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# Fabrication and evaluation of silica-based ceramic scaffolds for hard tissue engineering applications

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ABSTRACT

In recent decades, bone scaffolds have received a great attention in biomedical applications due to their critical roles in bone tissue regeneration, vascularization, and healing process. One of the main challenges of using scaffolds in bone defects is the mechanical strength mismatch between the implant and surrounding host tissue which causes stress shielding or failure of the implant during the course of treatment. In this paper, space holder method was applied to synthesize diopside/forsterite composite scaffolds with different diopside content. During the sintering process, NaCl, as spacer agent, gradually evaporated from the system and produced desirable pore size in the scaffolds. The results showed that adding 10 wt.% diopside to forsterite can enormously improve the bioactivity, biodegradability, and mechanical properties of the composite scaffolds. The size of crystals and pores of the obtained scaffolds were measured to be in the range 70–100 nm and 100–250 μm, respectively. Composite scaffolds containing 10 wt.% diopside showed similar compressive strength and Young's modulus (4.36 ± 0.3 and 308.15 ± 7 MPa, respectively) to that of bone.

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## 1. Introduction

Bone tissue engineering offers an alternative approach to treat and repair bone defects by stimulating bone formation from the surrounding tissue [1]. Cells, scaffold and growth factors are three important elements in tissue engineering which depend on the features of the tissue at the location of the defect [2]. Scaffolds are considered as biological substitutes in bone defects which improve and accelerate the healing procedure of surrounding tissue [3]. Based on the location of defects and the desired function, scaffolds with different mechanical and biological properties have been developed; however, for load bearing applications, high mechanical properties as well as a controlled biodegradability are required [4–7]. Scaffolds with interconnected macroporosity have osteal structure (such as trabecular bones) which can provide an appropriate environment for cell migration, vascularization, and diffusion of nutrients [8]. It is of a great interest to design scaffolds with pore diameters of a hundred microns with bioresorption capability to stimulate the bone ingrowth and simultaneously degrade in situ [9].

Previous studies have demonstrated that bioceramics including $CaO$-$MgO$-$SiO_2$ groups have excellent mechanical and biological properties in comparison with hydroxyapatite (HA) [10]. Magnesium, silicon and calcium elements play crucial roles in human body. Silicon is an essential element in skeletal development [11]. The findings showed that silicon (>5 wt.%) was uniquely localized in the active growth areas in the bones of young rats (when the Ca/P ratio was 0.7) and involved in the early stage of bone calcification in physiological conditions [12]. Magnesium is closely associated with mineralization of calcified tissues and indirectly influences mineral metabolism [13]. It is worth noting that Mg can motivates osteoblast proliferation with using of insulin-like growth factor [14]. Calcium is also a well-known element which is needed to build and maintain strong bones [10].

Forsterite ($Mg_2SiO_4$) is one of the interesting ceramics for biomedical applications. Nanostructure forsterite has been introduced as a biocompatible and bioactive bioceramic with high mechanical strength in compression to HA [15]; however, micron size forsterite is not bioactive and shows low biodegradability [16–17]. In order to synthesize forsterite scaffolds with a high compressive strength, sintering the scaffolds at high temperatures is required. On the other hand, holding the samples at high temperatures decreases the pore size of the scaffolds and increases grain growth which inversely affect the vascularization and bioactivity of the material, respectively. Many studies have been performed to overcome these problems.

Kharaziha et al. [18] used two-step sintering method to prepare dense nanostructure forsterite. In this technique, the grain growth was controlled and the produced forsterite had a high compressive strength. Ni et al. [19] developed a β-$CaSiO_3$/forsterite nanocomposite which showed higher cell proliferation and degradability in comparison to pure micron sized forsterite. It was found that utilizing secondary phases can be helpful in the sintering process and providing higher mechanical strength. The presence of a secondary phase can decrease the

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required sintering time and temperature. As a result, less grain growth happens during sintering and grains remain in a nano-scale.

Diopside ($CaMgSi_2O_6$) is a member of pyroxene family in the $CaO$-$MgO$-$SiO_2$ system. Recently, diopside has received a great attention as an alternative bioceramic for biomedical applications. It has been proved that diopside is nontoxic, biocompatible and bioactive, which helps proliferation and differentiation into different pathways. Formation of apatite on the surface of diopside just after 3 days shows its higher bioactivity compared to forsterite [20–21].

Different procedures have been developed to synthesize single-phase diopside and forsterite scaffolds such as foaming method [22–23], gel-casting method [9,24] and space holder method [25]. Space holder method is of a great interest to many scientists due to the capability of this method in providing interconnected porosities with controlled size and appropriate mechanical properties [26]. Furthermore, bio-glass and bio-ceramics were produced by the space holder method with different porogen agents such as polyethylene, rice husk, ammonium nitrate, sucrose, gelatin and sodium chloride [27].

In this study, for the first time, the potential of space holder method as a new technique to fabricate diopside/forsterite scaffolds was studied. Moreover, the effects of adding various diopside content on mechanical, physical, size of crystals, bioactivity and biodegradability of forsterite ceramic were investigated as well. Diopside acts as a sintering aid and decreases the required sintering temperature and subsequently reduces grain growth which occurs during the sintering process. Also it has a higher bioactivity compared to forsterite and therefore it can provide a biological fixation in a shorter period of time which prevents the movement and failure of the implant.

## 2. Experimental

### 2.1. Preparation of forsterite nanopowder

Mechanical activation (MA) method was used to produce the forsterite nanopowder. Talc ($Mg_3Si_4H_2O_{12}$, 98% purity, Merck) and magnesium carbonate ($MgCO_3$, 99% purity, Aldrich) powders were selected as initial materials. In order to produce forsterite nanopowder, $MgCO_3$, and Talc powders were mixed in a high energy ball mill machine utilizing zirconia vial and balls with the diameter of 2 cm. Talc and $MgCO_3$ with the molar ratio of 5:1 were mixed to produce stoichiometric forsterite powder. The ball to powder weight ratio was 10:1 and the rotational speed of the main disc was 300 rpm. The time of MA was 15 h. The ball milled powders was sintered at 1000 °C for 1 h to produce forsterite nanopowder [28–31]. The obtained forsterite powder was called NF.

### 2.2. Preparation of diopside nanopowder

In order to synthesize diopside nanopowder, a combination of sol-gel and MA was used. Magnesium chloride hexahydrate ($MgCl_2 \cdot 6H_2O$, 98% purity, Merck), calcium nitrate tetrahydrate ($Ca(NO_3)_2 \cdot 4H_2O$, 99% purity, Merck), tetra ethyl orthosilicate (TEOS) ($(C_2H_5O)_4Si$, 99% purity, Merck) and ethanol ($C_2H_5OH$, 99.9% purity, Merck) were used as the starting materials. First, calcium and magnesium nitrate powders with the molar ratio of 1:1 were dissolved in 150 ml ethanol and then stirred for 30 min at 80 °C. Subsequently, TEOS was added to the solution with the molar ratio of Si:Mg equal to 2:1 and stirred for 24 h. The resulting gel was maintained at 100 °C until it was dried completely. The dried gel was sintered at 800 °C for 2 h. Finally, the powder was milled for 5 h with the rotational speed of 250 rpm and the ball to powder weight ratio of 10:1 [32]. The obtained diopside powder was called ND.

### 2.3. Preparation and characterization of diopside/forsterite scaffolds

Diopside/forsterite scaffolds were prepared by the space holder method. In this method, sodium chloride (NaCl, 99.9% purity, Merck) (300–420 mesh size) was used to prepare the scaffolds with the interconnected porosity. Complete specifications of scaffolds are shown in Table 1. Forsterite and diopside powders were mixed with 85 wt.% of NaCl as the spacer and mixed together with an amalgamator. Subsequently, the powders were uniaxially pressed at the pressure of 65 MPa by a universal testing machine (HOUNSFIELD: H50KS). Finally, the obtained specimens were sintered at 1200 °C for 3 h with the heating/cooling rate of 3 °C/min. The compressive strength and Young's modulus of the scaffolds with the height and diameter of 20 mm × 10 mm were evaluated by Hounsfield, H25KS instrument.

Phase evaluation was performed by X-ray diffractometry (XRD, Philips X′ pert) with Cu Kα radiation ($\lambda = 0.154$ nm at 40 kV and 30 mA). The XRD patterns were recorded in the 2θ range of 20–80° with the step size and time per step of 0.05° and 1 s, respectively. The size of crystals was measured based on Scherrer equation as follow [31]:

$$\beta \cos\theta = \frac{0.9\lambda}{D} \quad (1)$$

where $\theta$ is Bragg diffraction angle, D is the size of crystals, λ is the wavelength of the radiation, $\beta$ is the half of diffraction peak width, and 0.9 is the Scherrer constant. The morphology and the size of crystals of the scaffolds were evaluated by transmission electron microscopy (TEM, Philips EM208S with 100 kV operating voltage). The surface morphology of the scaffolds was investigated by scanning electron microscopy (SEM) in a Philips XL30 with the acceleration voltage of 30 kV. The porosity of scaffolds was measured according to the Archimedes principle as described in [33].

### 2.4. In-vitro test

This part of the study was carried out using the standard in vitro procedure described in Kokubo and Takadama [34]. The scaffolds with different weight percentages of diopside were soaked in a simulated body fluid (SBF) with pH = 7.38 for 1, 7, 14, and 21 days. All samples with the scaffolds to solution ratio equal to 1.5 mg/ml were kept in Ben Marry bath at the temperature of 37 °C. After soaking, the scaffolds were dried at 70 °C for 5 h.

The apatite formation ability on the surface of the scaffolds was evaluated by scanning electron microscopy (SEM), energy-dispersive spectroscopy (EDS) and Fourier transform infrared spectroscopy (FTIR, JASCO 680 PLUS). FTIR analysis was performed in the range of 400–4000 $cm^{-1}$. PH change of SBF was examined by a pH meter every 2 days. The atomic absorption spectrometer (AAS, 3030) was utilized to determine the concentration of Ca and P of the optimize scaffold after immersing in the solution.

The degradability of samples was evaluated in the phosphate buffered saline (PBS) solution at 37 °C in a soaking Ben Mary bath for 7, 14 and 21 days. The media was changed every 3 days. The samples were dried at 100 °C for 24 h. The weight loss of each specimen was expressed as a percentage of the initial weight.

### 2.5. Statistical analysis

All data were examined as average values ± standard deviation for n = 3. Statistical analysis was performed using GraphPad Prism. Data

**Table 1**
The specifications of diopside/forsterite nanocomposite scaffolds.

| Sample | Diopside (wt.%) | Forsterite (wt.%) | NaCl (wt.%) |
|---|---|---|---|
| FD | 0 | 15 | 85 |
| FD1 | 0.75 | 14.25 | 85 |
| FD2 | 1.5 | 13.5 | 85 |
| FD3 | 3 | 12 | 85 |
| FD4 | 6 | 9 | 85 |
| FD5 | 12 | 3 | 85 |

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