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ORIGINAL ARTICLE

Physicochemical properties and biocompatibility of silica doped β-tricalcium phosphate for bone cement

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Abstract *Background/purpose:* β-Tricalcium phosphate (β-TCP) is an osteoconductive material. Earlier reports revealed that silica plays an important role in bone mineralization, and its incorporation would enhance the biocompatibility of implants.
Materials and methods: In this study, silica doping at various concentrations (0 wt.%, 10 wt.%, 20 wt.%, and 30 wt.%) was performed. Si-â-TCP was obtained upon calcining the as-prepared powders at 1400°C. To check its effectiveness, different Si-â-TCP samples were prepared to make new bioactive and biodegradable biocomposites for bone repair. Formation of bonelike apatite, the diametral tensile strength, ions released, and weight loss of composites were considered before and after immersion in simulated body fluid. We also examined the behavior of human dental pulp cells (hDPCs) cultured on these materials.
Results: The results showed that the apatite deposition ability of the β-TCP was enhanced as the Si content was increased. For composites with >20% Si content, the apatite layer covered the samples. At the end of the immersion point, weight losses of 52%, 45%, 33%, and 26% were observed for the β-TCP containing 0%, 10%, 20%, and 30% Si, respectively. *In vitro* cell experiments showed that the Si-rich cement promotes hDPC proliferation and differentiation. However, when the Si content in the cement is >20%, the amount of cells and osteogenesis protein of hDPCs were stimulated by Si released from Si-β-TCP.
Conclusion: The degradation of β-TCP and osteogenesis of Si give us a strong reason to believe that these Si-based cements may prove to be promising bone repair materials.

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Introduction

The suitable bioactivity and appropriate degradation of materials are required to conform to different clinical requirements for hard tissue repair. Autograft possesses all the characteristics indispensable for new bone formation, osteogenesis, osteoconductivity, and osteoinductivity, and it is currently considered the gold standard in this field. In spite of its strengths, however, there are still various disadvantages when using this material for certain medical clinical applications. This approach regrettably minimizes reconstructive healing and, even worse, sometimes promotes foreign body reaction. Therefore, efforts are ongoing to design better biomaterials that allow good control of material physicochemical properties.

Silicate (Si)-based cements have received a considerable amount of positive attention in recent years because these materials have a better bioactivity than calcium phosphate-based materials.[1,2] Recently, several studies have indicated that Si-based materials can play an important role in bone formation, at least based on these materials' Si ion release[3] and fast apatite formation ability.[4,5] Si-based materials can promote human mesenchymal stem cells, human dental pulp cells (hDPCs), and osteoblast-like cell adhesion, proliferation, and differentiation.[6–11] Furthermore, the Si-based cement was able to activate angiogenesis[12] and antiosteoclastogenesis.[13] These studies demonstrate that Si-based cement has the ability to promote the apatite layer to form *in vitro*[14] and also shows it is better at promoting bone regeneration *in vivo*.[15] However, the slow degradation rate of Si-based materials may result in a decrease in osteoconductivity, which may restrict their application in clinical practice.[16] In order to ameliorate its relative disadvantage in regard to material degradation, we used β-tricalcium phosphate (β-TCP) as an additive to see how this would affect its rate of decay. β-TCP is a bioceramic material that is widely used for hard tissue repair. It has a chemical composition similar to apatite present in bone tissue, and has been applied extensively as a bone grafting material.[17,18] Previous studies have shown that wollastonite-doped TCP bioceramic has a higher degradation rate and promotes new bone formation better than TCP *in vivo*.[15] Wang et al[19] assert that the composite scaffolds containing50% and 80% calcium silicate not only have good osteoconductivity, but also promote rapid bone formation compared with pure β-TCP and calcium silicate scaffolds.

Thus, to obtain both osteostimulation and osteoconductivity by taking advantage of the favorable bioactivity of silica and the high degradability of β-TCP, Si-doped β-TCP cements have been produced in the hopes that the right mixture can help to control the degradation rate and improve interactions of the material with human tissue. In this study, Si-doped β-TCP cements with varying ratios were prepared so that we could observe the changes in physiochemical properties, bioactivity, *in vitro* degradation behavior, osteogenesis, and antibacterial activity with different ratios of Si doped. It is our hope that this knowledge may help in the design of optimal biomaterials for bone regeneration.

Materials and methods

Preparation of Si-doped β-TCP cement

The reagent grade SiO_2 (Sigma-Aldrich, St. Louis, MO, USA) and β-TCP (Sigma-Aldrich) powders were used as matrix materials. The SiO_2 and β-TCP mixtures were sintered at 1400°C for 3 hours using a high-temperature furnace, and then ball-milled in ethyl alcohol using a centrifugal ball mill (S 100; Retsch, Hann, Germany) for 6 hours. The specimen codes "Si10", "Si20" and "Si30" were used to indicate cement containing 10 SiO_2/90 β-TCP, 20 SiO_2/80 β-TCP, and 30 SiO_2/70 β-TCP (in wt.%), respectively. Si0 is the β-TCP group. The powder was mixed using a liquid/powder ratio of 0.25 mL/g. After mixing with liquids (1M Na_2HPO_4), the cements were molded in a Teflod mold (diameter, 6 mm; height, 3 mm). The cement quantities were such they fully covered each well of the 24-well plate (GeneDireX, Las Vegas, NV, USA) to a thickness of 2 mm for cell experiments. All samples were stored in an incubator at 100% relative humidity and 37°C for 1 day of hydration.

Setting time and strength

After the powder was mixed with liquid, the cement was placed into a cylindrical mold and stored in an incubator at 37°C and 100% relative humidity for hydration. The setting time of the cements was tested according to the standards set by the International Standards Organization 9917-1. The setting time was recorded when the Gilmore needle failed to create a 1-mm-deep indentation in three separate areas.

After being taken out of the mold, the composite specimens were incubated at 37°C in 100% humidity for 1 day. Diametral tensile strength (DTS) testing was conducted on an EZ-Test machine (Shimadzu, Kyoto, Japan) at a loading rate of 1 mm/minute. The maximal compression load at failure was obtained from the recorded load–deflection curves. At least 10 specimens from each group were tested.

In vitro soaking

To evaluate the *in vitro* bioactivity, the cements were immersed in a 10-mL simulated body fluid (SBF) solution at 37°C. The solution in the shaker water bath was not changed daily under a static condition. After soaking for different time durations (from 3 days to 3 months), specimens were removed from the tube and evaluated for several physicochemical properties.