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Manufacture And Characterization Of Hydroxyapatite From Quail Eggshell Using Precipitation Methods

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***Abstract*— Hydroxyapatite (HA) is a biomaterial used to treat bone defects. The utilization of HA in medicine is currently significantly increased this is because HA has high biocompatibility properties when used as a bone graft. Various kinds of bone grafts from HA are currently available in the market. However, its use is often bumped at a high price and is an imported product. This will ultimately increase the burden of financing that must be borne by hospitals and the government. HA sources can be obtained through extraction from natural products, such as eggshells. The purpose of this study was the creation and characterization of hydroxyapatite from quail egg shells using precipitation methods. The research stage consists of 4 stages, Stage 1 is the calcination of quail egg shells so that CaO compounds are obtained. Stage 2 is the synthesis of HA compounds from CaO compounds produced in the previous stage using wet precipitation or deposition methods. Stage 3 is sintering HA compounds produced in the previous stage using a furnace for five hours at a temperature of 900 ºC. Stage 4 is the characterization of the HA compounds produced. The characterization is done using XRD and SEM. Hasil yang diperoleh yaitu Efisiensi dari senyawa HA yang dihasilkan sebesar 52,34%. Based on the characterization carried out using XRD it was obtained that the HA compound was successfully synthesized this is characterized by the peak of the quail eggshell difaktogram that is the same as the peak of the standard HA difractogram which is found at an angle of 2θ: 26.00ºC, 31.90ºC, 32.31ºC, 33.02ºC, 34.19ºC, 46.81ºC, 49.59ºC with the crystal phase and the size of the lattice parameters, namely the lattice a = b = 9.4234 Å and c = 6.8801 Å. But in addition to the peak of HA there are also other peaks that show the existence of the impurities phase at 2θ : 10.01°, 21.85°, and 53.01°. Based on SEM characterization found that the size of the resulting HA particles is not homogeneous, meaning that there is a difference in the smallest particles with a size of 1,497 µm and there are the largest particles with a size of 60.98 µm. If observed the shape of a single particle tends to be round (Shperical). Thus it was concluded that the manufacture and characterization of hydroxyapatite from quail egg shells using precipitation methods has been successfully carried out.**

Keywords— Hydroxyapatite; Quail Egg Shell; Precipitation; Bone Defects.

I. INTRODUCTION

Bone defects are a condition of loss of part of the bone, which can be caused by trauma or certain post-surgery, such as surgical removal of bone tumors. Defects in bones that are not handled properly and correctly will cause disability in patients later in life. In orthopaedic practice, bone *graft* is one of the procedures performed for handling defect cases on the bone, to restore the missing bone part. This procedure is proven to be effective and able to help in the healing process and strengthen bone stability. Hydroxyapatite (HA) is a material that is widely used as a *bone graft* to repair defects in bones. HA with the chemical formula $Ca_{10}(PO_4)_6(OH)_2$ is one of the calcium phosphate compounds and belongs to the group of apatite minerals that are currently being widely developed. With the high need for this biomaterial, HA synthesis becomes a thing that is quite useful to do.

Ha manufacturing generally uses solids or calcium oxide (CaO) powders. In medical application, it is known that the body's response to implanted materials in the form of calcium phosphate compound groups is related to the mass ratio of Ca/P and

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crystallity of the compound. Therefore, the selection of hydroxyapatite manufacturing technology needs to consider whether the product produced from that technology is close to the required specifications or not. For implantable materials, the desired specification of hydroxyapatite is one that has a Mass Ratio Ca / P of 1.67 and has the same crystal arrangement as animal / human bones [1]. The advantage of hydroxyapatite material is that it has a crystalline composition and structure similar to bone and is currently the most widely used material in biomedical applications [2]. Calcium phosphate has complex properties, can be in various phases, can also be in the form of a solid solution. In addition, calcium phosphate can be in the form of nonstoikiometry in the presence of impurities that replace lattice ions in crystals. In general, calcium phosphate is in the form of amorphous and various crystals. HA is used because it is the most stable biokeramic calcium phosphate.

The application of HA as a bone graft shows satisfactory results due to its osteoconductive and bioactive abilities.Various kinds of bone grafts from HA are currently available in the market. However, its use is often bumped at a high price and is an imported product. This will ultimately increase the burden of financing that must be borne by hospitals and governments. HA sources can be obtained through extraction from natural products, such as eggshells. Eggshells are currently a waste that is barely utilized. HA derived from eggshells has been shown to have good biocompatibility, low risk of disease transmission, easy to use and has abundant availability. The egg shell has the potential as a precursor in the manufacture of HA with economic value that is very affordable and has abundant availability.

Bone graft from eggshells becomes an alternative to the cost-effective and efficient bone graft option, which can be prepared in a very economical way. In addition, egg shells come from natural ingredients that are safe and easy to obtain.Eggshell waste has abundant availability and can be obtained at a very cheap cost. Bone grafts derived from eggshells have proven to be much more efficient and can cut healthcare costs. For comparison, the price of 5 cc imported bone graft with the trademark "X" ranges from Rp 1,870,000. Meanwhile, another trademark "Y" ranges from Rp 5,225,000. The manufacture of bone graft from eggshells has a production cost of Rp 520,000, which is able to produce 50 cc bone graft, which means that every 5 cc bone graft from the eggshell has a price range of only Rp 52,000. The use of bone graft from the egg shell becomes an effective and efficient solution for use in handling defect cases on the bone [3].

Increased protein consumption for the community is important, especially for people who cannot afford food from animals that are generally relatively expensive. Animal protein that is cheap enough for people to buy includes quail meat and eggs (Coturnix-coturnix japonica). As a food, quail eggs have better quality because they have a relatively higher protein content for each egg than chicken eggs [4].Increased consumer demand for quail eggs resulted in an increase in quail egg shell waste. Quail egg shells are household and industrial waste that has not been utilized to the fullest [5]. If quail egg shell waste is directly discharged into the environment it will damage the aesthetics of the environment, increase the volume of waste, and potentially pollute the environment. Quail egg shell waste also has the potential to cause pollution [6]. Quail egg shells contain $CaCO_3$ (55.46%), $MgCO_3$ (0.84%), $Ca(PO_4)_2$ (0.75%) and protein (amino acids) [7][8]. Unfortunately during this time the benefits of quail egg shells are still rarely even not so noticed at all. Though the calcium contained in quail egg shells is high enough that it can be used as a source of calcium in the manufacture of biomaterials such as HA. So the purpose of this study is the creation and characterization of hydroxyapatite from quail eggshell using precipitation methods.

II. Experiment

2.1. Material

Quail egg shells used as sources of calcium are collected as waste from various chicken egg utilization activities in the Campusarea, Universitas Kristen Indonesia (UKI). The chemicals used in the study were aquadest and diamonium hydrogen phosphate $(NH_4)_2HPO_4$.

2.2. Instrumentation

The tools used in the study are hammer, crussible, furnace, mortar, digital balance sheet, erlenmeyer tube, magnetic stirrer, pumpkin measuring, statif, aluminum foil, filter paper, funnel, burrete, furnace, petri dish, sample plastic, label paper, spatula, X-ray diffractometer (XRD) and electron microscope (SEM).

2.3. Procedure

Stage 1. The sample used was a quail egg shell around the UKI campus. The sample weighed about 300 gr then washed thoroughly and dried indoors for 24 hours. Then the egg shell is put in the furnace for six hours at temperatures of 700 °C and 1000 °C .This is done to remove impurities so that CaO compounds are produced. Next the sample is pounded with mortar until smooth.

Stage 2. Take a CaO sample then weighed 2.96 grams and transferred into an erlenmeyer tube then dissolved in 100 mL of aquadest. This solution is labeled the name of the first solution on the erlenmeyer tube. It is further weighed as much as 3.96 grams of diamonium hydrogen phosphate $(NH4)_2HPO_4$ and transferred into an erlenmeyer tube then dissolved in 100 mL aquadest. This solution is labeled the name of the second solution on the erlenmeyer tube. The precipitation method is done by mixing the second solution into the firstsolution. The mixing process is carried out on a magnetic stirrer by dripping with a flow rate of 10 ml/minute for 100 minutes using a burret and stirred at a speed of 350 rpm using a magnetic stirrer so that the solution becomes homogeneous. After the mixing process is completed then the solution is closed using aluminum foil, then deposited for 12 hours. The solution that has been precipitated is filtered using filterpaper, so that the sample sediment is obtained. Next washed the sediment using aquadest. The purification process is carried out 3 times. This is done to remove chemical compounds attached to the precipitate.

Stage 3. Then the precipitate is dried by putting it in the oven for 3 hours. So that the sediment will turn into powder. The resulting powder is then put in the furnace for five hours at a temperature of 900 °C.

Stage 4. Furthermore, the sample is ready to be characterized for the resulting HA purity meanalise. The characterization is done using XRD and SEM.

III. RESULTS AND DISCUSSION

3.1. Acquisition Hydroxyapatite

CaO compounds from the sintering stage with temperature variations of 700 °C and 1000 °C for 6 hours each are powders with characteristics such as table 1. Characteristics of CaO obtained from the calcination process at a temperature of 700 °C for 6 hours are in the form of blackish-gray powder, and the size of the powder produced roughly because the size of the powder is relatively large. Unlike the characteristics of CaO compounds obtained from the calcination process at a temperature of 1000 °C for 6 hours, namely in the form of powder with a bright white color and the size of the powder is smooth because the size of the powder is relatively small. So it can be concluded that the results of calcination at a temperature of 1000 °C for 6 hours have produced CaO compounds with good characteristics [9].

TABLE I. THE CHARACTERISTICS OF CALCINATION POWDERS EACH FOR 6 HOURS ON QUAIL SHELLS AT A TEMPERATURE OF 700 °C AND A TEMPERATURE OF 1000 °C.

Sample Type	Calcination Temperature	Color	Powder Size
Quail eggshell	700 °C	Blackish gray	Rough and large powder size
	1000 °C	Bright white	Smooth and small powder size

The efficiency of the calcination results of CaO compounds obtained from the sintering stage at a temperature of 1000 °C for 6 hours can be seen in table 2. It is seen that the mass of CaO compounds produced from the sintering process is relatively small, which is only about 10.81 grams from the initial mass of $CaCO_3$ compounds about 20.65 grams. This is caused by the sintering process of carbon compounds removed so that shrinkageoccurs. The efficiency of the CaO compound produced is 52.34%. This CaO compound will later be used as a material for making HA compounds by precipitation methods.

TABLE II. EFFICIENCY OF CAO COMPOUNDS RESULTING FROM CALCINATION OF QUAIL SHELLS AT A TEMPERATURE OF 1000 °C

Sample Type	Mass (gram)		Efficiency (%)
	$CaCO_3$	*CaO*	
Quail eggshell	20.65	10.81	52.34

HA compounds are produced from chemical reactions between CaO from the egg shell and diammonium hydrogenphosphate by wet method (precipitation) according to equation 1.

$$10Ca(NO_3)_{2(aq)} + 6(NH_4)_2H(PO)_{4(aq)} + 8NH4OH_{(aq)} \longrightarrow Ca_{10}(PO_4)6(OH)_{2(s)} + 20NH_4NO_{3(aq)} + 6H_2O \quad (1)$$

Synthesis (mixing) between CaO powder and diamonium hydrogen phosphate $(NH4)_2HPO_4$ with a Ca/P ratio of 1.67:1 will produce HA powder solids. The efficiency of the resulting HA compounds is indicated by table 3. CaO compounds used following the Ca/p ratio are 2.83 grams and diamonium hydrogen phosphate $(NH_4)_2HPO_4$ compounds used following the Ca/p ratio are 3.97 grams both of these compounds produce HA compounds of 4.43 grams. The resulting HA mass is noticeably much smaller than the total mass of CaO and diamonium hydrogen phosphate(NH4)2HPO_4.This is likely caused when the process of purifying HA deposits there are some HA compounds that are wasted. In addition, in the final stage there is also a sintering process that causes evaporation and mass development. The efficiency of ha compounds produced is 63.68%. This value is good enough because the value is above 50%.

TABLE III. EFFICIENCY OF HA SAMPLES WITH CAO SOURCES

Source CaO	Mass (gram)			Efficiency (%)
	CaO	*$(NH_4)_2HPO_4$*	*HA*	
Quail eggshell	2.83	3.97	4.43	63.68

3.2. HA Characterization results with XRD

To find out the characteristics of ha compounds obtained, XRD characterization is carried out.The difractogram of each HA synthesis is then compared to the standard HA difraktogram.

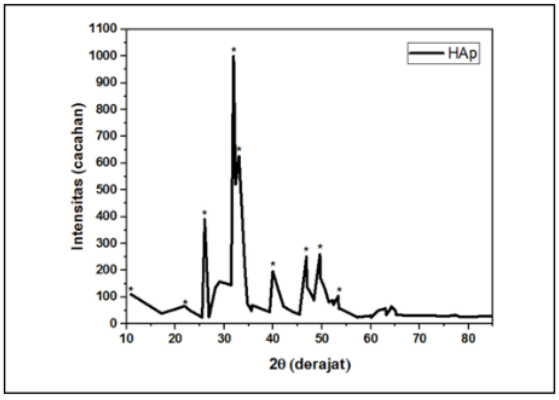

Fig. 1. XRD HA diffraction pattern from quail eggshell

The peak of the standard HA difactogram that quail egg shell samples also have is found at an angle of 2θ: 26.00 °C, 31.90 °C, 32.31 °C, 33.02 °C, 34.19 °C,46.81 °C, 49.59 °C (Figure 1). From the results of comparison with the standard HA difractogram

obtained information about the structure of ha crystals formed, namely hexagonal. The size of the grid parameter is grid $a = b =$ 9.4234 Å and $c = 6.8801$ Å. But in addition to the typical peaks of HA appear other peaks that show the existence of an impurities phase at 2θ of 10.01°, 21.85°, and 53.01°. But this impuritie is not analyzed further, because the intensity is low so it will not affect the quality of HA obtained.

3.3. HA Characterization results with SEM

The surface morphology of HA compounds resulting from the synthesis of characterization with SEM is shown in Figures 2 through 4. This characterization is done to get information such as particle size and uniformity of ha particles obtained. Figures 2 to 4 show the surface morphology of ha compounds produced with magnifications of 500x, 1000x and 5000x.

From figures 2 to 4, it was found that the size of the resulting HA particles is not homogeneous, meaning that there is a difference in the smallest particles with a size of 1,497 µm and there are the largest particles with a size of 60.98 µm.If observed the shape of a single particle tends to be round (Shperical). The HA particles shown in the image also help agglomeration or experience clumping according to research conducted by Hui in 2010 [10]. In addition, in terms of particle size is also still relatively large because generally the synthic HA compounds can be smaller, namely up to the nanometer scale as reported in previous research, namely the size of HA particles reported by Binnaz and Koca in 2009 which is about 210-410 nm[11].

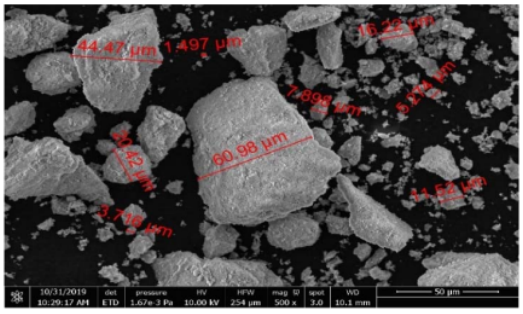

Fig. 2. SEM HA results with 500x magnification

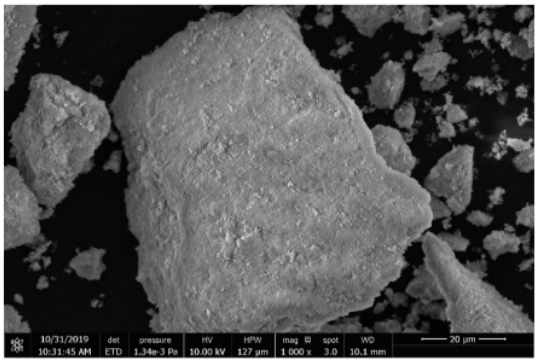

Fig. 3. SEM HA results with 1000x magnification

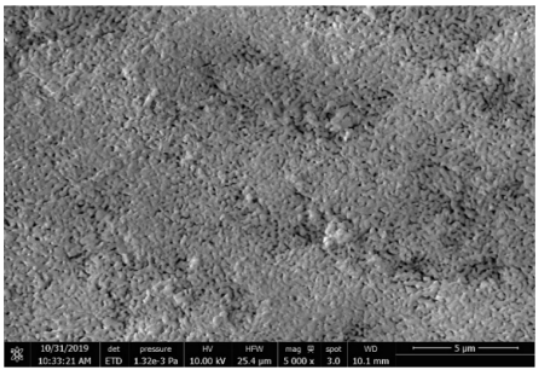

Fig. 4. SEM HA results with 5000x magnification

IV. CONCLUSION

The manufacture and characterization of hydroxyapatite from quail egg shells using precipitation methods has been successfully carried out. The efficiency of the ha compound produced is 52.34%. Based on the characterization carried out using XRD it was obtained that the HA compound was successfully synthesized this is characterized by the peak of the quail eggshell difaktogram that is the same as the peak of the standard HA difractogram which is found at an angle of 2θ: 26.00 ºC, 31.90 ºC, 32.31 ºC, 33.02 ºC, 34.19 ºC, 46.81 ºC, 49.59 ºC with the crystal phase and the size of the lattice parameters i.e. lattice a = b = 9.4234 Å and c = 6.8801 Å. But in addition to the typical peaks of HA appear other peaks that show the existence of an impurities phase at 2θ of 10.01 °, 21.85°, and 53.01°. Based on SEM characterization found that the size of the resulting HA particles is not homogeneous, meaning that there is a difference in the smallest particles with a size of 1,497 µm and there are the largest particles with a size of 60.98 µm. If observed the shape of a single particle tends to be round (Shperical).

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