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Preparation of Hydroxyapatite and Bioactive Glass Ceramic to Get Biocomposite by Using a Genetic Algorithm Method

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Abstract: In this study, hydroxyapatite was prepared synthetically by the sol-gel approach. The precursors used were calcium nitrate tetrahydrate and di-ammonium hydrogen phosphate. The bioactive glass ceramic was obtained by melt derived process. Particle size analyzer was used to determine the size of the prepared powders and XRD have been used to characterize the prepared powder phases and the crystallites size. Biocomposite was prepared via. mixing different proportions of bioactive glass ceramic 0, 5, 15, 25 wt.% with hydroxyapatite by using 5% bioactive glass ceramic, hydroxyapatite was phase transformed to β-TCP. Thus, bioactive glass ceramic works only as a sintering aid while at using 15 and 25% bioactive glass ceramic, it reacts with HA to give sodium calcium phosphate $Na_3Ca_6(PO_4)_5$ which is a new bioceramic on which research is still going on 25% the bioactive glass was used with HA obtained from bovine bone and it gave the same phase (sodium calcium phosphate $Na_3Ca_6(PO_4)_5$).

Key words: Hydroxyapatite, bioactive glass ceramic, sol-gel approach, melt derived process, genetic algorithm method, tetrahydrat

INTRODUCTION

Biomaterials such as Hydroxyapatite (HA) and bioactive glass ceramic such that based on Na_2O-CaO-P_2O_5-SiO_2 have been increasingly used as a bone substitute due to their biological properties like biocompatibility, bone bonding ability and similarity to the mineralized phase of the bone. Bioceramics are materials that often used for the medical and dental applications. They can be classified according to their bioactivity into surface active, inert and resorbable. The bioactive material is an intermediate between biodegradable and bioinert material. These materials form a strong bond with the host tissue through forming a Carbonated Hydroxyapatite layer (CHA) (Xian, 2010; Ogawa *et al*., 1988).

Hydroxyapatite is a calcium phosphate ceramic which has the formula $Ca_{10}(PO_4)_6(OH)_2$, it has a composition with Ca/P ratio of 1.67 (Bilton *et al*., 2010). HA has widely used bioceramic due to biocompatibility, excellent bioactivity and easily bonding to the bone and surrounding tissues (Hoque *et al*., 2014). Some of the classical methods for HA synthesis include hydrothermal technique, direct precipitation, solid state reaction, hydrolysis of calcium phosphates and wet precipitation (Agrawal *et al*., 2011). Sol-gel process is a wet chemical method that doesnt need very high sintering temperature and offer a molecular mixing of the phosphorus and calcium that improves the chemical homogeneity. Also, the sol-gel powder reactivity reduce the processing temperature (Feng *et al*., 2005).

The first material prepared to bond to bone was bioglass®45s5 by Larry Hench consisting of (46.1% SiO_2, 24.4% Na_2O, 26.9% CaO and 2.6% P_2O_5 in mol.%) (Wu *et al*., 2005). It has attraction due to good biocompatibility and bond strongly to bone due to forming a layer of carbonated hydroxyapatite (Yu *et al*., 2013). Bioactive glass-ceramics are obtained by crystallizing bioactive glass at high temperatures (Kokubo *et al*., 2008).

The Genetic Algorithm (GA) developed by Goldberg was inspired by Darwins theory of evolution which states that the survival of an organism is affected by rule the strongest species that survives (Vishwakarma, 2012). The GA is one of the most succeeded optimization techniques that were used to solve combinatorial optimization problems. However, the GA differs from the traditional search and optimization methods by the following: it search a population of points are not a single point does not need derivative information or auxiliary knowledge, the direction of the search is affected only by objective function and fitness levels and it uses probabilistic

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transition rules and it works on encoding of the parameter set (Al-Dujaili *et al*., 2017b). GA is not commonly used in ceramic field. The GA approach starts with a set of individuals that are randomly generated in a named population. Each individual was called a chromosome which was evaluated through using a certain function named the fitness function. After that a new population is generated by using the GA operator (selection, crossover and mutation) that replaces the old population. Repeating this process would gain the optimum solution (Al-Dujaili *et al*., 2017a).

The study is adding different percentage may control the particle size and particle size distribution. In the case of multimodal distribution when is increasing the concentration the modal of the coarser particles becomes the major modal (Al-Jabar *et al*., 2017). For that reason, this research aims to study the effect of adding different amounts 0, 5, 15 and 25 wt.% of bioactive glass ceramic on the phases of hydroxyapatite.

MATERIALS AND METHODS

The study was prepared the materials as shown in Table 1.

Hydroxyapatite preparation: HA was prepared by sol-gel method using calcium nitrate tetrahydrate $Ca(NO_3)_{2.4}H_2O$ and Di-Ammonium hydrogen Phosphate (DAP) with the formula $(NH_4)_2\ HPO_4$ as precursors, 500 mL of distilled water have been used to dissolve 1 M of $Ca(NO_3)_{2.4}H_2O$ and 0.67 M of $(NH_4)_2HPO_4$ in a separate beakers. The first precursor was added rapidly to the DAP under stirring at 75°C, the pH of this solution is (~6). The final product was stirred for 45 min then NH_4OH was added drop wised to the solution under stirring until the PH is in the range of 9-11 and stirred for 5 h. The obtained gel was aged for 24, then washed with distilled water and ethanol. Finally, the gel was filtered and dried at 80°C for 6 h:

$$6\left(NH_4\right)_2\ HPO_4+10Ca\left(No_3\right)_2.4H_2O+$$
$$8NH_4OH \rightarrow Ca_{10}\left(PO_4\right)_6\left(OH\right)_2+20NH_4NO_3+6H_2O \quad (1)$$

Bioactive glass ceramic preparation: Calcium oxide was obtained from an oyster shell by grinding.

Table 1: Characteristics of raw materials

Materials	Formulas
Calcium nitrate tetrahydrate	$Ca(NO_3)_{2.4}H_2O$
Di-ammonium hydrogen phosphate	$(NH_4)2HPO_4$
Ammonium hydroxide solution	$NH_4(OH)$
Silica foam	SiO_2
Oyster	$CaCO_3$
Sodium carbonate	Na_2CO_3
Phosphorous pentoxide	P_2O_5

The shells by ceramic mortar and then calcined at 900°C for 2 h to obtain an off-white quicklime according to:

$$CaCo_3(s) \rightarrow CaO(s)+Co_2(g) \quad (2)$$

45S5 bioglass® was synthesized by mixing 45 wt.% SiO_2, 24.5 wt.% CaO, 24.5 wt.% Na_2O and 6 wt.% P_2O in a planetary ball mill for 4 h. Then, the powder was melted at 1200°C for 4 h with a heating rate of 15°C/min in an alumina crucible and allowed to cool down. Then, it was heat treated at 800°C for 5h with 5°C/min heating rate to complete the full crystallization. The obtained bioactive glass ceramic was ground by the planetary ball mill for 4-5h (Fig. 1).

HA/bioactive glass-ceramic preparation: Different percentages from the bioactive glass ceramic 0, 5, 15, 25% by weight were added to hydroxyapatite and mixed thoroughly. Then, the obtained mixture was pressed uniaxially in a die with a diameter of 10 mm to a pressure of 114 Mpa according to ASTM-D695. And sintered at 1200°C for 4 h with heating rate of 15°C/min. The tests results were achieved in the labs of Department of Ceramics and Building Materials, College of Materials Engineering, University of Babylon (Fig. 2).

Materials characterization: The particle size distribution was determined via. using Bette rsize 2000 laser particle size analyzer (Bettersize Instrument Ltd., China in Department of Ceramics and Building Materials, College of Materials Engineering, University of Babylon). The phases were evaluated via. using X-ray diffractometer (XRD 6000, Shimatzo, Japan in Department of Ceramics and Building Materials, College of Materials Engineering, University of Babylon) at room temperature by using Cuk α radiation (λ = 1.5405 Å) with scanning speed of 5°/min and applied power of 40 kV/30 mA (Fig. 3 and 4).

Fig. 1: High speed shimmy ball mill SFM-1 (QM-3SP2)

Fig. 2: Sintering (heat treatment) furnace

Fig. 3: Laser particle size analyzer

Fig. 4: X-ray diffractometer

RESULTS AND DISCUSSION

Prepared particle size analyses results: Figure 5a and b was shown the result of the particles size distribution

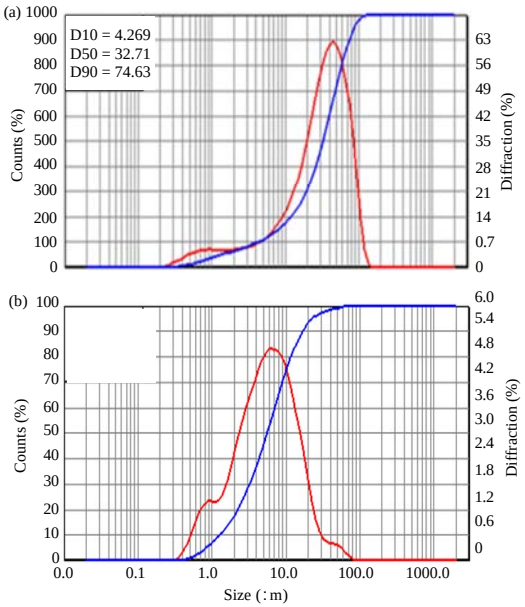

Fig. 5: Particle size distribution of: a) Bioactive glass ceramic and b) Hydroxyapatite

analysis for the hydroxyapatite and bioactive glass ceramic, respectively. The particle size distribution of bioactive glass ceramic was over the range 0.829-94.91 μm and the D10, D50, D90 are 4.269, 32.71, 74.63. For hydroxyapatite, the particle size distribution was over the range 0.686-28.68 μm and the D10, D50, D90 are 1.272, 5.538, 16.64.

X-ray diffraction results: The phases of prepared powders were obtained via. using XRD. Figure 6a represents the phases of hydroxyapatite obtained by sol-gel, the broad peak is an indication of being nanoparticle and it agrees with JCPDS, card No. 09-0432. The peaks illustrated in Fig. 6b represents the sodium calcium silicate crystalline phase of the formula $Na_2Ca_2Si_3O_9$ which agrees with JCPDS, card No. 22-1455 and $Na_2Ca_2Si_3O_8$.

Figure 6c shows the beta-tricalcium phosphate ($Ca_3(PO_4)_2$) that agrees with JCPDS, card No. 09-0169 obtained via. adding 5% bioactive glass ceramic due to the decomposition of the hydroxyapatite by sintering. Figure 6d, e was shown the phases of the biocomposite formed through adding 15 and 25%, respectively, sodium calcium phosphate ($Na_3Ca_6(PO_4)_5$) and it agrees with the JCPDS, card No.11-0236.

This is because the reaction between bioactive glass ceramic and beta tricalcium phosphate which forms due to the decomposition of HA. Figure 6f shown the XRD for the biocomposite prepared from adding 25% bioactive

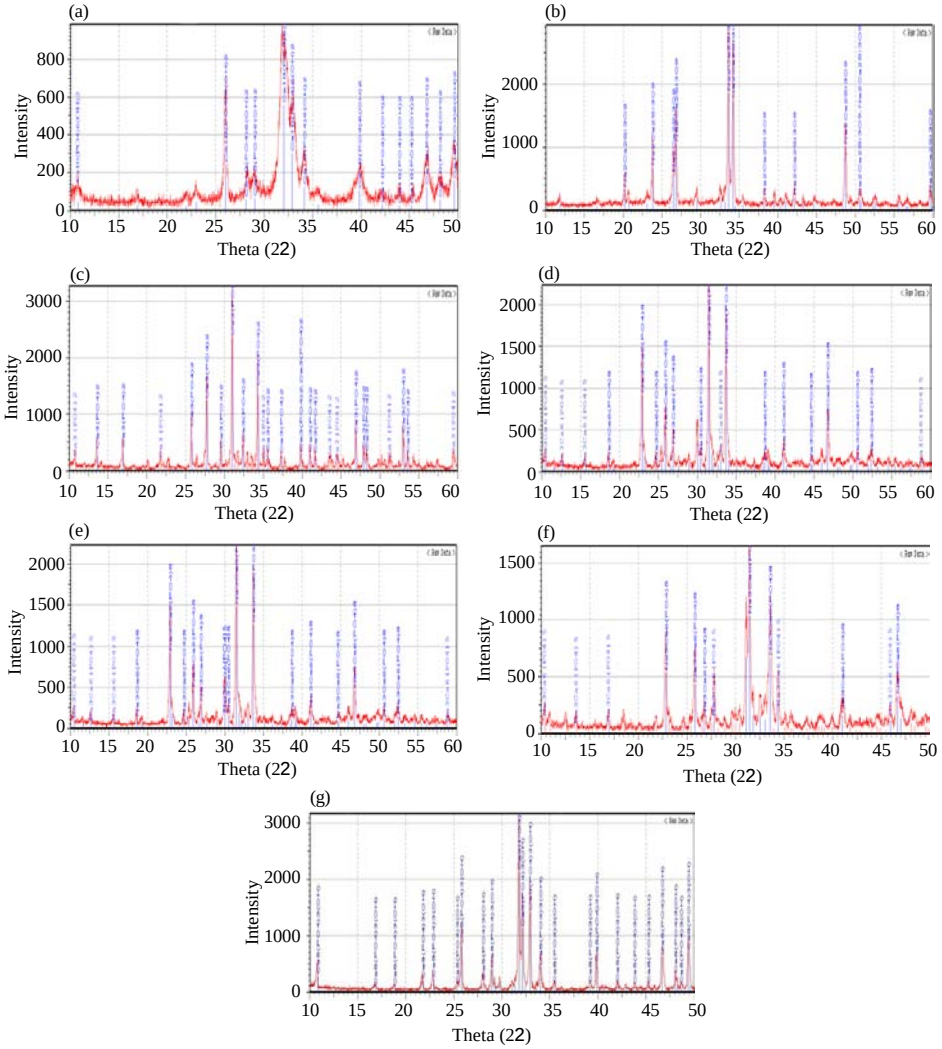

Fig. 6: XRD patterns of: a) Hydroxyapatite; b) Bioactive glass ceramic; c) 5% bioactive glass ceramic and HA; d) 15% bioactive glass ceramic with HA; e) 25% bioactive glass ceramic with HA; f) Bioactive glass ceramic with HA obtained from bovine bone and g) HA obtained from bovine bone at 1300°C

glass ceramic to bovine bone HA. This also produces ($Na_3Ca_6(PO_4)_5$) due to the reaction between HA and bioactive glass ceramic, though; HA that was prepared naturally from bovine bone starts to decompose at a temperature of about 1300°C as shown in Fig. 6g.

Regression function or fitness function: A porosity prediction model was established by using Minitab Software by taking the porosity as a dependent variable and ratio of the bioactive glass ceramic as an independent variable (the input):

$$P = 29.424-0.1932R \quad (3)$$

Where:
p = The porosity
R = The ratio of the bioactive glass ceramic

The R^2 is 3.45%, R-adj is 2.22%. The prediction model for density is made by taking density as dependent ariable. While the ratio of the bioactive glass ceramic as an independent variable (input) as follows:

$$\rho b = 2.6161+0.01152R \quad (4)$$

The R^2 is 5.59%, R-adj is 4.38%.

Analysis the regression equation: The optimization of the porosity and density were obtained through using genetic algorithm with support by experimental work Table 2. This is because the study looking for finding values of the processing parameters for the bioactive glass ceramic concentration (Table 3).

Table 2: Regression equation analysis of porosity

Terms	Coefficient	SE coefficient	t-values	p-values	Vif
Constant	12.34	1.3600	9.08	0.000	-
Concentration	0.1535	0.0909	1.67	0.099	1.00

Table 3: Regression equation analysis of density

Terms	Coefficient	SE coefficient	t-values	p-values	Vif
Constant	2.6161	0.07930	32.98	0.000	-
Concentration	-0.01152	0.00536	-2.15	0.035	1.00

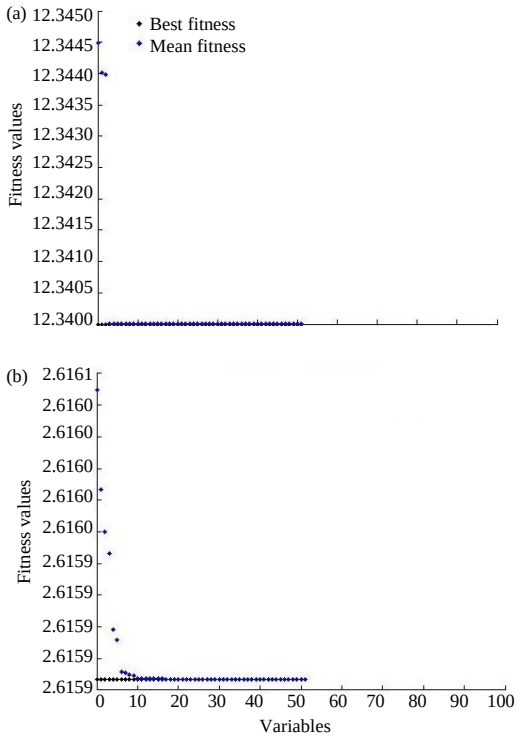

Fig. 7: The regression versus the fitness value for: a) Porosity (Best: 12034, Mean: 1234) and b) Density (Best: 2.61587, Mean: 2.61587)

The genetic algorithm approach: The GA option from the optimization method via. MATLAB Software was used to present the solution of the optimization problem. This software is available in the computer cluster in Department of Ceramics Engineering and Building Materials, College of Materials Engineering, University of Babylon. The best individual values and generation versus fitness value were shown in Fig. 7a and b.

CONCLUSION

HA/bioactive glass ceramic was successfully prepared via. using the solid state reaction. When the study was added 5% bioactive glass ceramic. The phase produced was beta-tricalcium phosphate ($Ca_3(PO_4)_2$) which means that bioactive glass ceramic act only as a sintering aid and helps the decomposition of HA. While when the study was added 15 and 25% bioactive glass ceramic. The sodium calcium phosphate was obtained ($Na_3Ca_6(PO_4)_5$) due to the reaction between beta tricalcium phosphate and bioactive glass ceramic.

The same phase was obtained when was using 25% bioactive glass ceramic and HA synthesized from bovine bone. This means that the bioactive glass ceramic act to decompose the HA to beta tricalcium phosphate and then react with it. Although, at the same sintering temperature (1200°C), HA doesnt decompose. The best result of porosity is 12.34 compared with the range of real values 1.35-27.25 and 2.61587 for the density compared with the range 1.8738-3.2843. Immerse the samples in the Simulated Body Fluid (SBF) and study its effect upon the biological properties. Study of the morphology of the sample by Scanning Electron Microscope (SEM) before and after soaking in (SBF).

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