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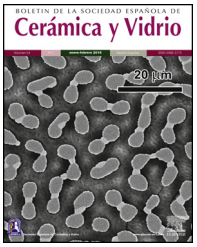

Characterization of β-tricalcium phosphate powders synthesized by sol–gel and mechanosynthesis

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ABSTRACT

β-Tricalcium phosphate (β-TCP) is one of the most investigated calcium phosphates to be used in bone tissue regeneration. Nowadays, β-TCP has been synthetized by different methods that allow obtaining different morphologies with chemical compositions similar to the bone tissue, broadening the possibilities of use. In this work, β-TCP powders were obtained using two different routes: mechanosynthesis and sol–gel. Results shown that both methods had significant effects on morphology and size particle of the obtained powders. The average size particles obtained using mechanosynthesis at 24 and 12 h of milling time were 1000 and 170 nm, respectively. The average size particles of the powders fabricated using sol–gel was 350 nm, and the average powder particle size of the sigma reagent was 1450 nm. Besides, sol–gel and sigma reagent powders were observed residual phases, instead mechanosynthesis shown only tricalcium phosphate beta phase. The formation of agglomerates with different sizes was observed in both methods by sol–gel and mechanosynthesis.

Caracterización de polvos de fosfato tricálcico fase β sintetizados por sol-gel y mecanosíntesis

Palabras clave:
Sol-gel
Tamaño de partícula

RESUMEN

El fosfato tricálcico fase β (β-TCP) es uno de los fosfatos de cálcico más investigados para el uso de la ingeniería de tejidos óseos. Hoy en día, el β-TCP ha sido sintetizado por diferentes métodos, que han permitido obtener diversas morfologías con composiciones químicas similares a las del tejido óseo ampliando las posibilidades de uso.

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Fosfato tricálcico fase β
Polvos Molienda

En este trabajo se obtuvieron polvos de β-TCP usando dos rutas diferentes: mecanosíntesis y sol-gel. Los resultados mostraron que ambos métodos presentan efectos significativos sobre la morfología y el tamaño de partícula de los polvos de fosfato tricálcico. Las partículas de tamaño promedio obtenidas por la ruta de mecanosíntesis con condiciones de 12 y 24 h presentaron valores de 1000 y 170 nm, respectivamente. Las partículas de tamaño promedio de los polvos fabricados por sol-gel fueron de 350 nm y el tamaño promedio del reactivo sigma fue de 1450 nm. Además, se observó que los polvos obtenidos por sol-gel y el reactivo sigma presentaron fases residuales, en cambio por mecanosíntesis mostró solo fosfato tricálcico fase beta. La formación de aglomerados se observó en ambos métodos tanto en sol-gel como en mecanosíntesis.

Introduction

Tricalcium phosphate (TCP) is one of the variations of the calcium phosphate compounds with more applications in bone tissue regeneration [1–3], due to its chemical composition similar to the natural bone tissue [4]. Different synthesis routes can produced TCP, e.g., mechanosynthesis, wet methods, microwave irradiation, sol–gel, etc. [5–8]. Each of these methods provides specific characteristics to the TCP, such as particle size, mechanical properties, morphology and crystalline structures. Among them, mechanosynthesis and sol–gel procedures present the best performance taking into account particles size and homogeneity. These two methods are controlled by variables that modify the powders feature: in the case of mechanosynthesis, the powders can be changed by the reagents selection, ball/mass ratio, powder/mass ratio, wet or dry milling, milling time, rotational speed, etc. On the other hand, variables to consider in the sol–gel method are pH, reagents, aging temperature, and calcination. Then, the resulting particle sizes obtained by sol–gel and mechanosynthesis can measure from nanometers to some microns in size, also broadening the range of properties such as chemical reactivity, dissolution and mechanical properties [5,9]. It has been reported that using these synthesis techniques it can be obtained high purity and crystalline phases, as a consequence of the chemical composition of the reagents [6,8,10,11]. Then, it is essential to correlate the synthesis mechanisms and their variables with the final characteristics and properties of the obtained materials. Therefore, the objective of the present work was to achieve a detailed characterization of tricalcium phosphate powders by mechanosynthesis and sol–gel methods. Besides, to compare the sigma reagent tricalcium phosphate with the powders manufactured with the procedures already mentioned.

Materials and methods

Powders preparation

Two different processes were performed for obtaining TCP powders with beta phase: (i) sol–gel, (ii) mechanosynthesis at 12 and 24 h at 350 rpm. β-TCP Sigma–Aldrich reagent with a purity of 96% was used as a reference parameter to compare with sol–gel and mechanosynthesis powders. The nomenclature used for this research was 12 M and 24 M for the milling time of 12 and 24 h respectively. The experimental conditions for each procedure are specified below.

Mechanosynthesis method

All the reagents used in this research were from Sigma–Aldrich: calcium carbonate ($CaCO_3$), and calcium dibasic phosphate ($CaHPO_4$), both of them with a purity higher than 98%. The mechanical activation was achieved in a Retsch PM400 planetary ball mill. Calcium carbonate and calcium dibasic phosphate powders were mixed in a roller mill for 30 min (Ca/P = 1.5). Furthermore, the ratio of ball-to-powder was of 8:1. The mechanosynthesis was carried out for 12 and 24 h with a speed of 350 rounds per minute (rpm). Finally, the beta phase (β) of the TCP was obtained through heat treatment at 900 °C for 3 h and then, it was cooled with a speed of 3 °C per minute to room temperature in a Thermolyne FB-1315 M benchtop muffle furnace.

Sol–gel method

For this case, the reagents were also from Sigma–Aldrich, calcium nitrate tetrahydrate: $Ca(NO_3)_2{\cdot}4H_2O$, citric acid monohydrate $C_6H_8O_7{\cdot}H_2O$, and diammonium phosphate $(NH_4)2HPO_4$. The amounts used were 0.0926 mol $Ca(NO_3)_2{\cdot}4H_2O$, 0.0926 mol $C_6H_8O_7{\cdot}H_2O$ and 0.0617 mol of $(NH_4)2HPO_4$. The three components were dissolved in 100 mL of water and agitated for 30 min. The pH was maintained around 2 adjusting with nitric acid (HNO_3) dropwise. The resulting product was vaporized at 80 °C until appearing a transparent gel. Subsequently, the gel was heated at 100 °C to eliminate rest of water [12]. The TCP xerogel was heated at 900 °C for 3 h and then, it was cooled with a speed of 3 °C per minute to room temperature.

Powders characterization

The tricalcium phosphate powders obtained by mechanosynthesis at 12, and 24 h, sol–gel and β-TCP Sigma–Aldrich powders were characterized by different techniques. Crystalline structures were analyzed using a D500 X-ray Diffractometer (XRD) with Cu Kα radiation at 30 kV and 25 mA in the 2θ interval from 20° to 40°, at scanning speed of 0.2°/min. Diffraction patterns were analyzed using EVA V4.2.1

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