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Short communication Pomacea sp shell to hydroxyapatite using the ultrasound–microwave method (U–M)

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February 2014; received in revised form 28 March 2014; accepted 28 March
2014 Available online 3 April 2014 Abstract The shell of

Pomacea sp has a potential application as raw material for the production of hydroxyapatite (HA). The
preparation of HA was carried out at low temperature using a combination of ultrasound and microwave
assisted radiation methods. In this experiment, hydroxyapatite was synthesized using citric acid as an
organic modifier and Na2HPO4 as the source of PO34 . The obtained products were analyzed by
X-ray Diffraction (XRD), Fourier Transform Infrared (FTIR), and Scanning Electron Microscope (SEM) to confirm the morphology and crystalline phase of HA. & 2014 Elsevier Ltd and Techna Group S.r.l. All rights reserved. Keywords: Hydroxyapatite; Pomacea sp.; Characterization; Microwave 1. Introduction Over decades thousands of studies have been devoted to find or explore the production of various kinds of chemicals using natural or biomass resources as the raw materials [1,2]. Hydroxyapatite (Ca$_{10}$(PO$_4$)$_6$(OH)$_2$) is an inorganic material that is widely used in various biomedical applications. It is used primarily by orthopedists and dentists because of its close similarity to the inorganic mineral component of bone and teeth [3,4]. Because this material has so many applications in the biomedical area, considerable research has been directed towards alternative methods for its preparation. Many different raw materials and methods can be used for the preparation of HA. Raw materials have been used included natural sources such as fish scale [5], eggshells [6–8], animal bone [9,10] etc. While, the methods include solid state reactions [7], wet nCorresponding authors. nnCorresponding author. Tel.: þ 62 31 3891264; fax: þ 62 31 3891267. E-mail addresses: felyciae@yahoo.com (F.E. Soetaredjo), yhju@mail.ntust.edu.tw (Y.-H. Ju), suryadiismadji@yahoo.com (S. Ismadji).

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chemical precipitation [11–13], and mechanic-chemical methods [14,15]. The wet chemical precipitation process is used to prepare HA powder because it is simple, versatile and cheap [16]. In the wet precipitation method, the precipitated powder is typically calcined at high temperature up to 1100 °C in order to develop a well-crystallized HA phase [17]. The chemical reactions take place between calcium and phosphorus ions under a controlled pH and solution temperature.

[18,19].

Slow titration and diluted solution must be used to improve chemical homogeneity within the system.

[20]. Pomacea sp. (also known as golden snail) is a pest in rice farming areas. Even though this mollusk is considered as pest, in countries such as Indonesia, Philippines and Thailand, the meat of this creature can be utilized as the food source for human consumption because it has exceptionally high protein content. The Pomacea sp. shell contains a high amount of calcium carbonate (96.83%) and a small amount of metallic
Impurities (3.17%) [21]. The shell of this mollusk is an excellent source of raw material for the preparation of HA since it contains such a high amount of calcium carbonate. 11454 J.N. Putro et al. / Ceramics International 40 (2014) 11453–11456 A combination of wet synthesis, ultrasound, and microwave irradiation methods were used to synthesize HA from the Pomacea sp. shell. In the preparation of HA, it is crucial to control the shape and particle size distribution of HA by proper mixing and the use of ultrasound during the mixing. This reduces particle aggregation. Microwave irradiation improves the physical property of HA and shortens processing time. At present, there are no other studies using a combination of ultrasound and microwave irradiation methods to prepare HA using Pomacea sp. shell as the raw material.

2. Experimental

2.1. Material Pomacea sp shell waste was collected from Pati, Central Java, Indonesia. The shell waste was cleaned using tap water and immersed in boiling water for 30 min to remove the surface contaminants and organic components. Subsequently, the shell waste was dried in the oven and crushed into powder. The calcination of the shell powder was conducted at a temperature of 900 1C for 3 h. For the preparation of HA from Pomacea sp shell waste, the following chemicals were used: citric acid monohydrate (C6H8O7 H2O) with the purity of Z 99.0% (Sigma Aldrich), Sodium Hydroxide (NaOH) with the purity of Z 98.0% (Sigma Aldrich), sodium hydrogen phosphate (Na2HPO4) with the purity of Z 99% (BioXtra, Sigma Aldrich). All the chemicals were directly used without any further purification process.

2.2. Preparation of HA The preparation of hydroxyapatite (HA) using Pomacea sp shell waste as the precursor was carried out using ultrasound and microwave irradiation methods. One gram of calcined Pomacea sp shell powder was mixed with distilled water and stirred for 30 min. The solution was then mixed with citric acid at different concentrations with constant stirring condition. Subsequently the mixture was placed into temperature controlled ultrasound equipment. The mixture was subjected mechanical stirring within the ultrasound device, and solution of sodium hydrogen phosphate (Na2HPO4) was added into the mixture (ratio Ca/P 1.67). The temperature of the process was kept constant (651, 751, and 85 1C). After the addition of Na2HPO4 was completed, the reaction mixture was stirred further for varying times within the ultrasound device. The reaction mixture was kept constant at pH 11 by addition of NaOH solution. After the ultrasound process was completed, the mixture was refluxed in a domestic microwave for 5 min at 700 W. Subsequently the resulting mixture was filtered and solid particles were repeatedly washed with distilled water and dried in an oven at 100 1C for 4 h. The powders were then calcined at 1000 1C.

2.3. Characterization of HA The resulting products were characterized using X-ray diffraction (XRD), scanning electron microscopy (SEM), and Fourier transforms infrared (FTIR) methods. Diffraction spectra of the HA samples were obtained on a Philips X’Pert diffractometer in 2θ angle between 51 to 601. CuKα was used as a radiation source. The crystalline phase of HA was determined by comparing the diffraction spectra with standard data for HA from International Center for Diffraction Data (ICDD-PDF no: 09-0432). The surface morphology of the HA samples was observed on a field emission scanning electron microscope JEOL JSM 6390. The SEM apparatus was equipped with a backscattered electron detector. The accelerating voltage of the detector was set at 20 kV at working distance of 12 mm. Qualitative analysis of HA samples was carried out by FTIR method using the KBr technique. The FTIR spectra of the samples were obtained in the wavenumber range of 4000 to 500 cm using a Shimadzu 8400s FTIR instrument.

3. Results and discussion

The XRD patterns of two samples HA obtained at two different process conditions (Citric acid 1 M, 75 1C, 30 min; and citric acid 2 M, 75 1C, 30 min) are given in Fig. 1. This figure clearly shows that three crystalline phases are observed in both samples. These crystalline phases are hydroxyapatite, calcium oxide, and calcium hydroxide, and the main phase is hydroxyapatite. The presence of small quantity of calcium oxide indicates that the transformation of
calcium oxide into HA was not complete—possibly due to low temperature and short reaction time or short exposure to microwave irradiation. During the cooling process of calcined HA, the basic site of unconverted CaO adsorbed the water from the ambient atmosphere, and the reaction between adsorbed water and basic site of CaO to form Ca(OH)2 could not be avoided. The peaks characteristic of HA in both samples (Fig. 1) are consistent b Hydroxyapatite (211) CaCO3 Ca(OH)2 (002) a (202) (310) (222) (004) 10 20 30 40 50 60 2θ Fig. 1. The XRD patterns of HA from Pomacea sp. Shell, (a) citric acid 1 M, 75 1C, 30 min, (b) citric acid 2 M, 75 1C, 30 min. J.N. Putro et al. / Ceramics International 40 (2014) 11453–11456 11455 Fig. 2. SEM images of HA ((a) citric acid 1 M, 75 1C, 30 min, (b) citric acid 2 M, 75 1C, 30 min). with standard data of HA from International Center for Diffraction Data (ICDD-PDF no: 09-0432). The XRD patterns of the synthesized HA from Pomacea sp. shell correspond to crystal orientation planes of (0 0 2), (2 1 1), (2 0 2), (3 1 0), (2 2 2), (3 1 2), and (0 0 4). The combination of ultrasound and microwave to assist in the wet synthesis of HA has advantage compared to traditional wet chemical precipitation. It is much faster (shorter process- sing time) due to higher heating rate. As mentioned by Nazir et al. [4] the microwave irradiation process enables fast volumetric heating that ensuring uniform heating with almost no thermal gradient. High temperature and rapid heating rate accelerates the formation of thermodynamically stable crystal phase and avoids the formation of metastable phases [4]. While the ultrasound process reduces the particle aggregation during the reaction, shock waves produced by cavitation shortens the contact time among particles and prevents their bonding together [22].

In the synthesis process, ultrasonic irradiation caused formation, growth and implosive collapse of bubbles, creating localized spots. These spots alternate low-pressure and high-pressure waves in liquids, leading to the formation and violent collapse of small vacuum bubbles and breakdown the Wavenumber Wavenumber Fig. 3. FTIR spectra of HA ((a) citric acid 1 M, 75 1C, 30 min, (b) citric acid 2 M, 75 1C, 30 min). particle aggregate [23] and as the result different particle size (0.46–2.5 μm) was obtained. The concentration of citric acid seems has a small effect on the crystallinity of HA as seen in the XRD diffraction pattern (Fig. 1), both of the samples exhibit similar diffraction patterns. During the reaction process, the presence of citrate groups in the solution surrounds the calcium ions and develops stable and supersaturated solutions [24]. Since the citrate groups have strong coordination ability, the spontaneous precipitation of calcium phosphate due to the reaction between calcium and phosphate is retarded [24]. The SEM micrographs of synthesized HA are shown in Fig. 2. The hydroxyapatite synthesized from Pomacea sp. shell consists of a mixture of particles with different sizes. The crystal structure of HA is also observed from this figure. The FT-IR spectrum of synthesized HA is shown in Fig. 3. The vibrations of characteristic PO34 appear in two vibrational modes at bending 1044–1098 cm 1(v3) and 540–580 cm 1(v4) 11456 J.N. Putro et al. / Ceramics International 40 (2014) 11453–11456 which indicate the formation of the hydroxyapatite phase. The bands of CO23 were spotted at peak of 869–876 cm 1 and 1432–1447 cm 1, FTIR spectra. These are the characteristic peaks corresponding to OH vibrations at librational mode 694–702 cm 1 and its stretching vibrations at range 3525–3745 cm 1. The spectrum of 2500–3000 cm 1 corresponds to H2O in the synthesized HA. These peaks indicate the form of b- type HA mineral. 4. Conclusion Pomacea sp. shell has been successfully used as raw material for the preparation of hydroxyapatite using an ultrasound–microwave method. The synthesized HA was characterized by SEM, XRD and FTIR, and the product confirmed as b-type carbonated HA. Acknowledgements The authors
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