

SONOCHEMICAL SYNTHESIS OF HYDROXYAPATITE USING HOMOGENIOUS PRECIPITATION AGENT- UREA

M. Jevtić^{1a}, M. Mitrić^{2b}, N. Ignjatović^{1c}, D. Uskoković^{1d}

¹Institute of Technical Sciences of the Serbian Academy of Sciences and Arts, Belgrade, Serbia

²The Vinča Institute of Nuclear Sciences, Laboratory for Theoretical and Condensed Matter Physics, Belgrade, Serbia

Introduction

Synthetic hydroxyapatite- HAP is biomaterial with excellent biocompatibility, bioactivity and similarity to live bond. Its mechanical properties allow using this biomaterial for medical and dental applications [1-4].

Ultrasonic irradiation is a novel precipitation method for nanocrystalline HAP preparation [5]. The chemical effects of ultrasound derive primarily from acoustic cavitations (the formation, growth and collapse of bubbles) [6]. Synthesis of HAP nanoparticles in ultrasonic precipitation and influence of temperature, [Ca²⁺], Ca/P ratio and ultrasonic power on its morphology and crystalline has been recently reported by Li-yun, Chuan-bo Zhang and Jian-feng Huang Cao [5,7]. In this work we analyzed ability for HAP synthesing using homogenous precipitation method in the field of ultrasonic irradiation. We researched ability of using urea for precipitation agent. We defined basic synthetic parameters: temperature, concentration, power of ultrasound field, time and dynamics of ultrasound field effect.

Materials and methods

In the first step, Ca(NO₃)₂ and NH₄H₂PO₄ were dissolved in distilled water, by stirring on stirrer, to make homogeneous solutions with Ca/P ratio at 2 and [Ca²⁺] at 0.02M. Solutions were pounded into reaction vessel and heated at 88°C.

Next, different volumes of 12% urea were added to regulate pH value and to allow homogeneous precipitation (Table 1). Parameters of ultrasonic liquid processor were set (Table 2). The sonic horn, made of Ti, was dipped into solution and ultrasonic irradiating started.

Precipitates were aged for 15 h, separated from the mother liquid and washed with distilled water by centrifugal processing method. Sludges were dried in dryer for 5h at 80°C.

Results and discussion

Figure 1 shows XRD of powders synthesized with different amount of precipitation agent. Samples A, B and C consist two phases- HAP and OCP (octa calcium phosphate). OCP is intermediate for HAP synthesis. Sample D is monophasic HAP which indicate that reaction is finished. Graph 1 shows increasing of HAP content with increasing of urea amount i.e. concentration of OH⁻ ions according figure 1.

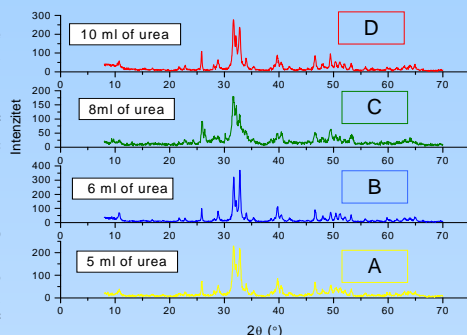


Figure 1: XRD spectra of samples

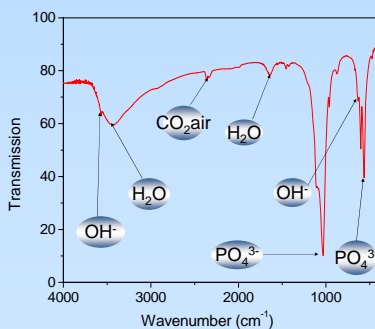


Figure 2: IR spectra of HAP monophasic

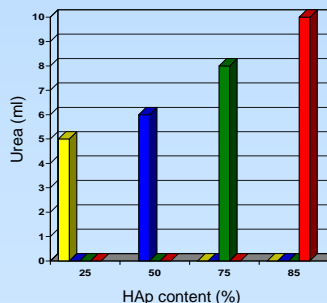
Table 1: Parameters of ultrasonic liquid processor

T(°C)	t (min)	amp.(%)
60-80	2	80
80-90	3	80
80-90	3	80
82-90	3	80
80-90	3	85

Figure 2 represents IR spectrum of sample D. The characteristic bands of HAP are exhibited:

632.42 cm⁻¹ and 563.88 cm⁻¹ for PO₄³⁻ bending, 961.76 cm⁻¹, 1108.1 cm⁻¹ and 1032.5 cm⁻¹ for PO₄³⁻ stretching, 3568.8 cm⁻¹ and 632.43 cm⁻¹ for OH vibrations. The broad band at 2500-3600 cm⁻¹ and band at 1642.5 cm⁻¹ are reflection of absorbed and associated water in HAP powder. Bands at 874.93 cm⁻¹ and 1455.1 cm⁻¹ are vibrations of CO₃²⁻ group which presence is normal concerned with organic precipitation agent. The band at 2359.2 is air CO₂ vibration.

There are no bands in IR spectrum or peaks in XRD patterns belong to urea or any urea composition. That indicates that urea is totally decomposed.



Graph 1: Relationship between urea and HAp content

Table 2: Volume of 12% urea in different samples

sample	urea (ml)
1	100
2	15
3	10
4	8
5	6
6	5



Conclusion

In this work we synthesized Hap using sonochemical method. As starting components we used Ca(NO₃)₂, NH₄H₂PO₄ (Ca/P ratio was 2) and homogeneous agent was urea (NH₂CONH₂). With following parameters: T= (80-90)°C, P= 600W, E= 400 kJ and ratio Ca:P:Urea = 2:1:9 we obtained HAP and OCP mixture. With increasing of urea content, HAP percentage increased, too. In the final product we obtained HAP monophasic. Optimal parameters were: T= (80-90)°C, P= 640W, E= 500 kJ and ratio Ca:P:Urea = 2:1:10. Homogeneous precipitation method in the field of ultrasonic irradiation is very effective for monophasic HAP synthesis.

References

- [1] X. Y. Pang, X. Bao, Cancer Letters, 107 (1996) 11-18
- [2] K. J. L. Burg, S. Porter, J. F. Kellam, Biomaterials, 21 (2000) 2347-2359
- [3] A. K. Dash, G.C. Cudworth, Journ. Pharmacol. Toxicol. Methods, 40 (1998) 1-12
- [4] W. Suchanek, M. Joshimura, Journ. Mater. Res., 13 (1998) 94-117
- [5] L. Cao, C. Zhang, J. Huang, Ceramics International, 31 (2005) 1041-1044
- [6] K. S. Suslick, G. J. Price, Annu. Rev. Mater. Sci., 29 (1999) 295-326
- [7] L. Cao, C. Zhang, J. Huang, Materials Letters, 59 (2005) 1902-1906