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On the mechanical and biological properties of bredigite-magnetite ($Ca_7MgSi_4O_{16}$-Fe_3O_4) nanocomposite scaffolds

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ABSTRACT

The aim of the present study was to study the mechanical and biologocal properties of the bredigite-magnetite ($Ca_7MgSi_4O_{16}$-Fe_3O_4) nanocomposite with various amounts of magnetite (0, 10, 20 and 30 wt%). According to the obtained results, the properties of the constructed scaffolds have an extreme dependence on the magnetite content. In this research, the bredigite-30 wt% magnetite as the optimum sample showed a fracture toughness of 2.69 MPa $m^{1/2}$ and a Young's modulus of 29 GPa. Increasing bredigite content led to the increase of pH values in the SBF solution. This was originated from the interchange/interaction of Ca^{2+} ion on the scaffold surface. The sample containing 10 wt% magnetite presented a rocky and irregular surface while that of 30 wt% illustrated a smooth and flat outer layer with coarse projections. The results confirmed that the biodegradation rate of the pure bredigite is more than that of 20 wt% sample. The event is originated from the dissolution of the Si ions of the bridigite particles in the absence of magnetite.

1. Introduction

Recently, various types of silicate bioceramics such as akermanite, diopside and baghdadite have been used in the biomaterials field for treating bone cancer disorder [1–4]. Several unique published papers showed that some of the glass ceramics which contain MgO, CaO, and SiO_2 have suitable biocompatibility and biological behaviour in comparison with calcium phosphate ceramics [5]. According to the literature [6], silicate bioceramics have a potential of chemical reactivity in the simulated body fluid for apatite formation on their surface. Calcium and magnesium as the main elements of the silicate bioceramics have the highest effect on the cells osteoblastic and the metabolism for the bone tissue engineering mechanism [6,7]. In addition, magnesium as an effective material in human's body can enhance the bone growth, for example.

An important silicate bioceramic named bredigite $Ca_7MgSi_4O_{16}$ was introduced for controlling the apatite formation as well as biodegradation rate of physiological saline [7–10]. Several researches illustrated that bredigite bioceramic may possesses proper bioactivity and apatite formation which make it a capable material for bone tissue implantations.

Wu et al. [10], produced pure bredigite powders via sol-gel technique. At a particular concentration of Ca, Si, and Mg ions dissolved in bredigite dissolution, the produced sample may stimulate the osteoblast proliferation [7]. Huang et al. [11], synthesized nanocrystalline bredigite powders via liquid combustion route. They showed that the produced bredigite powders had a range size of 230–460 nm with a relatively low synthesis temperature of 650 °C. According to their results, the apatite was formed on the surface of the bredigite scaffolds after soaking in the SBF solution for four days.

The old techniques for fabrication of scaffolds are freeze casting [12], gel casting [13], polymer sponge [14] and space holder [15,28]. In this research a bredigite-magnetite scaffold nanocomposite was fabricated via three-dimensional (3D) printing method. The effect of different various contents of magnetite on bioactivity and mechanical properties of the scaffold samples was investigated.

2. Material and methods

2.1. Preparation of bredigite nanoparticles

The bredigite nanopowders were synthesized using $Mg_3Si_4O_{10}(OH)_2$), (SiO_2) and $CaCO_3$ as starting materials. These materials were blended according to the stoichiometry content of the pure

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bredigite. Thereupon, the mixed powder was grinded in a planetary High Energy Ball Mill (HEBM) (Retsch, PM 100) for 10 h under ambient temperature. The milling media was consisted of zirconia vials and balls (d = 10–15 mm). In all grinding runs, the ball-to-powder weight ratio was set on 10:1 and the disk rotational speed was selected 650 rpm. The prepared milled powders were then kept in the furnace at 1300 °C for 4 h to complete bredigite synthesis.

2.2. Fabrication of magnetite nanoparticles

Magnetite nanoparticles (MNPs) were synthesized by the sol-gel method according to the previous research [16].

2.3. Preparation of bredigite-magnetite nanocomposite

A Bredigite-Magnetite nanocomposite containing various percentages of Fe_3O_4 (0 wt%, 10 wt%, 20 wt%, and 30 wt%) was fabricated using HEBM; with alcohol medium for obtaining a homogeneous combination. The grinding time was set on 60 min and the obtained powders were sintered at 1300 °C for 4 h to make homogenized nanocomposite.

2.4. Preparation of scaffolds nanocomposite

In this study, a nanocomposite scaffold was fabricated by three-dimensional printing (3DP) machine with cylindrical shapes. Scaffold prototypes with 12 mm height, 6 mm diameter, 0.8 mm pore size were designed using the 3D design software SolidWorks®2012. In this study, water was used as a binder (Zb63).

2.5. Fracture toughness evaluation

The fracture toughness and bending strength are estimated according to the previous practical experiments which were performed by Guazzato et al. [17]. Specimens with rectangular shape were designed (3 × 4 × 25 mm) to achieve three-point bending tests. Set points for the trial of bending strength were three-point tests which were conducted on a fracture test bench at with 0.5 mm/min speed using a load cell of 0.5 kN. The samples were polished with SiC paper (grits = 1200).

Finally, the bending strength was calculated using the following Eq. (1):

$$\text{Bending strength} = 3P_{max}L/2bh^2 \quad (1)$$

2.6. Compression strength evaluation

The compressive modulus is calculated from the first linear slope of the stress–strain curve. For measuring the compressive strength, the samples are designed in a cylindrical shape with dimensions of 6 mm × 12 mm (diameter × thickness) using a universal testing machine (ramp rate was 1 mm min^{-1}). Eq. (2) is used to estimate the compressive strength of the samples:

$$CCS(kg/cm^2) = F/\pi D^2/4 \quad (2)$$

Where D is the diameter of the sample and F is the applied force.

2.7. Porosity evaluation

The porosity of the nanocomposite scaffolds was calculated by Archimedes technique using following Eq. (3).

$$\text{Appearance Porosity} = \frac{(W2 - W1)}{W2 - W3)} \times 100\%, \quad (3)$$

Where W_1 is the weight of the scaffold nanocomposite in the air, W_2 is the weight of the samples in the water, and W_3 is the weight of samples suspended in water. The bulk density of both open and closed porosities was examined. Finally, the total of open porosities of the scaffold specimens were accurately reported.

Table 1
Compounds of the artificial blood, PBS, and SBF solutions.

Composition (g l^{-1})	Artificial blood	PBS	SBF
KH_2PO_4	–	0.2	–
NaCl	6.8	8.0	8.035
KCl	0.4	0.2	0.225
$CaCl_2{\cdot}H_2O$	0.2	–	–
$NaH_2PO_4{\cdot}H_2O$	0.026	–	–
$Na_2HPO_4{\cdot}H_2O$	0.126	–	–
$MgSO_4$	0.1	–	–
$NaHCO_3$	2.2	–	0.355
Na_2HPO_4	–	1.15	–
$MgCl_2{\cdot}6H_2O$	–	–	0.311
$K_2HPO_4{\cdot}3H_2O$	–	–	0.231
Na_2SO_4	–	–	0.072
$CaCl_2$	–	–	0.292
HCl (1.0 mol l^{-1})	–	–	39 ml
Tris-hydroxymethylamino	–	–	–
Methane	–	–	6.118

2.8. In vitro bioactivity studies

The bioactivity test in Simulated Body Fluid (SBF) is a technique that is used to evaluate the in vitro evaluation of materials. After immersion of the nanocomposite scaffold in SBF for a certain period of time (28 days), the bioactivity is evaluated by the amount of the apatite layer, which is formed on the surface of the samples. SBF was prepared according to procedure described by Kokubo [18]. Table 1 shows the elements and quantities used to prepare the SBF solutions.

2.9. Materials characterization

The size of porosities during the sintering process and apatite formation, were evaluated by scanning electron microscope (SEM). Samples were coated with gold (Au) for 3 min using spraying, in a high vacuum environment with a 40 kV accelerates voltage. The SEM was equipped with EDX microanalysis (FEI Quanta 200 ESEM) at Scientific Research Town Centre, at Isfahan University Technology) to measure Ca and P ions contents in order to estimate the rate of the apatite formation on the surface of samples. Inductive Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) instruments were applied for analyzing the experimental chemical changes.

3. Results and discussions

3.1. Phase and morphology characterization of sample

Fig. 1-a depicts the XRD patterns of the pure bredigite and bredigite-30 wt% magnetite composite samples. As can be seen in Fig. 1a, only bredigite peaks ($Ca_7MgSi_4O_{16}$, JCDP: 036–0399) are present in XRD pattern. As Fig. 1-b indicates the strain increases from 0 to 4 × 10^{-4}, while the lattice parameter showed a variated range of 0.43–0.68 (nm). At 1300 °C, by increasing the temperature of the sample, some extra nanocrystalline phase were appeared at 2θ = 32° and 54°. The regular calcination temperature for the bredigite sample was reported to be 1150 °C [8–10]. These additional peaks belong to CaO and carbon. Table 2 shows various parameters used to synthesize the bredigite powder. The pure bredigite powders were prepared in three steps. At the first step, while the milling time was 5 h and the sintering time was 10 h (at 1100 °C), the peak similarity of the XRD pattern was about 35%, in comparison with the initial pattern. At the second step, when the milling process increases to 8 h, the mentioned similarity reached