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Production of Highly Porous Fe-Si-Mg-C Alloy by Space Holder Method

Emre Atakan Meric [a], Muhammet Bayrak [a] and Ilven Mutlu [a*]

[a] Metallurgical and Materials Engineering Department, Istanbul University-Cerrahpasa, 34320, Istanbul, Turkey.

Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

In the present study, highly porous nonmagnetic bioresorbable Fe-5Si-8Mg-0.6C alloy was developed and produced for temporary biomedical implant applications. Iron alloy specimens with open porous structure were fabricated by powder metallurgy-based space holder method. Carbamide was used as a space holder. In general, Mg, Fe and Zn are biodegradable metals. Mg alloys biodegrade too fast with H2 evolution. Zn alloys show biodegradation rates in the middle of Mg and Fe alloys, but the Zn alloys are very brittle. Biodegradation rate of Fe alloys is too slow. Electrochemical corrosion behaviour of the foams was tested in simulated body fluid. Biodegradation rate was investigated by using weight loss and metal ion release measurements. Fe2+ ion release amounts of the specimens were lower than the upper limit for the humans.

Keywords: Corrosion; metal foam; biodegradation; fe alloy; powder metallurgy; temporary implant.

**Corresponding author: E-mail: imutlu@istanbul.edu.tr; imutlu@iuc.edu.tr;*

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1. INTRODUCTION

"Highly porous biodegradable scaffolds are used in the tissue engineering. Biodegradable scaffold provides mechanical support for the seeded cells and determines the shape of the tissues. Scaffold allows the transportation of the body fluids by the open pores. Biodegradable metals can be used in temporary implant applications. Permanent metallic implants show problems such as inflammation, stress shielding, secondary surgical operation" [1-3]. "Polymers have low strength, low wear resistance and release toxic reagents, while ceramics are brittle. Magnesium (Mg), iron (Fe) and zinc (Zn) are the biodegradable metals. Mg alloys biodegrade too fast with H_2 evolution. Zn alloys show biodegradation rates in the middle of Mg and Fe alloys, and their by-products are biodegradable. But, Zn is very brittle. Biodegradation rate of the iron is too slow. One method to fasten the biodegradation rate is addition of elements with lower standard electrode potential" [1-3]. "Open porous structure is important for the tissue ingrowth [4-8]. Open-cell foams can be manufactured by powder metallurgy. There are no solubility limitations in powder metallurgy as compared with casting" [9-19]. Francis et al. [15] studied "Fe alloys for stent applications. Fe alloys shows strength, high corrosion rate and biocompatibility. Fe alloy stents may biodegrade slowly while retaining their mechanical integrity". Orinakova et al. [16] produced "Fe alloy foams by replication method by using foamed polyurethane". Capek et al. [17] produced "Fe foams with 30-80 % porosity by powder metallurgy. Ammonium bicarbonate was used as a space holder". Lu et al. [19] produced biodegradable high nitrogen FeMnN alloy. It was found that, as the N content increased, the corrosion rate was enhanced. Elborolosy et al. [20] studied the biocompatibility, antibacterial effects of Fe-Mn-based alloys. Viability of epithelial cell line showed a significant increase. The highest percentage was in Fe-Mn-Co, followed by Fe-Mn-W then Fe-Mn-Cu.

In the present study, Fe alloy foams were fabricated by the space holder method. Although, there are studies on the Fe alloy foams, there is no study on the variation of the mechanical properties with time. In literature, there are several studies on the production of biodegradable Fe alloys, which are includes high Mn. The Mn addition produces austenite phase which eliminates ferromagnetic properties. Wang et al. [18] studied biodegradation properties of the Fe35Mn, and (Fe35Mn)5Ag alloys. Hermawan et al. [21] studied Fe-Mn alloys and concluded that the biodegradation rate lowered with increasing Mn content. In the present study, ferromagnetic properties were eliminated by lower alloying element addition. There is no study on the Fe-5Si-8Mg-0.6C alloy, which includes lower amounts of alloying elements.

2. METHODOLOGY

2.1 Alloy Production

"Fe-5Si-8Mg-0.6C alloy foams were fabricated by using Fe, Si, Mg, C powders. Metal powder mixtures were ball-milled with the zirconia balls for 15 hours with a rotational speed of 400 rpm. Metal powder to zirconia ball ratio was ten to one. Highly porous specimens were fabricated by space holder method. Carbamide powder in the range of 710-1000 μm was employed as a space

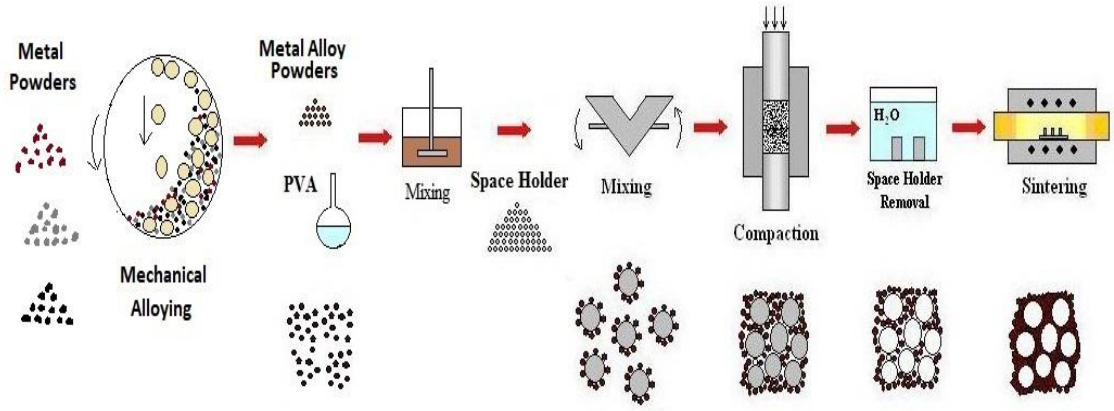

Fig. 1. Space holder method

holder material for its high solubility in water at room temperature. Carbamide is also nontoxic. Mixtures were pressed at about 190-200 MPa into the cylindrical shape. Green specimens were immersed in to water for 12 hours and then the carbamide was removed. Lastly, the samples were sintered for 1 hour at 1150 ºC" [18]. Fig. 1 illustrates the powder metallurgy based space holder method for the production of the open cell foams.

Schaeffler diagram can be used in order to predict the microstructural phases (austenite, ferrite, martensite) of the iron alloys, by using $Ni_{equivalent}$ and $Cr_{equivalent}$ values, which are given in equations (1) and (2). Implant materials must be magnetic resonance imaging compatible. Nonferromagnetic austenite phase must be obtained in the iron alloys. In the present study, nonferromagnetic austenite Fe-5Si-8Mg-0.6C alloy with the composition having $Ni_{equivalent}$ of 18% and $Cr_{equivalent}$ of 10% was produced [22].

$$Cr_{equivalent}=Cr+2Si+1.5Mo+5V+5.5Al+1.75Nb+1.5Ti+0.75W \quad (1)$$

$$Ni_{equivalent}= Ni+Co+0.5Mn+0.3Cu+25N+30C \quad (2)$$

2.2 Electrochemical Corrosion and Biodegradation Tests

"Simulated body fluid (SBF) solution was prepared from chemicals (Merck, Germany). The amounts of the chemicals (in g/L) were 8.0 NaCl, 0.3 $CaCl_2$, 0.2 KCl, 0.3 $MgCl_2$, 0.2 K_2HPO_4, 0.35 $NaHCO_3$, 0.07 Na_2SO_4, 6.0 tris and 1.0 M HCl. The pH of the SBF was 6.60. Electrochemical corrosion tests were done using a potentiostat (Interface, Gamry)" [23]. Graphite was counter electrode, saturated calomel electrode (SCE) was reference and the sample was working electrode. Initially, open-circuit potential was conducted. Tafel and linear polarization resistance tests were employed in order to determine the corrosion rates. Fe alloy specimens were dipped into the SBF for biodegradation tests. Solution volume to sample surface area ratio was constant. Weight loss values (%) were determined by gravimetric method.

2.3 Cytotoxicity Evaluation

"Evaluation of the cytotoxicity of the foams were studied by 3T3 neutral red uptake (NRU) assay, which is based on the ability of cells to uptake supravital neutral red dye. Living cells could be distinguished from dead cells. Positive (sodium lureth sulfate) and negative (polypropylene) samples were prepared to verify the system. Fe alloy test substances were exposed to immortalised mouse fibroblast Balb/c 3T3 cell line. Absorbance measurements were performed at 540 nm to determine the viability" [17]. Dulbecco's Modification of Eagle's Medium (DMEM) cell culture, bovine calf serum, trypsin/EDTA were the chemicals. The system is considered suitable if the viability for negative control specimen is >70 % of blank.

3. RESULTS AND DISCUSSION

3.1 Microstructure Characterization

Biodegradable highly porous Fe alloy specimens were fabricated for the temporary implant applications. Fig. 2 shows the optic microscope pictures of the Fe-6Mn-5Si-8Mg-0.5C alloy specimen. Microstructure consists of austenite main phase (bright, white areas) and some ferrite phase (brown areas). There are some micropores (dark areas), which are necessary for transportation of the body fluids.

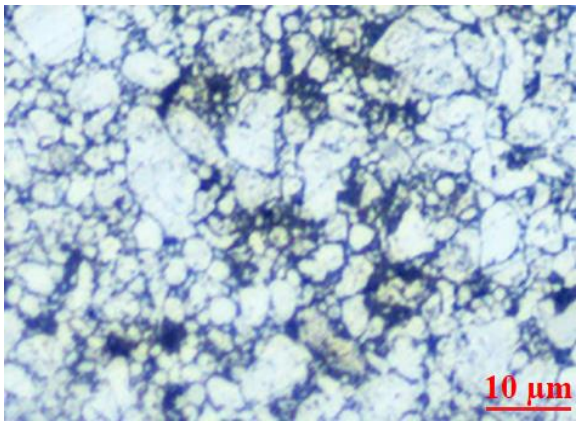

Fig. 2. Optic microscope picture of the specimen

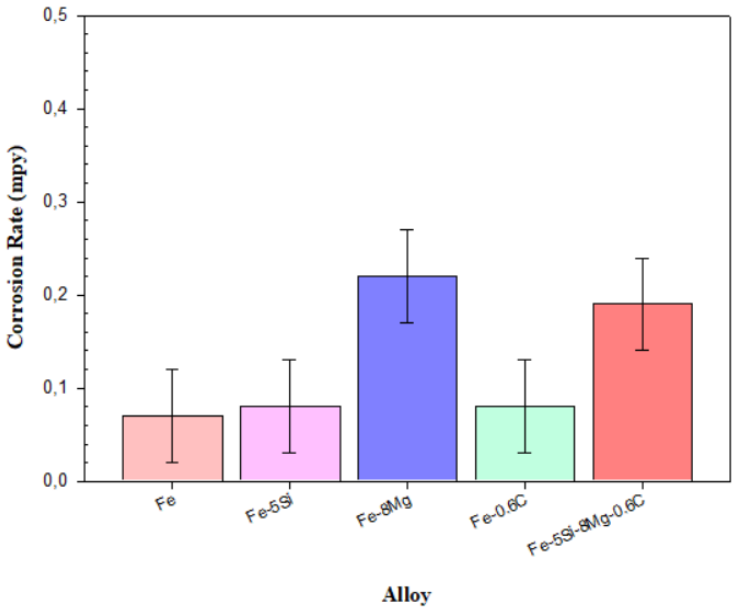

Fig. 3. Corrosion rates values of the Fe alloys

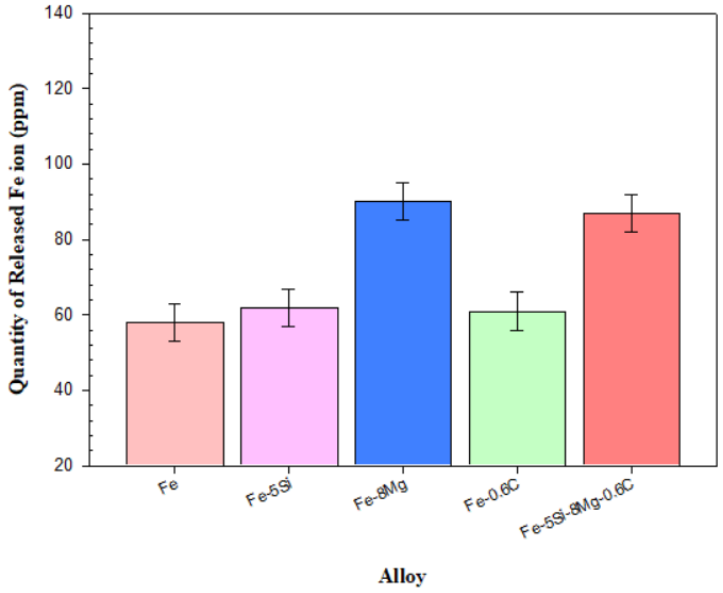

Fig. 4. Fe ion release values of the porous Fe alloys

Fig. 3 given below illustrates the electrochemical corrosion rates of the Fe alloys. Corrosion rate of the Fe-8Mg specimen was relatively high. Fe-5Si-8Mg-0.6C alloy also show high corrosion rate. Effect of Si and C addition was relatively neutral.

Fig. 3 shows the metal (Fe^{2+}) ion release values of the Fe alloys for 21 days of immersion in the SBF. Metal (Fe^{2+}) ion release and weight loss values were increased with time in the static immersion experiments. Metal (Fe^{2+}) ion release amount was lower than the daily limit for the iron (Fe^{2+}). Mg addition was increased the metal ion release.

Fig. 5. illustrates the weight change values of the highly porous Fe alloys for 21 days immersion in the SBF. Mg addition was increased the weight change values of the Fe alloy. Effect of C and Si addition was neutral.

Fig. 6 illustrates the variation of the Young's modulus values of the highly porous Fe alloys with immersion time in the SBF. Young's modulus values of the Fe alloys were deceased with immersion time. Increasing Mg content decreased the Young's modulus of the Fe alloy.

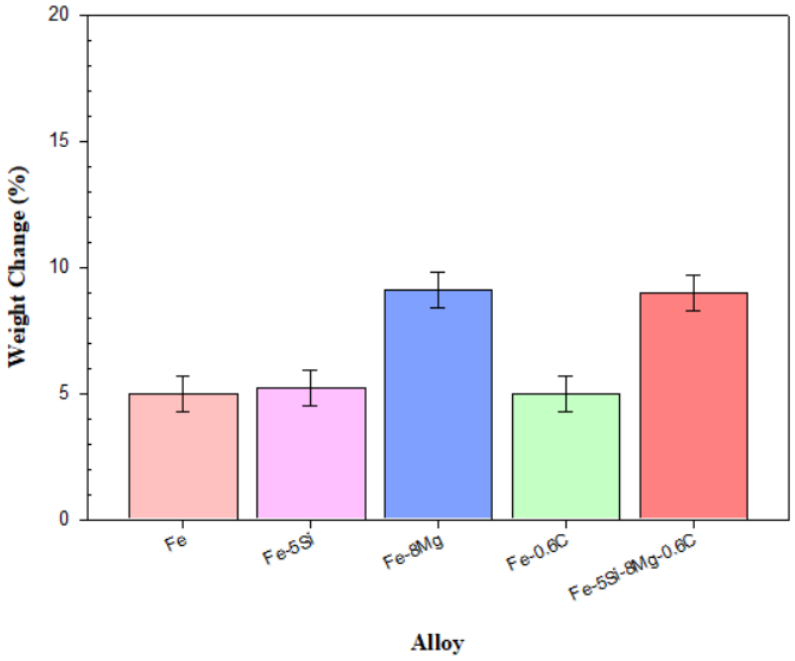

Fig. 5. Weight change values of the porous Fe alloys

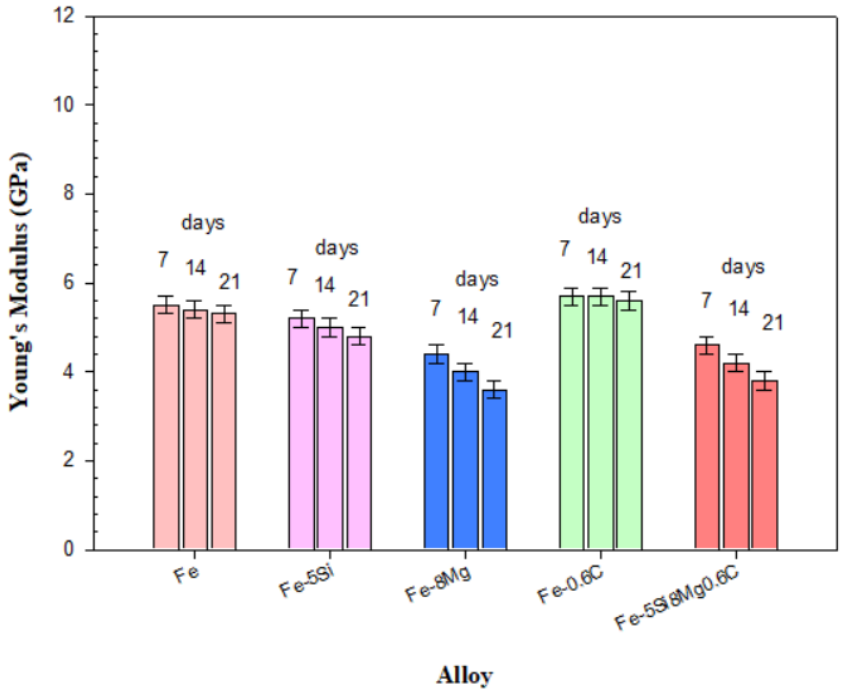

Fig. 6. Variation of the Young's modulus of the porous Fe alloys with immersion time

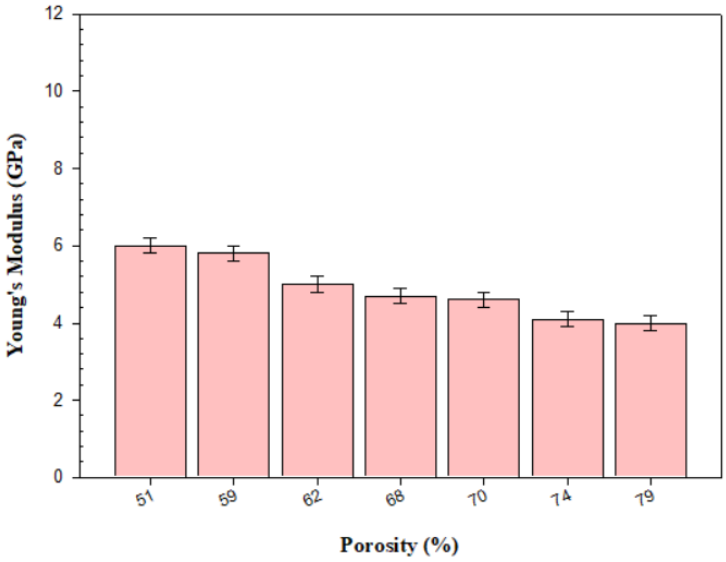

Fig. 7. Variation of the elastic modulus of the specimens with porosity

Fig. 7 shows the variation of elastic modulus of Fe-5Si-8Mg-0.5C alloy with porosity. Increasing porosity was decreased the elastic modulus. "Final pore size was related to the carbamide particle size. Particle size of carbamide was 800-850 µm, while pore diameter was 500-600 µm. Smaller pore sizes were attributed to crushing of the carbamide during compaction" [23].

In the present study, in vitro evaluation of the cytotoxicity of the Fe alloy foams were studied by 3T3 neutral red uptake assay. Viability of Fe-5Si-8Mg-0.5C alloy obtained was 77 % which is higher than the limit (70 %). This means that the Fe alloy has no cytotoxic potential. Cytotoxic values indicated the potential applications of the Fe-5Si-8Mg-0.5C alloy as implant material. Meanwhile, viability of the sintered pure Fe specimen was 81 %.

4. CONCLUSIONS

For use in temporary implant applications, biocompatible and biodegradable iron alloy foams were created in this work. Utilizing the space holder method, specimens with an open porous structure were created. The samples were subjected to electrochemical corrosion testing in SBF. We used the weight loss approach to evaluate the behavior of biodegradation. Fe^{2+} ion release amounts were negligible than the daily limit. Evaluation of the cytotoxicity was studied by neutral red uptake assay. Fe-5Si-8Mg-0.5C alloy does not have a cytotoxic potential. Elastic modulus of the specimens was deceased with immersion time. Elastic modulus values of the alloys were deceased with immersion time. Increasing content of alloying elements decreased the elastic modulus. Microstructure consists of austenite main phase and some ferrite phase.

DISCLAIMER (ARTIFICIAL INTELLIGENCE)

Author(s) hereby declare that NO generative AI technologies such as Large Language Models (ChatGPT, COPILOT, etc) and text-to-image generators have been used during writing or editing of manuscripts.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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