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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-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. 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 ß-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 $Na_3Ca_6(PO_4)_5$).

Keywords: Hydroxyapatite, Bioactive glass ceramic, Sol-gel approach, Melt derived process, Genetic algorithm method

I INTRODUCTION

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) [1, 2].

Hydroxyapatite is the most widely used calcium phosphate material with a Ca/P ratio of 1.67, it has a hexagonal unit cell having the formula of $Ca_{10}(PO_4)_6(OH)_2$. It is used in dental and implantation applications due to biocompatibility and the similarity of its bioactive behavior to the mineral component of the bone and the teeth [3].Various synthesis technique have been developed like direct precipitation from aqueous solution, sol-gel, hydrothermal synthesis, and emulsion and microemulsion routes. Sol-gel process is a wet chemical method that doesn't 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 [4].

The first bioactive glass material developed is named bioglass has developed by Larry Hench which has shown high bioactivity and can join easily even to soft tissues, it consists of 45 wt% SiO_2, 24.5 wt% CaO, 24.5 wt% Na_2O and 6 wt% P_2O_5. Bioactive glass-ceramics are obtained by crystallizing bioactive glass at high temperatures [5, 6].

The Genetic algorithm (GA) is one of the most succeeded 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 transition rules and it works on encoding of the parameter set [7].

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. [8]. For that reason, this work aims to

study the effect of adding different amounts (0, 5, 15 and 25 wt. %) of bioactive glass ceramic on the phases of hydroxyapatite.

II EXPERIMENTAL

A. HYDROXYAPATITE PREPARATION

HA was prepared by sol-gel method, using calcium nitrate tetrahydrate $Ca(NO_3)_2.4H_2O$ and di-ammonium hydrogen phosphate (DAP) with the formula $(NH_4)_2HPO_4$ as precursors, 500 mL of distilled water have been used to dissolve 1M of $Ca(NO_3)_2.4H_2O$ and 0.67M 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 45min 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 5h.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 6h.

$$6(NH_4)_2HPO_4 + 10\ Ca\ (NO_3)_2.4H_2O + 8NH_4OH \rightarrow Ca_{10}(PO_4)_6(OH)_2 + 20NH_4NO_3 + 6H_2O \quad (1)$$

B. BIOACTIVE GLASS CERAMIC PREPARATION

Calcium oxide was obtained from an oyster shell by grinding the shells by ceramic mortar and then calcined at 900˚c for 2h to obtain an off-white quicklime according to:

$$\mathbf{CaCO_3(s) \rightarrow CaO(s) + CO_2\ (g)} \quad (2)$$

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 4h. Then, the powder was melted at 1200˚c for 4h 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.

C. 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. For three points bending the die used having the dimensions (60×6×4 mm) and sintered at 1200˚c for 4h with heating rate of 15˚c/min.

D. MATERIALS CHARACTERIZATION

The particle size distribution was determined via using Bette rsize2000 laser particle size analyzer (bettersize instrument Ltd., China, department of Ceramics and Building Materials, University of Babylon). The phases were evaluated using x-ray diffractometer (XRD 6000, shimatzo, Japan, department of Ceramics and Building Materials, University of Babylon) at room temperature using Cukα radiation (λ = 1.5405 Å), with scanning speed of 5º/min and applied power of 40 kv/30 mA.

III RESULT AND DISCUSSION

A. PARTICLE SIZE ANALYSES RESULTS

Fig.1 (a and b) was shown the result of analysis of particles size distribution for the hydroxyapatite and bioactive glass ceramic respectively. The particle size distribution of bioactive glass ceramic is 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 over the range (0.686-28.68 µm) and the (D10, D50, D90) are (1.272, 5.538, 16.64).

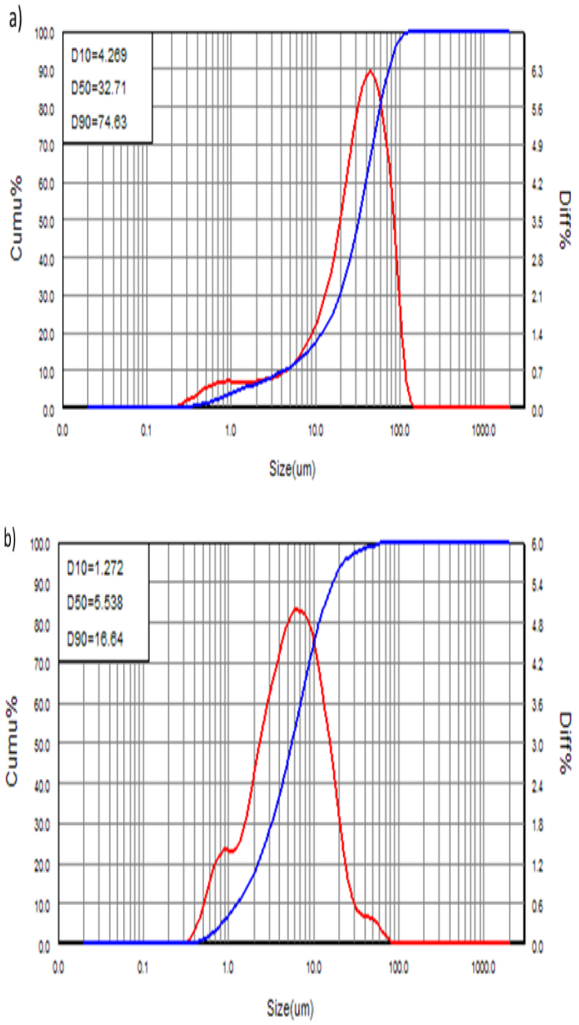
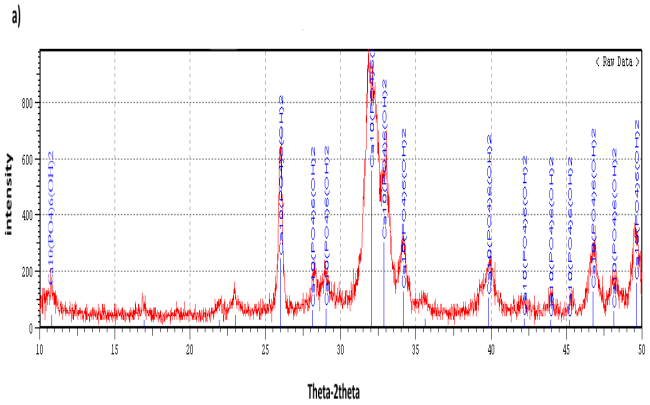


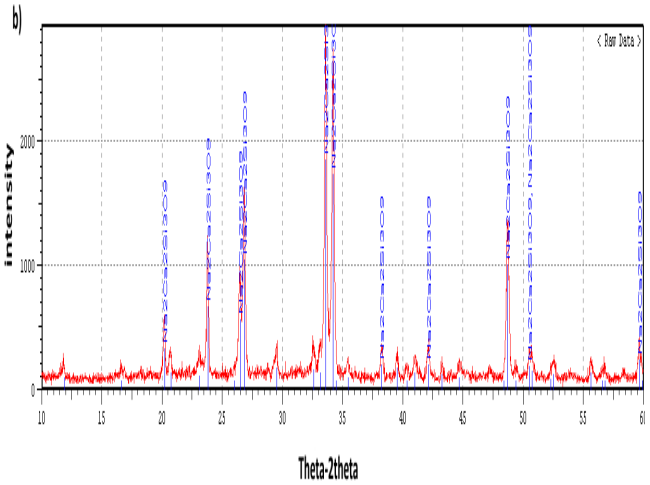
Fig1. particle size distribution of a) bioactive glass ceramic. b) Hydroxyapatite.

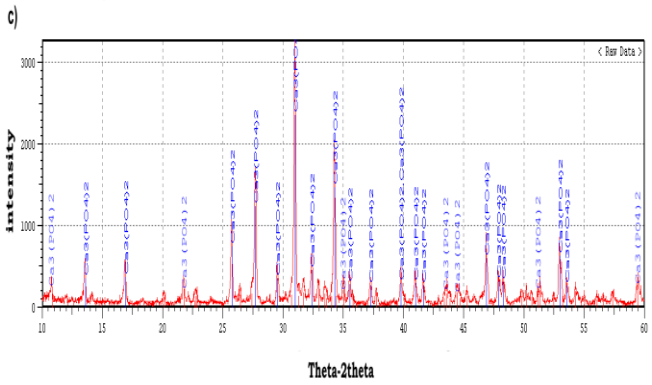
B. X-RAY DIFFRACTION RESULTS:

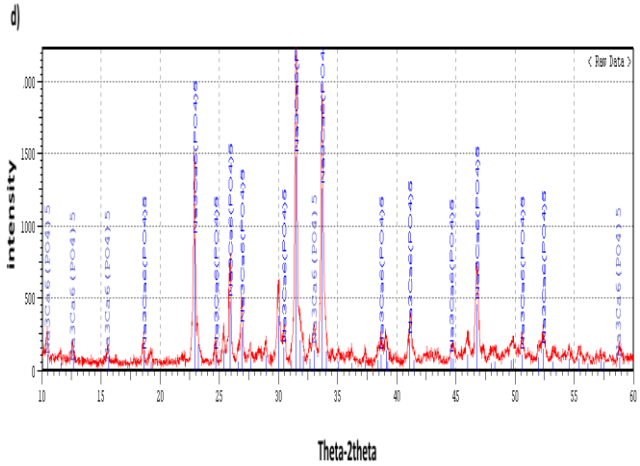
The phases of prepared powders were obtained by using XRD. Fig. 2a represents the phases of hydroxyapatite and it agrees with (JCPDS, card NO. 09-0432). The

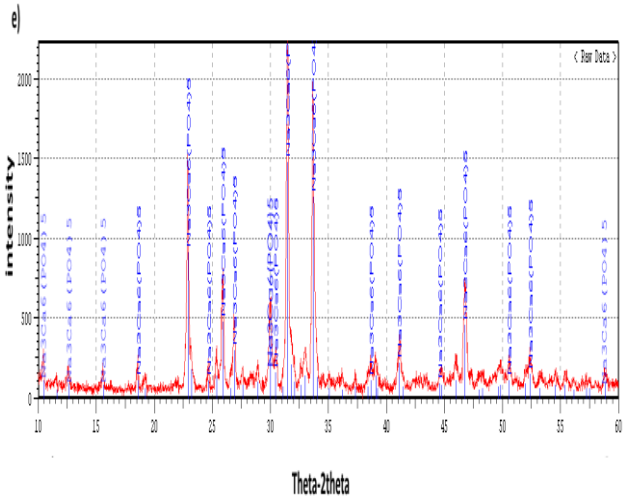
peaks illustrated in Fig 2b represents the sodium calcium silicate crystalline phase of the formula ($Na_2Ca_2Si_3O_9$) which agrees with (JCPDS, card NO. 22-1455) and ($Na_2Ca_2Si_3O_8$). Fig3c shown the beta-tricalcium phosphate ($Ca_3(PO_4)_2$) that agrees with (JCPDS, card NO.09-0169). Fig3 d, e) was shown the phases of the biocomposite formed by adding and 25% respectively, sodium calcium phosphate ($Na_3Ca_6(PO_4)_5$) and it agrees with the (JCPDS, card NO.11-0236). Fig3f shown the XRD for the biocomposite prepared from adding 25%bioactive glass ceramic to HA obtain from bovine bone.











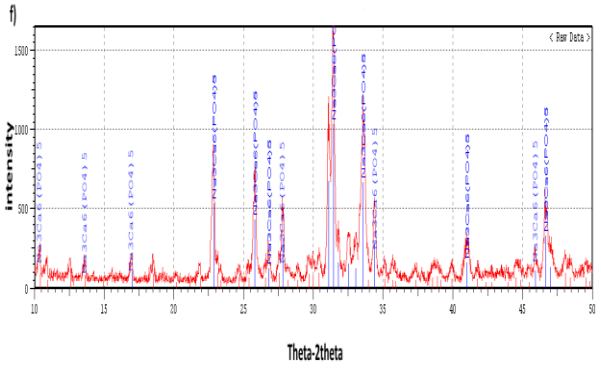


Fig.2: XRD patterns of a) hydroxyapatite, b)bioactive glass ceramic, c)5%bioactive glass ceramic and HA, d)15% bioactive glass ceramic with HA, e)25% bioactive glass ceramic with HA, and f) bioactive glass ceramic with HA obtained from bovine bone.

C. REGRESSION EQUATION ANALYSIS

A compression strength prediction model was established by using Minitab software is taking the compression strength as a depending variable and ratio of the bioactive glass ceramic as an independent variable (the input).

$\boldsymbol{\sigma} = 6.0508 + 0.06881R.$

Where: **σ** is the compression strength, R is the ratio of the bioactive glass ceramic. The R-Sq (98.79%), R-adj (98.19%).

The prediction models for flexural strength, density, porosity, and hardness are made by taking them as dependent variables. While, the ratio of the bioactive glass ceramic as an independent variable (input) as follows: For flexural strength, **σ**f = 5.227 + 0.0620R. The R-Sq (91.70%), R-adj (87.55%). For density, **ρ**b= 58.25 + 0.129R The R-sq (18.50%), R-adj (0.00%). For porosity, P= 29.424 - 0.1932R. The R-sq (97.97%), R-adj (96.95%).For hardness, Hv= 8.203 + 0.1153R The R-sq (97.97%), R-adj (96.95%).

D. ANALYSIS THE REGRESSION EQUATION FOR:

- Compression strength:

term	Coef	SE Coef	T-Value	P-Value	vif
constant	6.0508	0.0796	76.03	0.000	
Concentration	0.06881	0.00538	12.79	0.006	1.00

- *For flexural strength:*

term	Coef	SE Coef	T-Value	P-Value	Vif
constant	5.227	0.195	26.78	0.001	
concentration	0.0620	0.0132	4.70	0.042	1.00

- *For density*

term	Coef	SE Coef	T-Value	P-Value	Vif
constant	58.25	2.83	20.60	0.002	
concentration	0.129	0.191	0.67	0.570	1.00

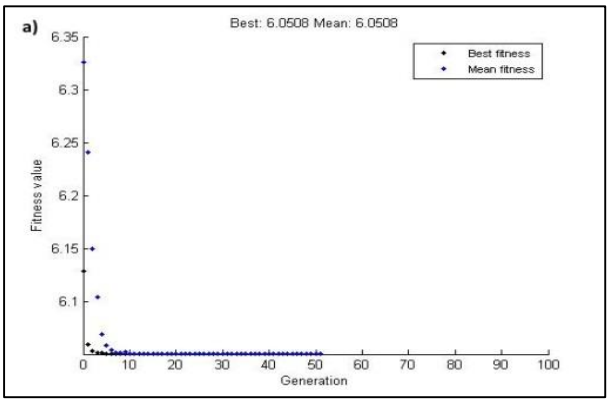
- For porosity

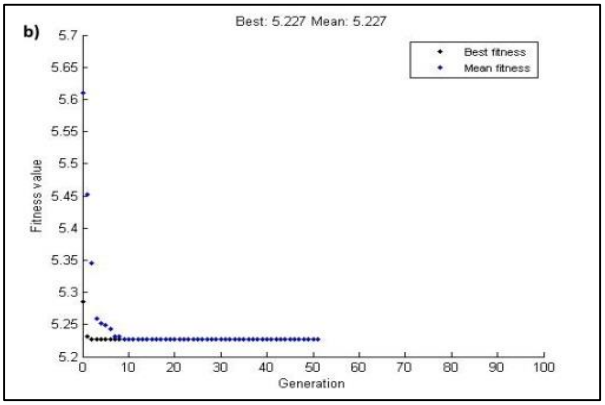
term	Coef	SE Coef	T-Value	P-Value	Vif
constant	29.424	0.540	54.49	0.000	
concentration	-0.1932	0.0365	-5.29	0.034	1.00

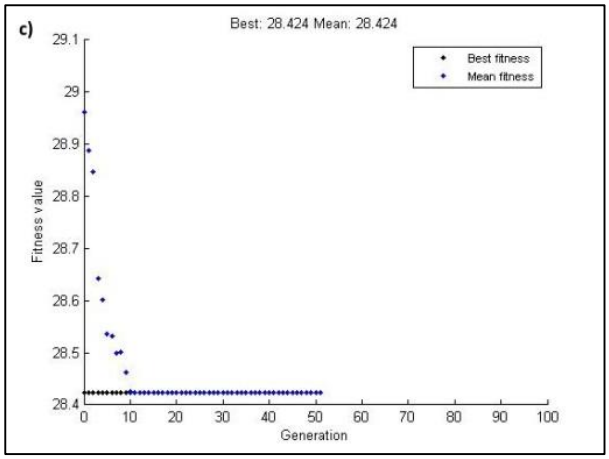
- For hardness

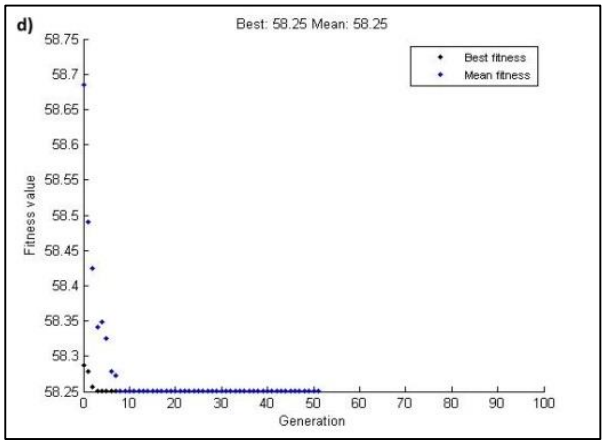
term	Coef	SE Coef	T-Value	P-Value	Vif
constant	8.203	0.174	47.23	0.000	
concentration	0.1153	0.0117	9.81	0.010	1.00

The genetic algorithm option from the optimization (regression) via Matlab software was used to present the solution of the optimization problem, the best individual values and generation versus fitness value are sown in the fig3.









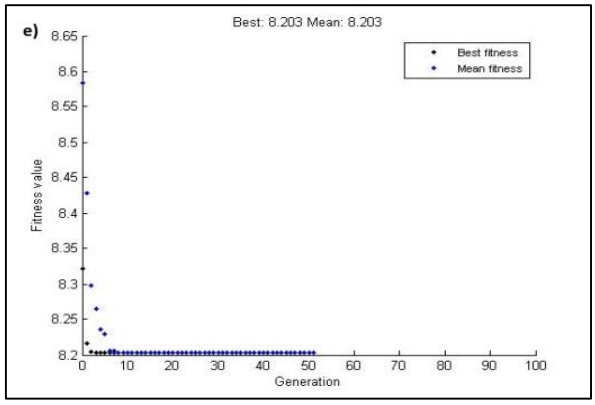


Fig3: the regression versus the fitness value for a) compression, b) flexural strength, c) porosity, d) density, and e) hardness.

IV CONCLUSION

HA/bioactive glass ceramic was successfully prepared by using the solid state reaction. This is because, when adding 5% bioactive was glass ceramic. The phase produced was beta-tricalcium phosphate (Ca_3 $(PO_4)_2$), which means that it react only as a sintering aid and helps the decomposition of HA. While, when are adding 15% and 25% bioactive glass ceramic, sodium calcium phosphate were obtained ($Na_3Ca_6(PO_4)_5$). The same phase was obtained when was used 25%bioactive glass ceramic and HA synthesized from bovine bone.

The result obtained from genetic was 6.0508 compared with the real values range (6-7.8). The best result for the flexural strength 5.227 compared with the range of real values (5-6.7), 28.424 for porosity compared with the range (30-25), 58.25 for density compared with the values (60-54), and 8.203 for hardness compared with the real values (8-11).

V FUTURE WORK

A Study of the morphology of the sample by scanning electron microscope before and after soaking in (SBF).

Put the samples in the simulated body fluid (SBF) and study its effect upon the biological properties.

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