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Investigation of 58S bioactive glass tablets

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Abstract

In this study, bioactive glass powders were successfully synthesized by using the sol-gel process and bioactive glass powders were tabletted by direct dry pressing method. The morphology and surface properties of bioactive glass tablets were examined via field emission scanning electron microscope (FE-SEM) devices. X-ray diffraction (XRD) was utilized to evaluate the phases formed in the sol-gel bioactive glass tablets. Surface characterization of the tablets immersed in simulated body fluid (SBF) was carried out with XRD, FE-SEM, and Fourier transform infrared (FTIR). XRD, FTIR, and EDS analysis proved that the sample contained hydroxyapatite. Also, the in vitro mineralization assay demonstrated that bioactive glass tablets are capable of inducing the creation of hydroxyapatite after dipped in SBF. All analyze results showed that bioactive glass tablets have good apatite-forming activity.

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1. Introduction

Bioactive glasses are osteo-productive materials that give the ability to repair damaged bone. These properties are due to the developing dissolution in the physiological environment where the release of calcium, sodium, and phosphate ions begins to form and the apatite layer will form a vigorous bond with the surrounding bone tissues [1]. Bioactive glasses are one of the most promising bone regeneration materials because they can connect easily bone and assistance bone growth [2]. In addition, these glasses are capable of forming chemical bonds to the surface layer of the scaffold to promote new bone growth. In the 90s, 58S bioactive glasses were produced with a sol-gel method, which showed similar properties with 45S5 and showed higher dissolution rates for apatite formation [3].

The sol-gel process is a chemical-based synthesis road in which the solution having the precursor for the composition is subjected to polymer-type reactions at room temperature to form the gel [4]. Sol-gel is an important method that makes it probable to manufacture glasses containing compositions that cannot be achieved by conventional melting methods [5]. It also allows the chemical composition of bioactive glasses to be significantly expanded compared to the conventional melting process [6]. The sol-gel method has determined to be an ideal technique for preparing bioactive glasses. It is a process that allows the synthesis of glassy materials at low temperatures [7]. The sol-gels are made utilizing low-temperature hydrolysis and condensation process. Solution chemistry provides an easy mixing and good homogenization of chemicals. Low reaction temperatures avoid crystallization and phase separation, thus let the formation of glass that cannot normally be prepared [2]. The sol-gel process allows the formation of a silica network around the polymer chains via introducing the polymer into the sol. TEOS has been added to control the degree of covalent bonding between inorganic and organic components and as a silica precursor [8].

Since dry press molding does not require a complex experimental process, it can be commonly used in some specific fields. Dry press molding is a method of forming the tablet form by mixing powder and additive which does not destroy the structure of the bioactive particles. The tableting of bioactive powder particles is the only physical reaction that the bioactive properties of the material are maintained. This demonstrates that bioactive glass tablets have excellent bioactivity as bone healing materials and can be used in biomedical applications [9]. The aim of this study is to form high bioactivity glass particles by the sol-gel method and to

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tablet these bioactive glass particles and finally characterize this material. In the mineralization study, the hydroxyapatite formation process was systematically investigated. The phases, microstructures, and structural analysis of the tablets were identified by XRD, FE-SEM, FTIR before and after immersion in SBF.

2. EXPERIMENTAL STUDIES

2.1. 58S bioactive glass powders and tablets

Triethyl phosphate, Tetraethoxysilane Merck, Calcium nitrate tetrahydrate Acros organics, Hydrochloric acid was obtained from Isolab chemicals company. The bioactive glass powder content was determined in moles, 36% CaO, 60% SiO_2, 4% P_2O_5, and Triethyl phosphate ($Ca(N0_3)_2.4H_20$), Tetraethoxysilane, hydrochloric acid, and deionized water were used in this formulation. This study was carried out in four stages according to the method used by Chen et al [9]. First, 2.32 g triethyl phosphate, 20.48 g tetraethoxysilane, and 14.04 g $Ca(N0_3)_2.4H_20$ were added to a mixture of 2.60 g hydrochloric acid (2 M) and 15.64 g water, respectively. The mixture was stirred in a continuous magnetic stirrer until homogeneous. Then the zeta potential value of the sample was determined as -0.0245 mV. Second, the sol was aged at room temperature for 48 hours to fully react hydrolysis of the polycondensation. During this period of aging, the sol solidified as a transparent gel. Transparent solidification is indicative of monomerization.

In the third step, the gel was kept in the oven at 60 °C and 120 °C, respectively, for 24 hours to remove excess water and ethanol. The sample exposed to this temperature changes from transparent to white color and this event indicated that the polymerization is complete. Finally, the dried gel was applied to thermal processing in an oven at 650 °C for 3 hours to remove unreacted organic material. Thus, there was a burning reaction in the sample. After grinding for 3 hours at 650 °C, the sample was ground in a mortar and 0.5 grams for each tablet was weighed. To prepare tablets of good mechanical strength, it is necessary to reduce the grain size of the large bioactive glass samples to reduce the stress concentration effect of the bioactive glass tablets. The milled 58S bioactive glass particles were then made into tablet form by applying 22000 pounds (10 metric tons) pressure on a Carver hand press using. Sol and solid bioactive glass tablets that solidify into a transparent gel are shown in Fig. 1.

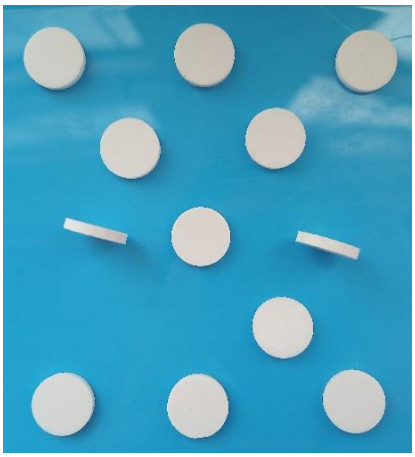


Figure 1. Bioactive glass tablets created by dry pressing method

2.2. In vitro bioactivity of bioactive glass tablets

The talent to form hydroxyapatite in vitro on the surface of the material was assessed by a simulated body fluid dipping test as described by Kokubo et al.[10]. The pH of the simulated body fluid solution was arranged to 7.40 with TRIS and HCl (1 M). The mineralization test with simulated body fluid was carried out at 37 °C for different periods of 7, 14, 21 and 30 days. Acetone was utilized to finish the mineralization process of tablets while removing from simulated body fluid. The tablets were then washed sequentially with ethanol and deionized water and finally left in a 60 °C oven for 1 hour to dry. Subsequently, the morphology of the newly formed hydroxyapatite on the tablet surface was investigated by field emission scanning electron microscopy. Mineral composition and crystal structure of hydroxyapatite were described with X-Ray Diffraction and FTIR spectroscopy analysis.

3. RESULTS AND DISCUSSION

3.1. Simultaneous thermal analysis (STA)

In order to determine the thermal processing temperature, simultaneous thermal analysis (STA) was done by examining the mass change of the organic-containing sample depending on the increasing temperature (Fig. 2). The mass reduction of the coating formed as a function of time or temperature was determined by Perkin Elmer STA 8000 brand device. Tg/DTA analysis was performed on the heat-processed sample at 650 °C for 3 hours. There were three stages of weight loss and a fragmentation reaction became in

Tg analysis. Because the analysis was carried out under a nitrogen atmosphere.

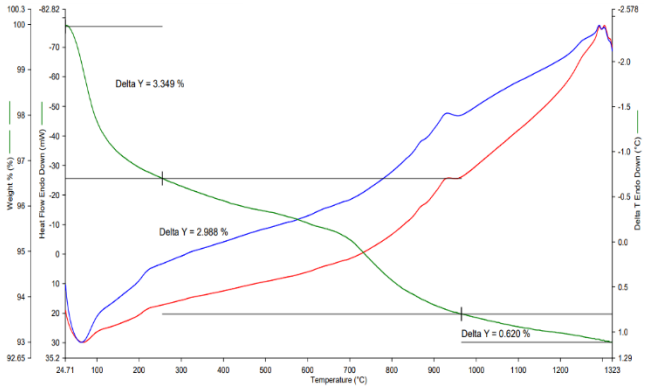


Figure 2. Tg / DTA Analysis

3.2. XRD analysis

The phases of the tablets obtained were characterized by the Panalytical Empyrean brand X-Ray diffraction (XRD) device. The XRD measurement was made in the range of 5-80 ° and the formed phases were calcite and hydroxyapatite before and after 7 days immersion in the SBF (Fig. 3). Fracture peaks observed at 2θ = 25.8°, 31.7 °, 39.8 °, 46.7, 48.6 °, 60.4 °, 65 °, 66.4 ° of hydroxyapatite crystals, It corresponded to the reflections of (002), (211), (310), (222), (320), (331), (511), (422) before immersion in the SBF. The prolongation of the hydroxyapatite peak length of the tablet immersed in simulated body fluid for 7 days is evidence of bioactivity. XRD results indicated that bioactive glass tablets have well bioactivity in vitro and mineralization products are hydroxyapatite.

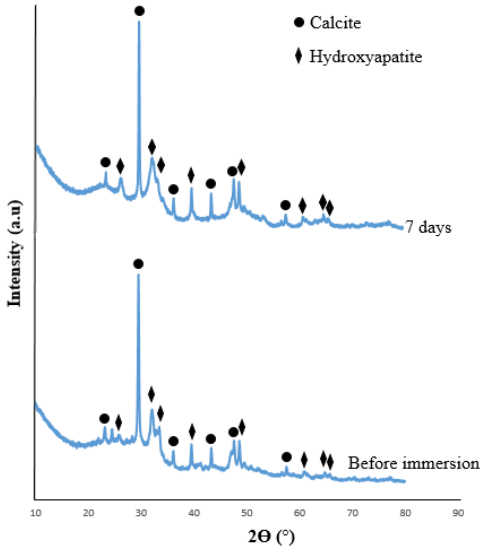


Figure 3. XRD analysis of the sample which was kept in SBF for 7 days and was not kept

3.3. FTIR analysis

Chemical characterization of the samples was performed using Perkin Elmer Spectrum Two brand Fourier Transform Infrared Spectroscopy in the range of 400-4000 cm^{-1}. Fig. 3 FTIR spectra of bioactive glass tablets before and after dipped in SBF may reflect the surface composition and structure of hydroxyapatite. Two peaks appearing at 603 cm-1 and 565 cm^{-1} before being dipped in SBF P-O is connected to bending vibration. The reason for the occurrence of a two-part peak is that the molecules in the crystal lattice are regulated in a regular manner due to the reinforced intermolecular interaction and eventually lead to band division. After being dipped in SBF for 7 days, these peaks length in the split phosphate band grew. When the mineralization time was increased to 7 days, the hydroxyapatite crystallization process was accelerated and the intensity of the peaks increased as the mineralization time increased. At the same time, these peaks approaching each other were seen in 571 cm^{-1} and 569 cm^{-1}. Furthermore, the peak corresponding to the P-O tensile vibration before submerged in the SBF appeared at 941 cm^{-1} and is clearly visible. Peaks at 875 cm^{-1} and 1644 cm^{-1} correspond to C-O tensile vibration. This indicates that the mineralization goods contain a carbon element. As a result, the mineralization product of bioactive glass tablets was hydroxyapatite after immersion in the SBF solution.

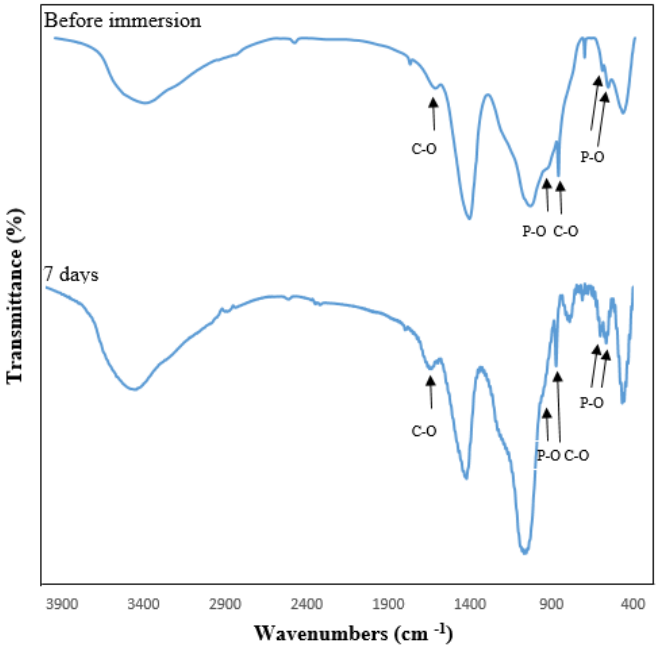


Figure 4. FTIR analysis of the sample which was kept in simulated body fluid for 7 days and not kept

3.4. FE-SEM analysis

The microstructure of the 58S bioactive glass tablet was examined using a Hitachi Regulus 8230 FE-SEM device. The FE-SEM Fig. 5 proved that the bioactive glass tablet has heterogeneous and irregular morphology. Vapor-liquid interfaces between bioactive glass powders occur with the drying process. Due to the impact of the interfacial stress, the curvature of the liquid surface has emerged between the particles. This was created a strong tensile force that caused the gel skeleton to precipitate. In the end, the particles were in close contact. This caused the soft and tough particles to come together. Therefore, the formed bioactive glass particles required to be pulverized before dry pressing molding.

FE-SEM analysis revealed a large number of inhomogeneous nanoparticles on the bioactive glass tablets (Fig. 5). Initially, cracks were not observed in the microstructure. Nuclei formed on the surface of the sample dipped in SBF for 30 days grew. The surface was completely covered with formed hydroxyapatite clumps. However, large bioactive glass particles began to disintegrate in the sample immersed in SBF for 30 days. Because the particles forming the tablets are not homogeneous, many micro-cracks have appeared on the surface of bioactive glass tablets (Fig. 7). After being dipped in SBF the tablets produced stress concentration and swollen. Cracks have been formed between the bioactive glass tablets and the SBF solution under the influence of water abrasion and ion exchange, as a result of physical and chemical reactions. While the Ca/P ratio was 10.30 before immersion in SBF (Fig. 5), this ratio was determined as 18.18 and 7.42 in the sample dipped for 7 days (Fig. 6). Calcite was formed from the excess of calcium on the surface of the tablets. The Ca/P ratio of the sample immersed for 30 days decreased to 3.58, 2.88, and 3.07 (Fig. 7).

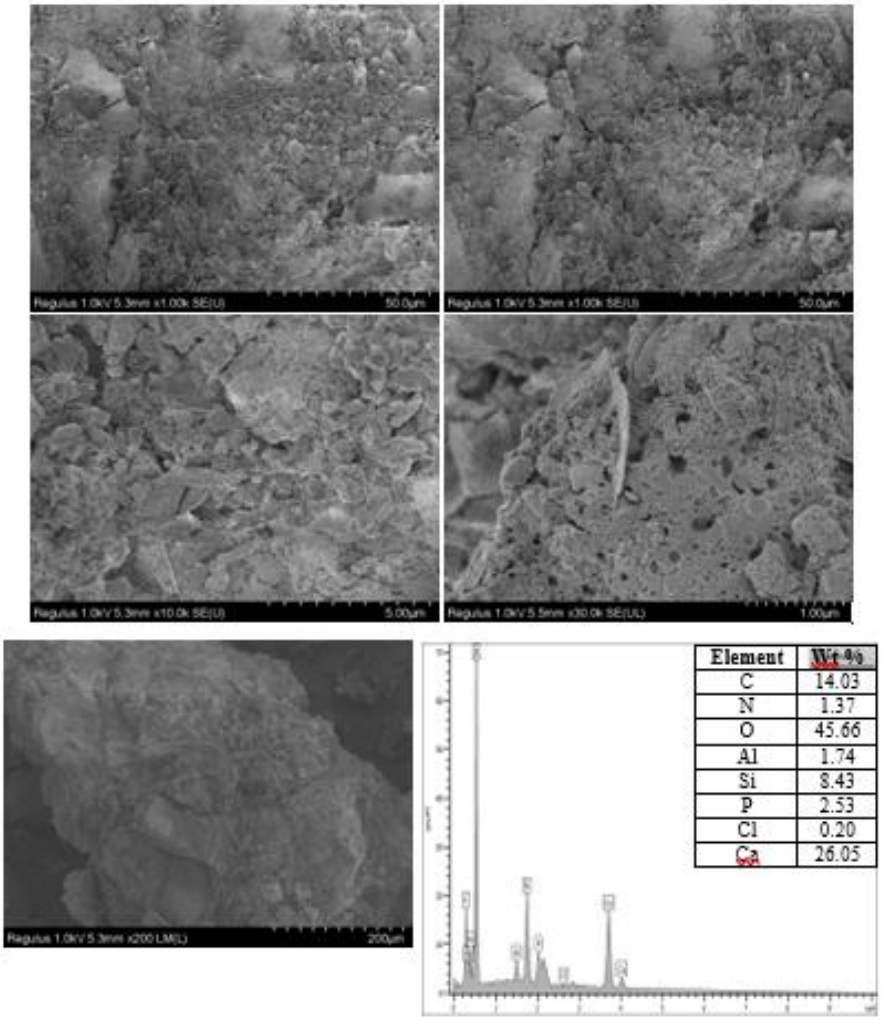
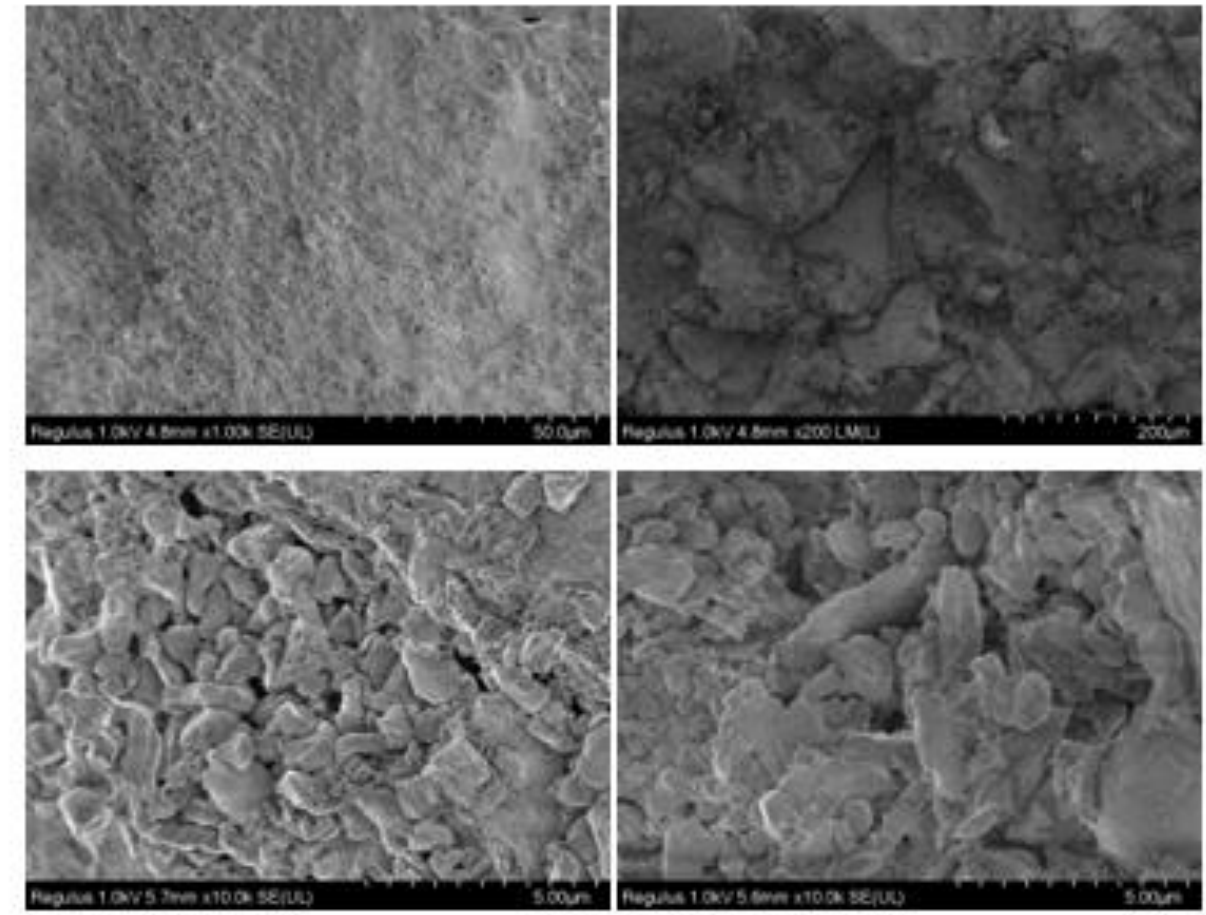


Figure 5. FE-SEM and EDS analysis of a heat-treated sample at 650 °C for 3 hours



Element	Wt %
C	12.06
N	0.53
O	38.19
Al	0.25
Si	7.08
P	2.18
Cl	0.07
Ca	39.65

Element	Wt %
C	7.85
N	0.71
O	27.76
Al	0.53
Si	31.62
P	3.71
Cl	0.28
Ca	27.54

Figure 6. FE-SEM and EDS analysis of the sample held in simulated body fluid for 7 days

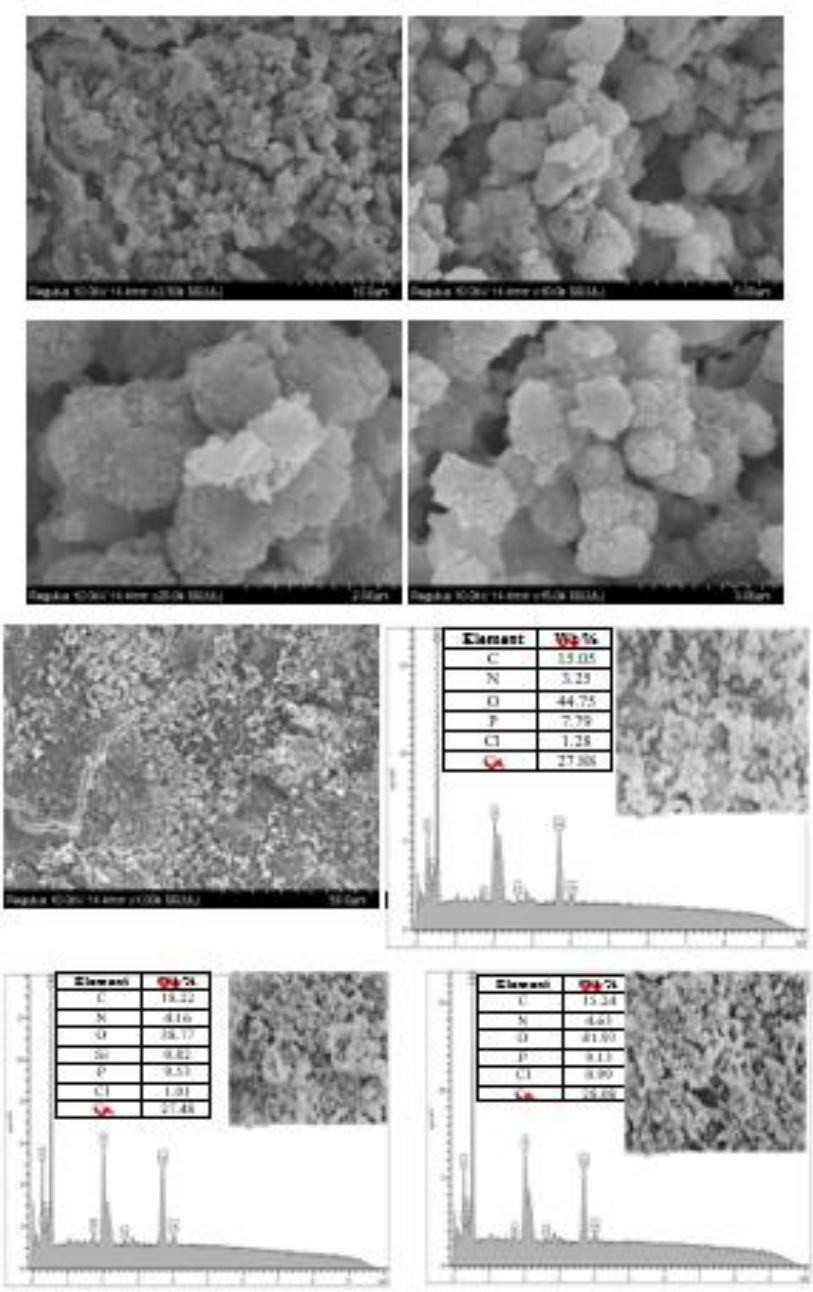


Figure 7. FE-SEM and EDS analysis of the sample held in simulated body fluid for 30 days

4. Conclusions

In this study, bioactive glass powders were successfully created via the sol-gel method. Powders were brought to the tablet form. It can be seen that the bioactive glass tablets made by dry pressing technology are pill-like and homogeneously structured, and the surface was slightly rough. In this study, 58S bioactive glass tablets were prepared and characterization studies were performed. Bioactivity of the formed tablets was appraised in vitro. The XRD, FTIR, and EDS analysis demonstrated that a dense layer of hydroxyapatite was formed on the surface of bioactive glass tablets after immersion in SBF and that the tablets had perfect bioactivity. The results show that the tablets created have the potential to use as a biological scaffold in tissue engineering. As a

consequence of the FTIR analysis, the formation of P-O bonds in the structures of the samples was determined. The peaks became sharp with an increase in the waiting time in the SBF. In other words, it has been determined that the formation of hydroxyapatite increases.

Acknowledgements

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Conflicts of Interest

The authors declare no conflict of interest.

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