SYNTHESIS AND CHARACTERIZATION OF HYDROXYAPATITE-CALCIUM HYDROXIDE FOR DENTAL COMPOSITES

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In this work hydroxyapatite was precipitated from calcium hydroxide and phosphoric acid. Calcium hydroxide forms in addition to hydroxyapatite in samples with calcium to phosphorus ratio more than the stoichiometric one (Ca/P = 1.67). The effect of changing the pH of the precipitation solution was investigated. Changing the pH of solution had no effect on the amount of compounds formed in the structure. In contrast, an increase in Ca/P ratio increases the total amount of calcium hydroxide which is suitable for dental composite application.

INTRODUCTION

Hard tissues repair has always been one of the most important issues in biomedical engineering [1]. Various biomaterials, such as different kinds of calcium phosphates, bioglasses, biopolymers, and synthetic polymers, have been studied in this area [2-5]. The most important properties of a biomaterials used for bone repair are osteoconductivity and osteoinductivity. Among biomaterials, calcium phosphate ceramics have been widely used in orthopedics and dentistry [6-11] and Hydroxyapatite (HA) is the most researched calcium phosphate biomaterial in hard tissue repair applications. HA comprising 60 to 70 percent of the bone tissue which reduces inflammation and immunogenic reactions at implantation site. Osteoconductivity is another property of HA and promotes osseointegration and contributes to osteogenesis.Some clinical applications of hydroxyapatite are: Bone defects repair, bone augmentation, and coatings for human body metallic implants [1-3, 9-20, 23, 25]. Therefore tendency to continue the research on HA as a biomaterial for hard tissue applications is growing.

There are various ceramic processing methods for HA synthesis such as precipitation, sol-gel, hydrothermal processing, etc. [4, 6, 10]. In this work, HA was precipitated from phosphoric acid and calcium phosphate and the stoichiometric calcium to phosphorus (Ca/P) ratio for HA synthesis is 1.67. Higher Ca/P ratio results in the formation of calcium hydroxide in addition to HA. The effect of different pH of the precipitation solution on the properties of HA was investigated. Properties of synthesized powders were characterized for use as dental composites.

EXPERIMENTAL

Samples were prepared by aqueous precipitation reaction. Suspensions of 0.5 molar calcium hydroxide were prepared using calcium hydroxide powder (Merck). The suspensions were degassed, vigorously stirred and heated to 40°C for two hours. A 0.3 molar phosphoric acid (Merck) solution was added drop by drop to the calcium hydroxide suspension and kept for an hour.

The obtained mixture was then stirred by a magnetic stirrer for 2 hours at the speed of 200 rpm, aged for 24 hours at room temperature and centrifuged to complete the precipitation process. The precipitate was dried at 100°C during 12 hours. The procedure was carried out with different ratios of calcium hydroxide and phosphoric acid to obtain desired Ca/P ratios. The pH of the mixture was adjusted using ammonium hydroxide (Merck). Powders were characterized using EDS, FTIR, and XRD spectra. Scanning electron microscopy (SEM) was used to characterize the morphology of the specimens.

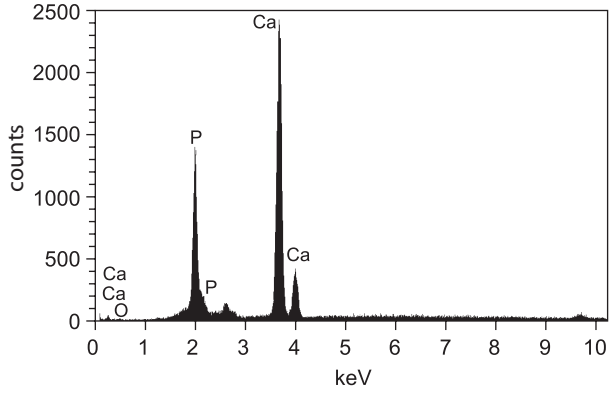
RESULTS AND DISCUSSION

Data gathered from the synthesized samples is shown in Table 1.

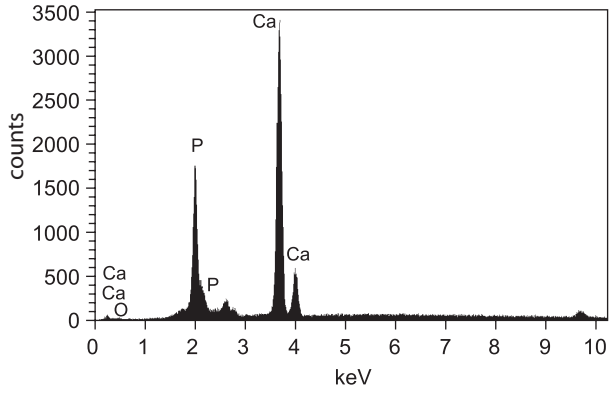
EDS spectra (Figure 1) shows the Ca/P ratio of the samples A, D, and G. XRD spectra of the samples are shown in Figures 2,3, and 4.As can be seen in the figures, calcium hydroxide is formed in addition to HA in samples with Ca/P ratio more than 1.67 (the stoichiometric ratio). FTIR spectra shows the formation of OH bonds due to the existence of calcium hydroxide in the structure (Figure 5).

Table 1. Characterization of the synthesized powders at 40°C..

Sample code	Ca/P ratio	precipitation pH
A	1.67	7.5
B	1.67	9.5
C	1.67	11.5
D	1.8	7.5
E	1.8	9.5
F	1.8	11.5
G	1.9	7.5
H	1.9	9.5
I	1.9	11.5



a)



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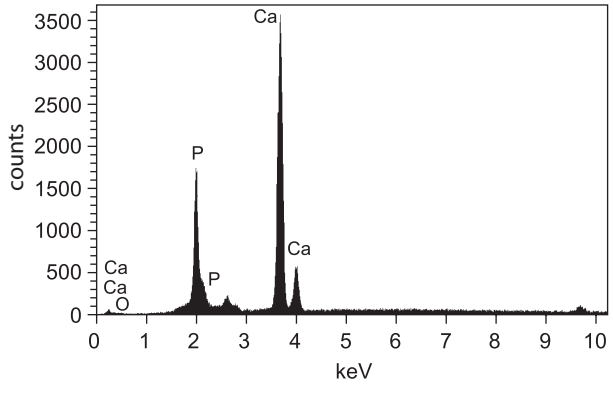
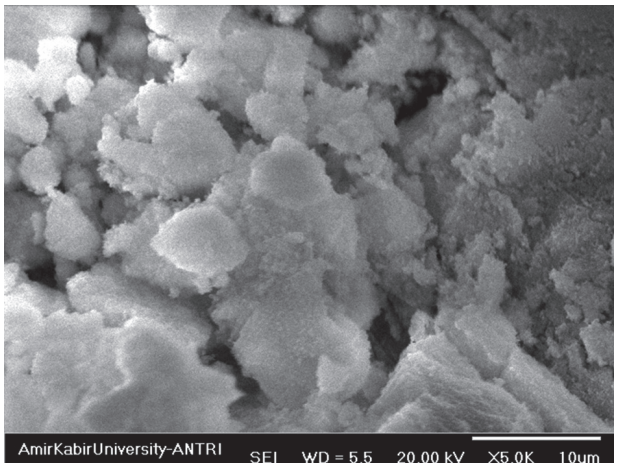


c)

Figure 1. EDS spectra of the samples with Ca/P ratio: a) 1.67, b) 1.8, c) 1.9.

Figure 2 depicts SEM micrographs of samples with the Ca/P ratios of 1.67, 1.8, and 1.9. Agglomerated hydroxyapatite crystals with needle-like and plate-like shapes can be observed in the figure.



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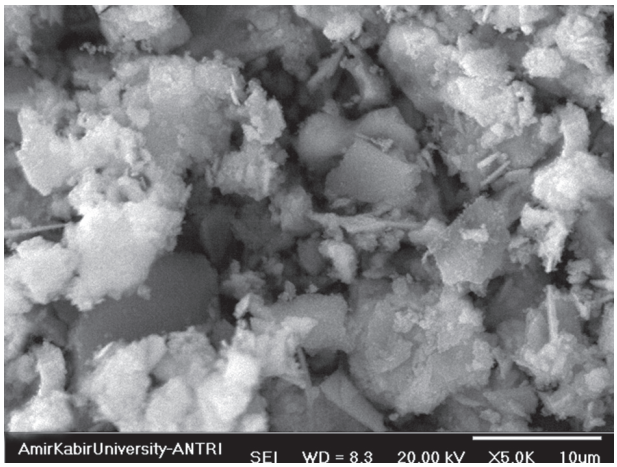

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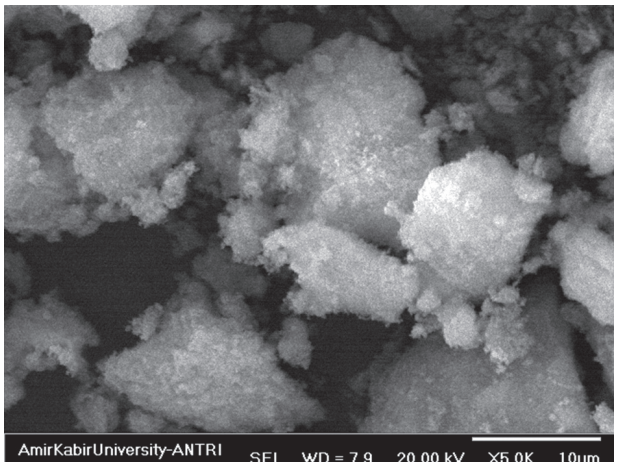

c)

Figure 2. SEM micrographs of the samples with Ca/P ratio: a) 1.67, b) 1.8, c) 1.9.

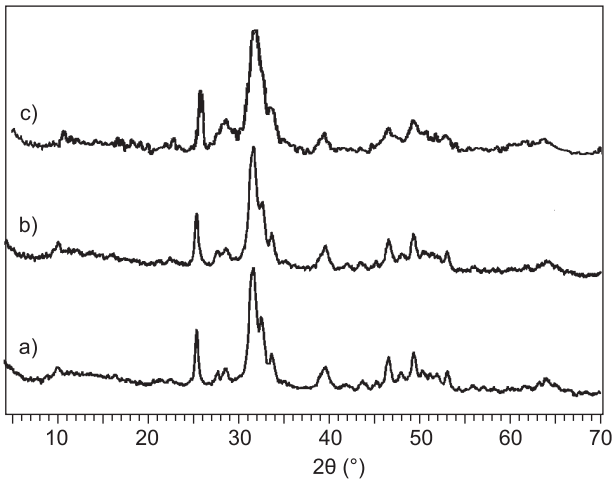

Figure 3. XRD spectra of the samples with Ca/P ratio 1.67 synthesized at pH: a) 7.5, b) 9.5, c) 11.5.

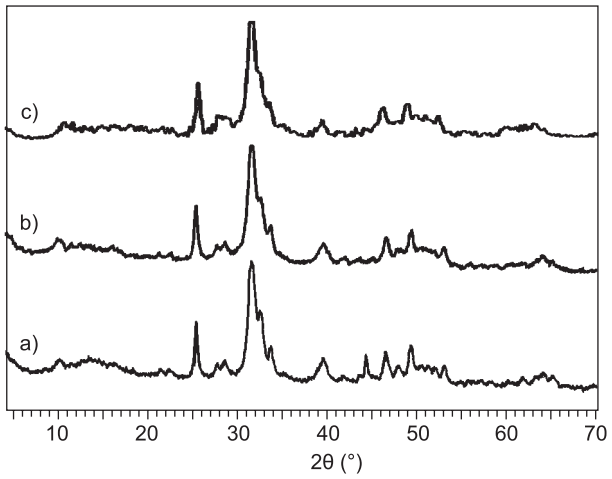

Figure 4. XRD spectra of the samples with Ca/P ratio 1.8 synthesized at pH: a) 7.5, b) 9.5, c) 11.5.

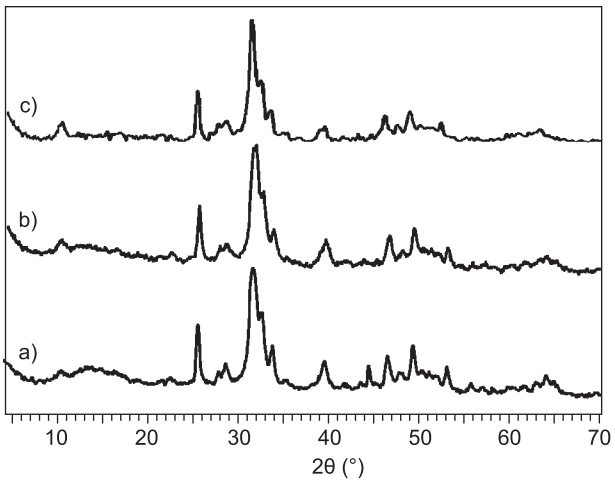

Figure 5. XRD spectra of the samples with Ca/P ratio 1.9 synthesized at pH: a) 7.5, b) 9.5, c) 11.5.

According to the results, in samples with Ca/P ratio more than the stoichiometric one, calcium hydroxide forms in addition to HA. The presence of calcium hydroxide in the structure is ideal for dental composites applications. In fact, this leads to an increase in the amount of free calcium hydroxide and its dissolution in the environment which increases pH and reduces bacteria growth. Calcium hydroxide has antibacterial characteristics, enhances enzymes and growth factors release, and increases the rate of drug release [24, 25].

As shown in XRD spectra (Figures 2,3, and 4), any change in the pH of the precipitation solution have no effect on the amount of the formed compounds but by increasing the Ca/P ratio, the amount of calcium hydroxide increases which is suitable for use in dental composites.

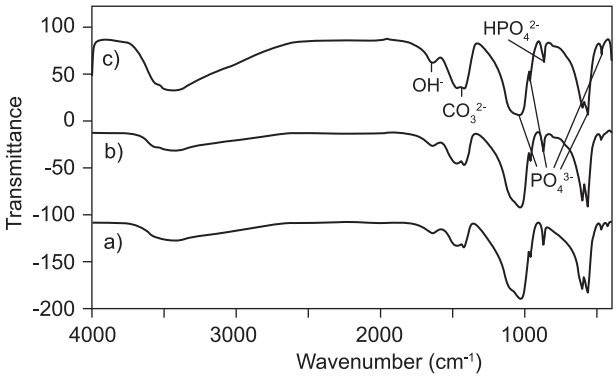

Figure 6. FTIR spectra of the samples with Ca/P ratio: a) 1.67, b) 1.8, c) 1.9 synthesized at pH = 11.5.

CONCLUSION

Calcium hydroxide is formed in the structure of hydoxyapatite precipitated from calcium hydroxide and phosphoric acid with Ca/P ratio more than the stoichiometric one (1.67). The presence of calcium hydroxide in the structure leads to an increase in the amount of free calcium hydroxide and its dissolution in the environment which increases pH and reduces bacteria growth. Calcium hydroxide has antibacterial characteristics, enhances enzymes and growth factors release, and increases the rate of drug release.

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