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Effect of Pressure Distribution on Hydroxyapatite (HAp) Based Hybrid Composites Made from the Milkfish Bones

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Keywords: Milkfish bones, Chanos-chanos forsk, Hydroxiapatite, self-propagating high temperature synthesis and Hybrid composites.

Abstract. The milkfish bones (chanos-chanos forsk/CCF) which contain of Ca 4%, P 3% and protein 32% are a waste material. It is necessary to process the waste into a new product so that it can be used optimally. The milkfish content contains various metallic elements which can be combined metallurgically with aluminum (Al) as a matrix and Titanium (Ti)/magnesium (Mg) as a wetting element which will give an interface between materials. With that advantages, the milkfish bones can be used as a composite material with a content of more than one type of matrix that is being used as a composite hybrid. Milkfish bone material calcined at temperature of 500-700^{O}C, and made into Hydroxyapatite (HAp). This material was made as reinforced by mixing on Al with Mg and lightweight metal as well so that it can be processed into hybrid composite material. The study refers to the influenced of the stress distribution at the process of composites based on CCF calcination on the mechanical properties that was produced by self-high propagating temperatures synthesis (SHS) methods. The experiment used 3 types of sample with different composition each sample. The first type of sample composition used 80% HAp, 10% Al, 4% Mg, 3% Ti and 3% Cu with a pressing operated at 461.2 MPa which resulted in maximum mechanical hardness of 167-239 HV10. The second type of sample composition used 80% HAp, 10% Al, 4% Mg, 2% Ti, 2% Cu, and an addition of 2% Al_2O_3 ceramics with operational pressure reached at 691.7 MPa which resulted in a hardness value of 229-349 HV10. The third type of sample composition used 80% HAp, 10% Al, 4% Mg, 2% Ti, 2% Cu and 2% SiC as a reinforced with operational pressure reached at 921.8 MPa and hardness value of 359-375 HV10.

Introduction

Milkfish bones in the material science term are called chanos-chanos forsk (CCF). CCF can be processed into hydroxyapatite (HAp) material which is useful for biomaterial applications. HAp material which contains compound of $Ca_5(PO_4)_3(OH)_2$ is an inorganic material that shows good biocompatibility for bone connection material [1]. Aluminum (Al) is non-ferrous metal that has been used for special application due to its special properties such as good ductility, lightweight and low melting point, compared to other materials. In this case, Al can be used as a matrix or reinforcement on composites materials, especially hybrid composites [2-3]. CCF is processed using calcination at temperature of 500-700ºC to transforms CCF into HAp. By used of the right temperature, hybrid composite microstructures of several metal elements are combined with HAp. The appropriate process for fabrication of hybrid composites that is made from CCF is self-propagating high temperature synthesis (SHS). SHS is a unique method for making certain advanced materials such as metals, alloys, ceramics, intermetallic compounds and especially an advanced composite material.

The SHS process is based on combustion of exotherms that has a high purity of products. The aim of SHS process is to obtain a metastable phase in synthesis and simultaneous densification as well. Based on the heat released from the process reaction (fuel-oxygen/oxidizer), the reaction is able to regenerate itself. While the high temperature that is being produced is sufficient for the synthesis of the desired material product [4].

In previous research, the heating process of 2 hours resulted in undistributed of SiC as a reinforced at each phase that is caused by the weak SiC bond. Heating process for 3 hours resulted in SiC as a reinforcement was surrounded by metal elements and stacked by magnesium which acts a wetting agent. During heating process at 4 hour resulted in a complex distribution between HAp, Al and other elements. The 4-hour heating condition produces SiC reinforcement grains to grow larger. Although all of powder materials are surrounded by metal elements, there is still agglomeration between granules. This condition caused mechanical properties to decreased [5]. Pressure parameter gives significant influence on properties that is produced. Pressure analysis used a finite element simulation to measure the impact of cold pressing effects and to produce relatively high mechanical properties. The impact was micro cracks occurred at the center point area so that the pressure distribution was less uniform [6]. Pressing in hot conditions will produce a distribution of mechanical properties, especially uniform hardness. The temperature used does not exceed the upper limit of the recrystallization temperature of the material, so that the material is free from *micro-cracking* [7]. This paper explores the effect of pressure on the mechanical properties produced, the distribution and changes on morphology of grains formed by results of the SHS method. Autodesk fushion type 360 analysis software was used to find out the pressure distribution. The optimum pressure in each type of composition has a different value, so the value of the pressure distribution ensured optimal characteristics of mechanical properties. Autodesk fusion type 360 Software is also used to determine the sample composition that needs to be calibrated.

Material and Experimental Method

The Calcination process of CCF (milkfish bones) into hydroxyapatite material. Milkfish bones (CCF) were washed using water and soap to removed fat and dirt from the bones. After that, the bones were boiled for ± 60-90 minutes, this depends on the fat content and dirt that need to dissolved in the water. Next, the heating of calcination process was applied at temperature of 500-700°C to get the HAp. Reading scale in the furnace was 600°C but outside the temperature of 700°C. After the temperature has reached the exact temperature and changes in white occurred, the CCF sample that was inserted in the furnace was issued to be prepared as a composite hybrid material. HAp that acts as a matrix is described in fig. 1.

a b c

Figure 1: Process of calcination on milkfish bone of Hap: a) milkfish bone after washing and drying b) calcined milkfish bone at an excess temperature of 700°C c) calcined milkfish bone at a temperature range of 500-600°C

The calcination process at a temperature of 600°C produces 2 different colors of hydroxyapatite. Based on fig. 1 the calcination process at an excess temperature will produce black color, because of organic compounds in the form of carbon found in fish bones. Organic components in fish bones include 30% of the material while 70% are calcium phosphate and HAp [1]. HAp in black has an irregular crystal structure (amorphous), whereas in the process of calcining milkfish bone ranges between 500-700°C hours have formed pure hydroxyapatite, this can be shown in white where carbon compounds and collagen have decomposed during the calcination process and in appropriate temperature, so that the process of degradation organic material did not occur again [5].

Materials used as supporting elements for hybrid compost materials. The manufactured of hybrid composite used 3 types of material, namely: HAp and SiC/Al_2O_3 ceramics as a matrix, reinforced by metal: Al and Ti and Mg/Cu acting as wetting, the use of experimental materials is presented in table 1. Experimental combination of composite hybrid materials have their respective role. Each element has a role in the manufacture of milkfish bone-based hybrid composites. Element Al as a reinforcement is useful to expand the area of ductility from static loading, to withstand plastic deformation. Mg as wetting has an effect on the bonding between metal and HAp. The main properties of Titanium are: strong at high temperatures, light-weight specific, corrosion resistant, and has the ability to withstand extreme temperatures. It was easily integrated with other metals, to produce high-performance alloys. Ti alloys on HAp that act as an oxidation protector is also able to bind the powder granular bonds as well, whereas Cu in HAP alloys acts as a catalyst in the bonding process - with other metal elements such as Mg and Al. Cu and Mg plays a role on wetting will work optimally.

Table 1. Specifications of constituent elements of hybrid composite

Materials	[Gram]	[Mesh]
Hydroxiapatite (HAp) *	500	200
Aluminum**	300	280
Copper (Cu)***	100	200
Titanium (Ti)**	200	320
Magnesium (Mg)***	250	180
Silicon Carbide (SiC)*	100	200
Alumina (Al_2O_3)*	100	200

*Matrix,
**Reinforcement,
***Coupling Agent

Application of the self-propagating high temperature synthesis (SHS) method. The experiment used the SHS method, as shown in fig. 2 HAp powder as matrix added of 99.9% pure Al as reinforced, as well as 99.9% magnesium mixed into the mold as a wetting agent. Mg as a wetting agent has an important influence in the bond between powders, so that a bond reaction occurs between each powder. The addition of Ti and Cu powder as an additional reinforcement to form hybrid composite was carried out using the SHS method. To add a strengthening effect, the HAp powder which acts as a matrix is added to each SiC and Al_2O_3 to be compared as well. The used of temperature ranges from 700-800°C using a muffle furnace. Making samples for the SHS process, namely by mixing using a shaker mill for 10 minutes, the goal is that all ingredients can be mixed evenly.

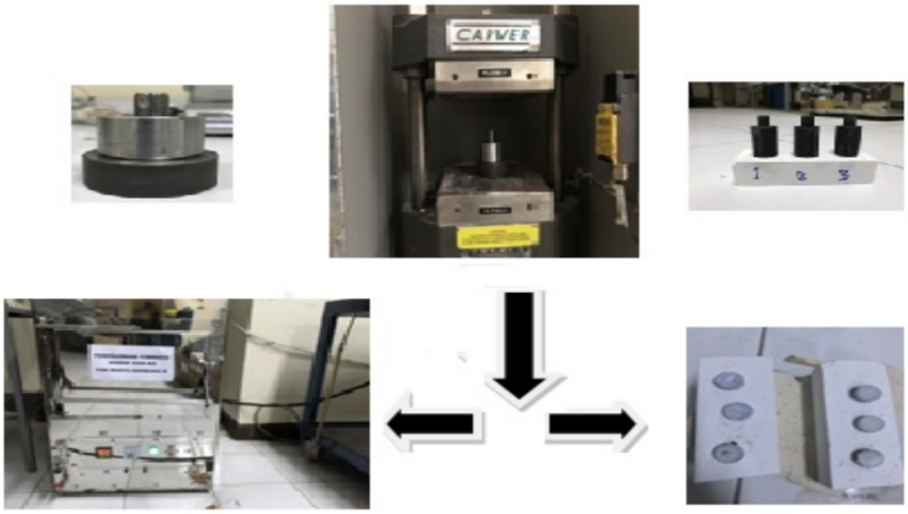
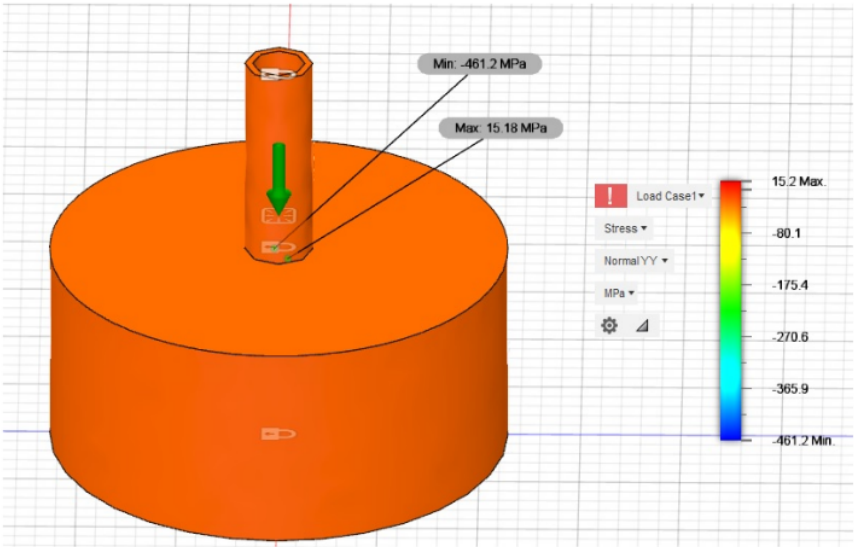


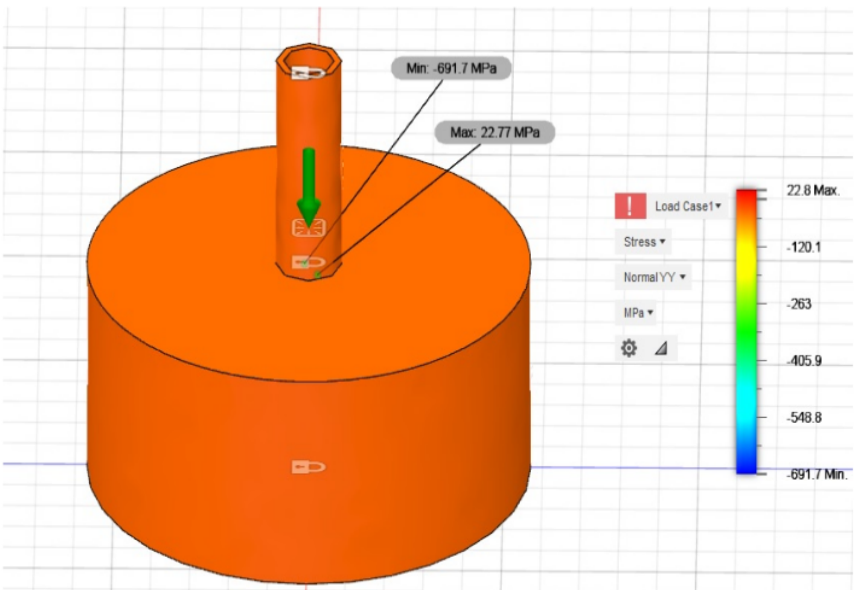
Figure 2: The SHS device consisting of mold/dies, compacting machine and muffle furnace accompanied by the results of a green hybrid composite samples

Mechanical desktop simulation to get stress distribution. The pressing process has an effect on microstructure and mechanical properties of hybrid composite materials. Software of Autodesk fusion 360 type, has been used for conducting pressure effect through distribution effect pressing [6]. Based on theoretical of journals [6-7], the flow stress is evenly distributed from the point of pressure to the end point. So that cracks will not arise [8]. This is proved by the absence of cracks on hybrid composite sample both from the composition without the matrix, and also the addition of SiC/Al_2O_3 as additional matrix. This has been done on experiments with Al_2O_3-reinforced Al-based composites. The compression provides an effect on mechanical properties especially on hardness value which are applied to hybrid composites. The initial pressure of a hybrid composite material applies a magnitude of 200 MPa, but after the pressing process runs, the pressure value that occurs has a difference.

The compression was applied to three different types of compositions. In hybrid composite materials: 80% HAp; 10% Al; 4% Mg; 3% Ti and 3% Cu results from the flow pressure that occurs, as seen in the simulation analysis of fig. 3. a. The moving distribution of flow stress from the value of 15.2: 80.1: 175.4: 270.6: 365.9 MPa to the maxima at 461.2 MPa, in this condition the flow stress works evenly and produces adequate mechanical properties between 167.07-229.21 HV10. The moving pressure follows the flow of forced, if there is no stress concentration, the resulting of pressure will be evenly distributed. In materials with a composition of 80% HAp. 10% Al; 4% Mg; 2% Ti; 2% Cu; 2% accompanied by additional Al_2O_3 ceramics, the pressure of 691.7 MPa, presented in figure 3. b, moving distribution of flow stress from the value of 22.8: 120.1: 263: 405.9: 584.8 MPa to the maxima at 691.7 MPa, in this condition the flow stress works evenly and produces adequate mechanical properties between 359.04-239.86 HV1. Al_2O_3 gives the effect of internal stress on the movement of the composite material powder, it is proven that the value of the resulting pressure was higher, this will improve the mechanical properties. Flow stress results with the addition of composition on 80% HAp; 10% Al; 4% Mg; 2% Ti; 2% Cu; 2% SiC, has the highest-pressure distribution value of 921.8 MPa, figure 3. c). flow stress distribution has a value that varies with results: 29.3: 160.9: 351.2: 341.4: 731.6 MPa and 921.8 MPa on last distribution stress. Compared with Al_2O_3, the role of SiC has more flow stress which is able to give a large tension for distribution effect. This proves that the surface contact pressure of SiC is greater than Al_2O_3


Min: -461.2 MPa
Max: 15.18 MPa
Load Case1
Stress
Normal YY
MPa
15.2 Max.
-80.1
-175.4
-270.6
-365.9
-461.2 Min.

(a)


Min: -691.7 MPa
Max: 22.77 MPa
Load Case1
Stress
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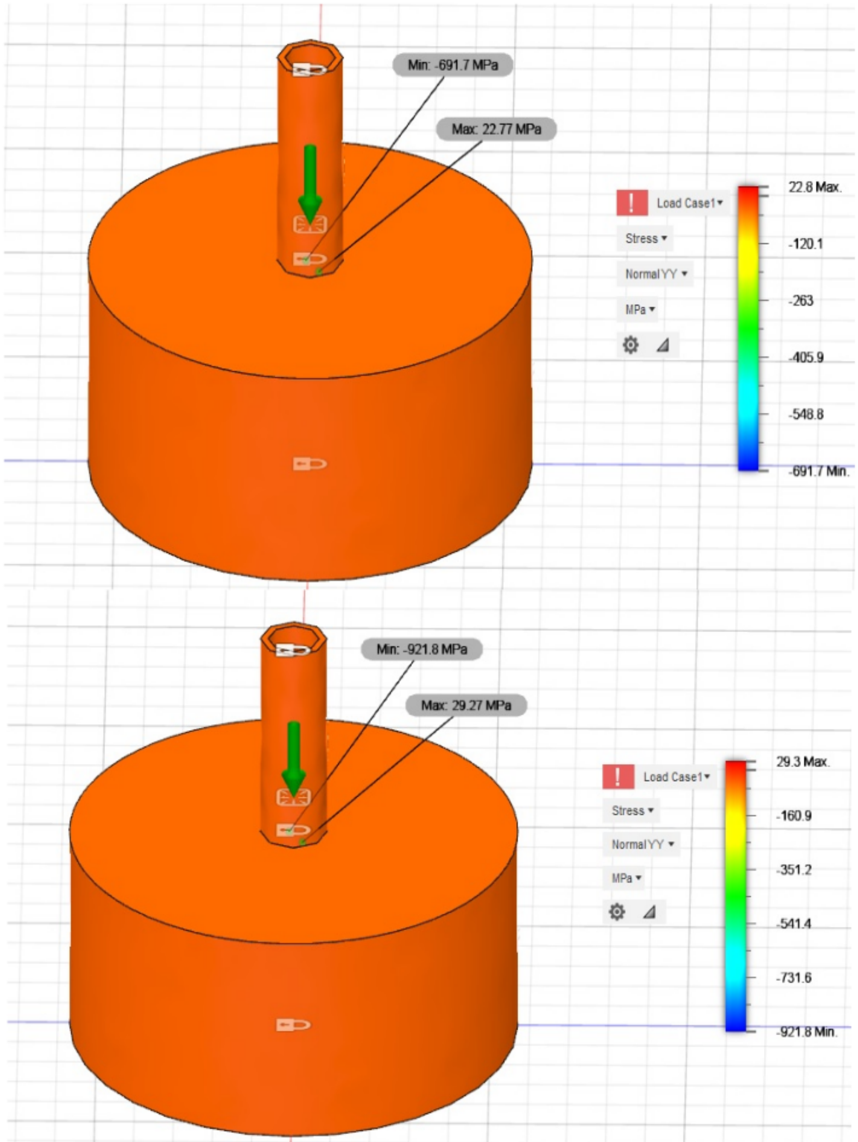
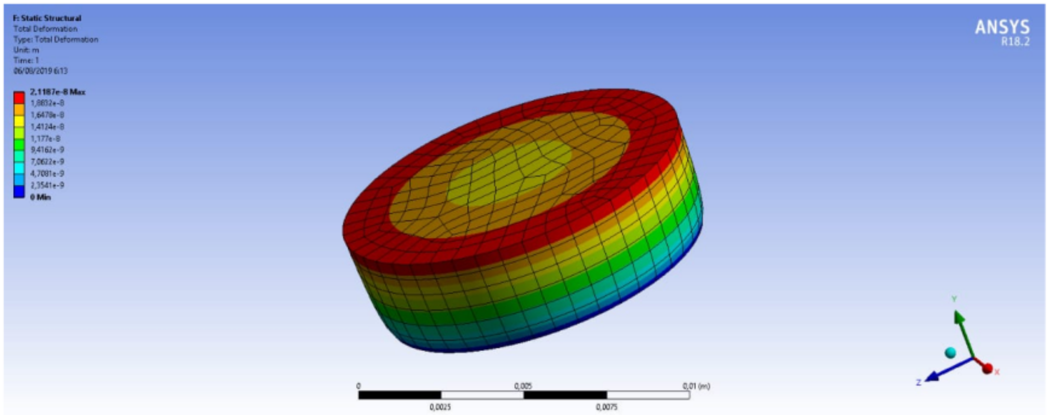
MPa
22.8 Max.
-120.1
-263
-405.9
-548.8
-691.7 Min.

(b)


Min: -691.7 MPa
Max: 22.77 MPa
Load Case1
Stress
Normal YY
MPa
22.8 Max.
-120.1
-263
-405.9
-548.8
-691.7 Min.
Min: -921.8 MPa
Max: 29.27 MPa
Load Case1
Stress
Normal YY
MPa
29.3 Max.
-160.9
-351.2
-541.4
-731.6
-921.8 Min.

(c)



(d)

Figure 3: Result of Autodesk fusion 360 simulation a). hybrid composite 1: 80% HAp; 10% Al; 4% Mg; 3% Ti; 3% Cu. b). Hybrid composite 2: 80% HAp; 10% Al; 4% Mg; 2% Ti; 2% Cu; 2% Al_2O_3. c). Hybrid Composite 3: 80% HAp; 10% Al; 4% Mg; 2% Ti; 2% Cu; 2% SiC. d) Validation of flow stress used ANSYS 18.2 software

Validate results software of measurements, used finite element type ANSYS R18.2, seen in fig. 3 d, there were differences in color that indicate the value of different deformation movements. Deformation flow starts from the top, in the picture in red, with a deformation of 2.1187e-8 m, continues to stage 2 in yellow, with values between: 1.6478e-8 to 1.177e-8. In the next stage, green was an area of equilibrium in green, valued at 9.6412e-9 and ending in grey at 2.3541e-9 m, in blue, the flow voltage stops, indicated by the value 0. This gives a sign that the deformation has stopped. This estimated total deformation provides information on the distribution of operational stresses that the voltage moves from top to bottom with the results transmitted to some central points of the deformation.

Characterization of Mechanical Properties.

Hardness measurements give information about homogeneity of strength [9]. It was determined by how much grain size can be produced uniformly, so that homogeneous strength can provide resistance to high loading [10]. The results of the measurement of hardness obtained several varied values. The hardness of Hybrid composite 1 with no-adding ceramics, resulted roughness was relatively low ranging between: 167.07-239.86 HV10. This is due to the effect of a relatively low compression effect that only has a value of 461.2 MPa compared to composites with the addition of ceramics as a matrix. Besides the HAp which is still fragile, pressure distribution that occurs is relatively small in the compression area, only 80.1-365.9 MPa based on image simulation 3.a, another factor because of reinforcement load of Al/Ti metal Cu and Mg is so complex, so HAp requires the addition of a second matrix as doping. Addition of Al_2O_3 in hybrid composite 2, hardness was 229.21-349.21 HV10. The hardness tends to increase; this is because the value of the compression absorbed is quite even. From the loading of 691.7 MPa, absorbed between 120.1-584.8 MPa, this will result in increased mechanical properties, due to the deformation resistance of the Al_2O_3 ceramic element which has medium hardness. The highest hardness produced by addition of SiC on composite hybrids 3: 359.04-375.01 HV10. The addition of SiC ceramic elements has an effect on the mechanical properties, especially hybrid composite toughness of the mixture of milkfish HAp on Al/Ti/Cu and Mg metals. This is caused by SiC elements that more closely blend alloy structures between ceramic metals and HAp, while Al_2O_3 is not sufficiently effective in forming densities in hybrid composites. Without addition of ceramics there were several micro-pores, indicated agglomerates on the metal with the addition of HAp from milkfish bone, this is shown in Figure 4. Results of hybrid composite microstructure made from HAp from milkfish bone alloy as matrix, metal as reinforcing and coupling agent and Al_2O_3 ceramic and SiC as reinforcement. Judging from the aspect of stress distribution, based on the simulation in fig. 3.c, amount of pressure that occurs is: 921.8 MPa. This results in a relatively large flow stress of 160.9-731.6. With the magnitude of the value, the compaction of the hybrid composite powder which is combined becomes increasingly compressible. So that the hardness increased significantly.

Table 2. Hardness and grain size of hybrid composite based CCF.

	Times	Hardness (VH10)	Grain Size (µm)
Hybrid composite 1*	(1 h - sinter)	167.07	2.66
Hybrid composite 2**	(1 h - sinter)	229.21	2.66
Hybrid composite 3***	(1 h - sinter)	359.04	1.28
Hybrid composite 1*	(2 h - sinter)	239.86	2.66
Hybrid composite 2**	(2 h - sinter)	349.21	2.65
Hybrid composite 3***	(2 h - sinter)	375.01	2.66

*Hybrid composites 1: 80% HAp; 10% Al; 4% Mg; 3% Ti and 3% Cu
**Hybrid composites 2: 80% HAp. 10% Al; 4% Mg; 2% Ti; 2% Cu; 2% + Al_2O_3
**Hybrid composites 3: 80% HAp; 10% Al; 4% Mg; 2% Ti; 2% Cu; 2% + SiC

Microstructures Analysis

The results of hybrid composites without addition of ceramics produced granules with size of 2.66 µm, the microstructure on fig. 4. a) For 1-hour heating, visible black HAp blobs. Reaching 2 hours heating shown fig. 4. b) Black HAp lumps shifted into the granular boundaries of metal. The same pattern also applies to the addition of Al_2O_3, at 1-hour heating figure 4. c), the HAp clumping is on each side of the field, after heating 2-hours the spread of HAp with Al_2O_3 shifts to the metal grain boundaries. The light on the optical microscope corresponds to the reflection of metal properties. The addition of SiC on Figure 4. e) With a heating time on 1-hour of SiC particles was seen to form symmetrical lines on metal grains while the HAp is on its sides. The addition of heating time reaches 2 hours fig 4. f) Then HAP and SiC were evenly distributed among the constituent elements. The grain size gives effects on mechanical properties especially to the particular hardness. In the grain structure produced by SiC composite, heating time of 1-hour produces the finest grain of 1.28 µm, but along by addition of a heating time on up to 2-hours then the item enlarged to reach 2.66 µm. On the addition of SiC with 2 hours of heating time, the highest hardness of 375.01 HV10 was obtained, from the microstructure influenced by the spread of constituent elements from hybrid composites such as; Al/Ti/Mg and Cu metals. The distribution of metal elements is able to cover the pores formed from HAp as a matrix with SiC which acts as an additional matrix so that the material structure becomes denser. The addition of SiC plays a better role than Al_2O_3, because the properties of SiC are better able to give even distribution than Al_2O_3. For hybrid composites that do not use ceramics as an additional matrix, there is still a hole that acts as a micro pore or can cause an indication of cracks in the samples.

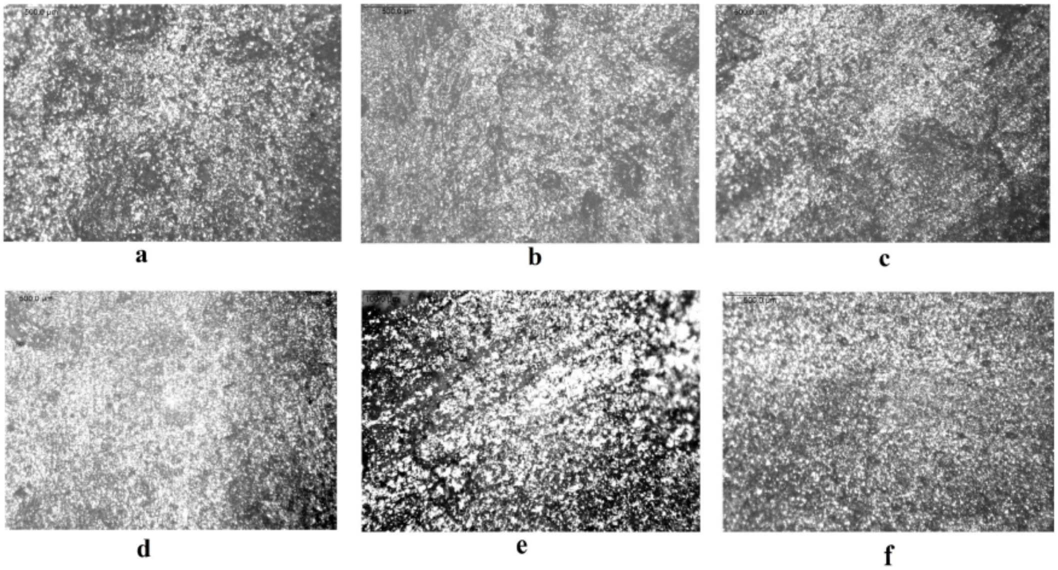


Figure 4: Micro-structures of hybrid composite based on HAp: a). HAp/metals no ceramic by 1-hour sinter: b). HAp/metals no ceramic by 2-hour sinter: c). HAp/metals add Al_2O_3 by 1-hour sinter: d). HAp/metals add Al_2O_3 by 2-hour sinter: e). HAp/metals add SiC by 1-hour sinter: f). HAp/metals add SiC by 2-hour sinter.

Summary

The first of Hybrid Composite experiment, consist composition of: 80% HAp; 10% Al; 4% Mg; 3% Ti and 3% Cu were pressured by the flow of stress, as seen in the simulation analysis. The distribution moves from the flow stress of the value 15.2: 80.1: 175.4: 270.6: 365.9 to the maximum at 461.2 MPa, in this condition the flow stress works evenly and produces the maximum mechanical hardness: 229.21 HV10.

Composition of hybrid composites was; 80% HAp. 10% Al; 4% Mg; 2% Ti; 2% Cu; 2% of accompanied by additional Al_2O_3 ceramics. The operating pressured reaches 691.7 MPa. Distribution of stress from the value of 22.8: 120.1: 263: 405.9: 584.8 in this condition the flow stress works evenly and produces mechanical hardness properties, namely: 359.04.

Pressure effect by addition of SiC to the composition of 80% HAp; 10% Al; 4% Mg; 2% Ti; 2% Cu; 2% has the highest distribution pressure of 921.8 MPa. The flow stress distribution has varying values with results: 29.3: 160.9: 351.2: 341.4: 731.6 When compared to Al_2O_3, the role of SiC has more flow stresses that can provide large stresses for a more even distribution effect.

Acknowledgments

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References

[1] Pramono A, Sulaiman F, Suryana and Milandia A. Calcination process on chanos chanos forsk (CCF) of milkfish bones to get Hydroxyapatite (HAp) as Composites Application. IOP Conf. Series: Materials Science and Engineering. Vol 532, 1 (2019): 1-9.

[2] Pramono A, Kollo L, Kallip K, Veinthal, R, Gomon JK 2014 Heat treatment of ultrafine grained high-strength aluminum alloy Key-Engineering Materials vol 604 pp 273-276:

[3] Pramono A: Investigation of Severe Plastic Deformation Processes for Aluminum Based Composites. Ph.D. Dissertation. Department Mechanical Engineering. Tallinn University of Technology, Tallinn-Estonia. (2016)

[4] Pramono A, Kommel L, Kollo L and Veinthal R. The Aluminum Based Composite Produced by Self Propagating High Temperature Synthesis. Materials Science (MEDŽIAGOTYRA) vol. 22, 1, (2016): 40-43

[5] Pramono A, Sulaiman F, Suryana and Milandia A. The Influence of Temperature on the Hybrid Composite Manufacturing Process Made from Milkfish. International Conference on Food Security Innovation (ICFSI), "Emerging a Sustainable Agriculture, Maritime and Local Resources Empowerment for Global Food Innovation". 23th October 2018. University of Sultan Ageng Tirtayasa

[6] Pramono A, Kommel L, Kollo L and Veinthal R. Hot and cold of pressing effect on ECAP parallel channel composite based on Al/ANF material, Advanced Materials Research vol. 1123 (2015): 343–347

[7] Pramono A. Dhoska K. Markja I and Kommel L. Impact pressure on mechanical properties of aluminum-based composite by ECAP-parallel channel. Pollack Periodica. vol 14, 1 (2019): 67-74.

[8] Comaneci R. Zaharia L and Chelariu R. Damaging Prediction of Difficult-to-Work Aluminum Alloys during Equal Channel Angular Pressing Journal of Materials Engineering and Performance vol 21, 3 (2012): 287-297.

[9] Pramono A, Kollo L, Kallip, Veinthal R and Gomon JK. Heat Treatment of Ultrafine Grained AA6061 Consolidation by Equal Channel Angular Pressing. Journal Applied Mechanics and Materials. vol 771 (2015): 252-256.

[10] Pramono A, Milandia A, Nugraha K and Fawaid M. Aluminum aloys by ECAP Consolidation for Industria Application. VANOS Journal of Mechanical Engineering Education. vol 2, 2 (2017): 117-126.

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