The Fe/ β -TCP bio-cermet samples were crushed into powders for phase analysis. The phase composition of powdered samples was determined by an X-ray diffractometer (XRD; X'Pert PRO, PANalytical Co., Netherlands) employing Cu-K α radiation (40 kV, 40 mA). Data were collected from 10° to 70° for 2θ with a step size of 0.0166° . Microstructure of the bio-cermet samples was observed under a field emission scanning electron microscope (SEM; Nava NanoSEM 430, FEI, Netherlands). Before SEM observation, the samples were mounted on an aluminum stub by carbon tape and then sputter-coated with gold. Accelerating voltages of 15 kV were employed.

2.3.2. Porosity measurement

Porosity of the samples was measured by the Archimedes method. The weight of dry samples was recorded as W_1 . The samples were immersed in deionized water under vacuum environment for 2 h, then hung in the water with a fine wire. The weight of the suspended samples in the water was recorded as W_2 . The samples were taken out from the water, and their surface was wiped with a saturated water sponge. The samples filled with water in air were recorded as W_3 . The porosity (P) was calculated as follows:

$$P = \frac{W_2 - W_1}{W_2 - W_3}$$

2.3.3. Mechanical test

The samples were cut into bars with geometry of 5 mm $\times$ 5 mm $\times$ 10 mm and 3 mm $\times$ 4 mm $\times$ 35 mm, which were used for measurement of compressive strength and fracture toughness, respectively. The compressive strength of the bio-cermet samples was measured using a universal material testing machine (Instron 5567, Instron, USA) at a crosshead speed of 0.5 mm/min. The fracture toughness of bio-cermet samples was determined by the single-edge notched beam (SENB) method under a universal material testing machine. The span dimension was 30 mm and the crosshead speed was 0.05 mm/min. Before measurement, a notch (depth ~ 2.5 mm; width ~ 0.2 mm) was produced on the surface of bar sample using a diamond wheel. The fracture toughness (K_{IC}) was calculated employing the ASTM C-1421 standard.

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