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Development and characterization of carbonate apatite/β-tricalcium phosphate biphasic cement

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ABSTRACT

Biphasic calcium phosphate consisting of carbonate apatite and β-tricalcium phosphate (β-TCP) is an excellent bone substitute with controllable bioresorbability. Fabrication of biphasic calcium phosphate with self-setting ability is expected to enhance the potential of applicability as bone substitute. In this study, mixtures of α-TCP/β-TCP powder after carbonation period 1, 3, and 5 h with 1.0 M $NaHCO_3$ were prepared. These powders were mixed with 0.25 M Na_2HPO_4 at a P/L ratio of 2, and then hardened at 37 °C and 100% RH up to 24 h. XRD patterns and FT-IR spectrum showed that the resulting hardened cement consisted of biphasic carbonate apatite and β-TCP. Diametral tensile strength of the cement decreased with the carbonation period.

1. Introduction

For bone regeneration, autografts are preferred for faster bone regeneration; however, this method encounters many challenges inherent to the process, such as the invasiveness of extraction of healthy bone, as well as a limited amount of starting material for the graft [1]. Calcium phosphate ceramics have been one of the most valuable artificial bone substitutes due to similarities of their crystalline structure and chemical composition to those of the inorganic phase of bone [2]. In addition, they have good biocompatibility and the ability of new bone formation [2,3]. Most of cements based on calcium phosphate ceramics set into hydroxyapatite (HA, $Ca_{10}(PO_4)_6(OH)_2$) [4–6]. In general, apatite existing in bone is not stoichiometric HA but carbonate apatite (CO_3Ap). Legeros et al. demonstrate that the solubility of apatite increases in accordance with the increase in the carbonate content in apatitic structure [7]. CO_3Ap, therefore, is high solubility than HA in a body environment. In the past study, carbonate apatite was fabricated by sintering the mixture of $CaHPO_4$ and $CaCO_3$ at 870 °C in a CO_2 [8]. However, there are few studies of calcium phosphate cement focusing on the self-setting ability forming CO_3Ap as a final product [9,10]. Matsuya et al. demonstrate that self-setting carbonate apatite is fabricated by using α-TCP [9]. In our previous study, it was successful to fabricate biphasic HA/β-TCP cement using heat-treated α-TCP powder as a main composition of cement due to its self-setting ability [11]. α-TCP/β-TCP powder with various content ratio was fabricated by using α-β phase transition of α-TCP powder at 1000 °C for various times. In the case of HA/β-TCP cement, α-TCP hydrolyzes to apatite, on the other hands, β-TCP is unreacted and embedded in the apatite matrix [3–5,11]. It is probable that the self-setting CO_3Ap/β-TCP cement was fabricated by means of α-TCP/β-TCP powder with carbonate ion.

Therefore, the primary aim of this study was to fabricate and to characterize a biphasic CO_3Ap/β-TCP cement. The cement powder was fabricated by using α-TCP/β-TCP powder was treated in 1.0 M $NaHCO_3$. the material characteristics, such as composition, morphology and mechanical property, were investigated.

2. Materials and methods

2.1. Preparation of cement powder

α-TCP/β-TCP mixed powder was prepared by using thermal treatment of α-TCP powder described previously [11]. Briefly, α-TCP (α-TCP-b: α-$Ca_3(PO_4)_2$) (Taihei Chem., Osaka, Japan) powder was heated at 1000 °C for 5 h to allow partial conversion of α-TCP to β-TCP [11]. In this condition, the conversion of α-TCP to β-TCP was the ratio of 40%. The size of α-TCP/β-TCP powder was from 5 to 10 μm. Ten grams of α-TCP/β-TCP powder were treated in 500 ml of 1.0 M $NaHCO_3$ (Wako Chem. Co., Osaka, Japan) for 1, 3 and 5 h at room temperature to introduce carbonate ion in the powder. After this treatment, the slurry was filtered under reduced pressure and was dried in an oven at 50 °C for 3 days. The

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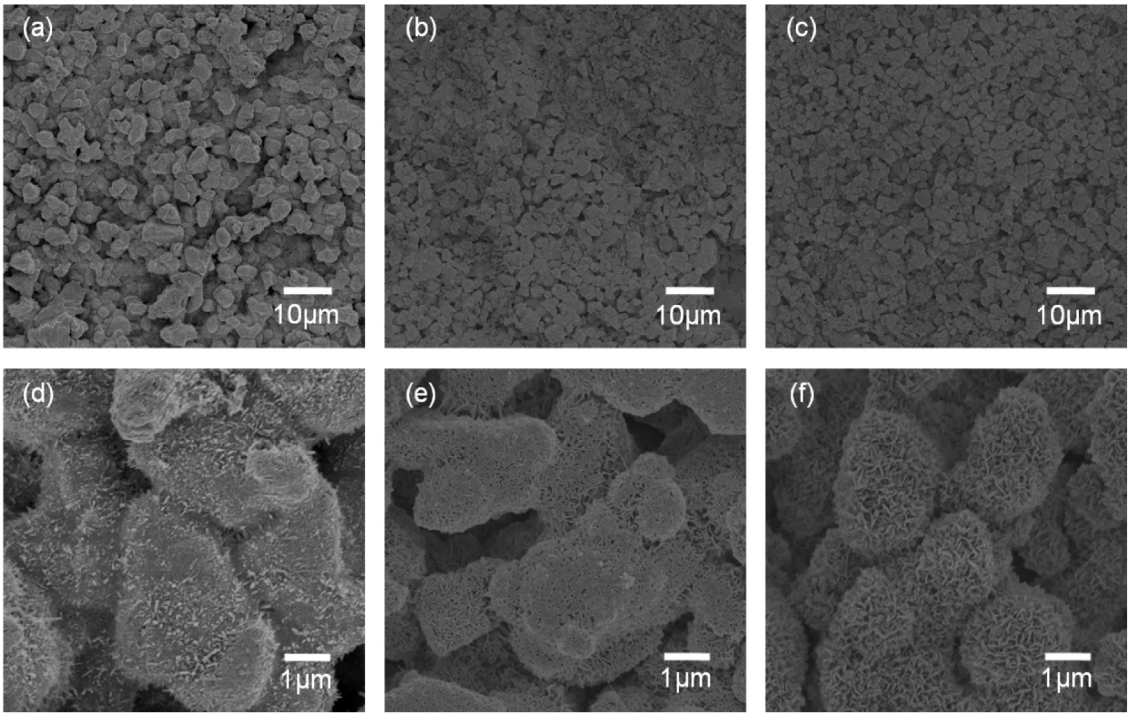

Fig. 1. FE-SEM micrographs of the surface of the CO_3Ap/β-TCP cement set for 24 h using CT1 (a, d), CT3 (b, e) and CT5 (c, f), respectively.

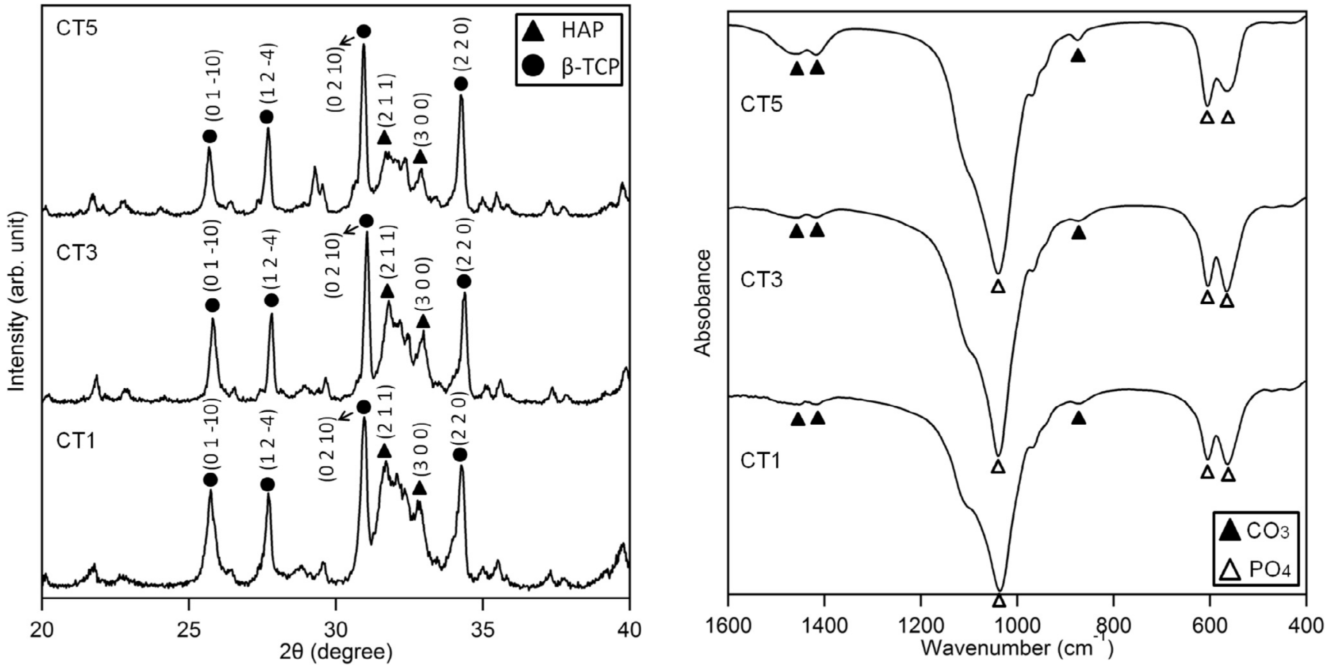

Fig. 2. XRD pattern and FT-IR spectra of the CO_3Ap/β-TCP cement set for 24 h.

α-TCP/β-TCP powder carbonated for 1, 3 and 5 h are hereafter identified as CT1, CT3 and CT5, respectively.

2.2. Fabrication of biphasic β-TCP/carbonate apatite cement

α-TCP/β-TCP powder obtained was mixed with 0.25 M Na_2HPO_4 solution at powder to liquid (P/L) ratio of 2 g/ml and packed into a stainless steel mold (6 mm in diameter and 3 mm in thickness), both ends of the mold were clamped with glass plates. The samples were placed into an incubator (SIB-35, SANSYO Co., LTD, Tokyo, Japan) kept at 37 °C and 100% relative humidity for 24 h.

2.3. Characterization of biphasic β-TCP/carbonate apatite cement

The set cements were ground into fine powders and characterized by X-ray powder diffraction (XRD) analysis. The XRD patterns of the cements were recorded with a vertically mounted diffractometer system (MiniFlexII Rigaku, Tokyo, Japan) using