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## Hydroxyapatite (HAp) From Tenggiri Fish Bones As Abrasive Material In Toothpaste Formula

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### Abstract

Cavities are one of the factors of dental and oral health problems that can be prevented by brushing teeth using toothpaste. Toothpaste can be made from a variety of chemicals, one of which is hydroxyapatite (HAp) which has good biocompatible properties. Hydroxyapatite can be obtained by utilizing tenggiri fish bone waste which has the main element of calcium. Fish bones soaked with NaOH and acetone are then calcined at 800 °C for 3 hours to obtain CaO powder and characterized its elemental content using X-Ray Fluorescence (XRF). CaO obtained was then reacted with (NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub> with a mole ratio of Ca/P 1.67 then heated at 90 °C, added NaOH up to pH 12, then the obtained deposits are filtered and calcined at 900 °C. The solids obtained from the calcination are then characterized using X-Ray Diffraction (XRD) and Scanning Electron Microscope (SEM). Hydroxyapatite (HAp) was formulated into toothpaste with the concentrations of 0% (F0), 45% (F1), 50% (F2), and 55% (F3). Toothpaste was evaluated using organoleptic tests, homogeneity tests, foam height tests, spreadability tests, pH and hedonic tests. XRD analysis shows that the resulting hydroxyapatite (HAp) has a crystal structure in accordance with ICSD standard No. 96-900-3549. SEM analysis showed that granular particles measuring 0.1 μm–0.3 μm in size. All formulated toothpastes (F0, F1, F2, and F3) meet the requirements of a good toothpaste. Hydroxyapatite (HAp) can be formulated into a good toothpaste with a concentration of 45%.

**Keywords:** Tenggiri fish bones, Hydroxyapatite (HAp), toothpaste

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### 1. INTRODUCTION

Dental and oral health is part of the overall health of the body and is inseparable from the health of the body in general. Teeth and mouth problems can affect the quality of life, including speech function, pain reduced confidence, and interfere with daily activities (Ahmad, 2017). Cavities are one of the factors that become dental and oral health problems. Cavities are caused by the erosion of dental emails, which can occurred due to many things, such as consuming sweet foods, microbial activity in the mouth, and also saliva activity (Wadu *et al.*, 2015). To prevent dental and oral health problems, brushing the teeth is one of the solution. Teeth brushing using toothpaste is recommended twice a day, i.e.

after meals and before going to bed. The use of toothpaste is chosen because it is more practical and can cover micro-sized holes in the teeth as well as reduce the potential for other holes to form (Wadu *et al.*, 2015).

Toothpaste contains a wide range of chemical compounds, one of which contains hydroxyapatite (HAp) which is effective for closing holes in the teeth (Wadu *et al.*, 2015). Hydroxyapatite (HAp) with the chemical formula Ca<sub>10</sub>(PO<sub>4</sub>)<sub>6</sub>(OH)<sub>2</sub> is often applied to the field of bones and teeth because it has good biocompatible properties in human tissues and its composition is almost the same as bone (Riyanto & Maddu, 2014).

One of the efforts to synthesis hydroxyapatite (HAp) is to utilize the waste of

tenggiri fish bone because many household industries in Jambi produce pempek, tekwan, and crackers that only use the meat of tenggiri. Fish bones have a high calcium (5.63 g/kg) and phosphorus content (2.38 g/kg) (Stanek *et al.*, 2013). Where calcium and phosphorus are the main elements of hydroxyapatite formation (HAp) (Susanti *et al.*, 2011). Hydroxyapatite can be synthesized from chemicals and natural materials such as from mollusk shell (Wattanutchariya & Changkowchai, 2014), mackerel bones (Anggresani *et al.*, 2018), chicken bones (Rajesh *et al.*, 2012), limestone of Bukit Tui in Padangpanjang (Anggresani, 2016), cockle shells (Azis *et al.*, 2015), tuna bones (Hanura *et al.*, 2017), and cow bones (Haris *et al.*, 2016).

The hydroxyapatite has been used in the pharmacy fields such as tablet making (Mustaqim, 2017), dental implant (Sidiqa *et al.*, 2012), injection manufacture (Budiatin *et al.*, 2016). However, there was not an investigation on making toothpaste from hydroxyapatite tenggiri fish bone, hence this study is conducted to investigate hydroxyapatite toothpaste that can cover the holes in the teeth.

## 2. MATERIALS AND METHODS

### The formation of Calcium Oxide (CaO)

8 kg Tenggiri fish bone waste (*Scomberomeres guttatus*) was cleaned and boiled for 45 minutes then rinsed. The bones were soaked with 0.1% NaOH 10 L for 7 hours and 50% acetone for 8 hours. Then, the bones are drained and dried for 7 days then crushed. The bones were calcined at 800 °C for 3 hours using a furnace until it formed the powder. The powder was crushed and sieved with mesh 80. The bones powder of mackerel (CaO) was then analyzed by XRF PAnalytical Epsilon 3.

### Hydroxyapatite Synthesis

337.0328 gram of CaO powder was diluted with 2 L aquadest. The solution was stirred for 30 minutes at 300 rpm so that the suspension of Ca(OH)<sub>2</sub> was formed. (NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub> was used as phosphate precursors. The suspension of Ca(OH)<sub>2</sub> mixed with (NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub> was generated with a 1.67 mole ratio of Ca/P and heated at 90 °C for 1 hour. NaOH 1 M added to contour the pH until it reaches 12. Let the mixture stands for 24 hours at room temperature. Then the formed deposit was filtered and dried using oven at 120 °C for 5 hours. The dry precipitate was calcined at 900 °C for 5 hours (Haris *et al.*, 2016). Precipitation process is an alkaline acid reaction that produces crystalline solids as well as water. This process requires only cheap raw materials, relatively simple chemical reactions as well as particle size and homogeneity that tends to be quite good (Haris *et al.*, 2016).

### Toothpaste Preparation

The synthesis of hydroxyapatite (HAp) toothpaste from mackerel bone begins with 1 part of Na-CMC expanded with hot aquadest in beaker glass and let remain for 15 minutes continue by stirred firmly. Hydroxyapatite (HAp) was crushed in mortar according to formula (F0 (CaCO<sub>3</sub> 55 parts), F1 (HAp 45 parts), F2 (HAp 50 parts), F3 (HAp 55 parts)), then added 0.1 part of titanium dioxide, 0.1 part of methyl paraben, and 0.2 parts of saccharine. The dispersion of Na CMC that has dissolved was crushed vigorously until homogeneous, then added 2 parts of SLS and 30 parts of glycerine. *Oleum menthae piperitae* dripped into the paste and crushed until homogeneous. After that, the paste preparation was inserted into the tube and evaluated.

**Table 1.** The formulation of toothpaste

Ingredients	Composition (%)			
	F0	F1	F2	F3
Hydroxyapatite	-	45	50	55
Calcium Carbonate	55	-	-	-
Na-CMC	1	1	1	1
Glyserin	30	30	30	30
Methyl Paraben	0.1	0.1	0.1	0.1
Sakaryn	0.2	0.2	0.2	0.2
Sodium lauryl sulfat	2	2	2	2
Titanium dioxide	0.1	0.1	0.1	0.1
<i>Oleum menthae Piperitae</i>	qs	qs	qs	qs
Aquadest	add 100	add 100	add 100	add 100

### Toothpaste Evaluation

Toothpaste preparations that have been made should be evaluated. Toothpaste evaluations include:

#### Organoleptic Test

Organoleptic observations of toothpaste include shape (consistency), color, and aroma that were objectively observed (Syurgana *et al.*, 2017).

#### pH Test

pH measurement was done using pH meter, which calibrated by using buffer phosphate solution with pH 10, buffer phosphate pH 7.0 and buffer phosphate pH 4.0 by dipping pH electrodes and temperature stick into the dapar solution. Once calibrated, the pH measurement of the aquadest is ready to use.. Weigh 1 gram of hydroxyapatite toothpaste (HAp) from the bones of the mackerel and diluted with 10 ml of aquadest. Dip the pH electrodes and temperature sticks into the prepared paste that has been diluted until the needle monitor shows a stable number and read the pH on the monitor (Ali *et al.*, 2015).

#### Homogeneity Test

Homogeneity test was done by means of toothpaste to be tested weighed as much as 0.1 grams and then placed between two layers of glass objects to be observed homogeneity. If there are no coarse grains on the glass of the object, then the toothpaste was declared homogeneous, while the presence of coarse grains indicates that the toothpaste was not homogeneous (Syurgana *et al.*, 2017).

#### Scatter Power Test

Scatter power test was carried out by placing  $a \pm 1$  gram of toothpaste preparation between two transparent plastics and then given a load from the smallest size to the largest size, then measured the size of the spread diameter formed at each load increase (2 g, 4 g, and 6 g) (Ismail *et al.*, 2014).

#### Foam Formation Test

Foam formation test was carried out by making a solution of 1% of the various concentrations of hydroxyapatite (HAp) toothpaste from the bones of mackerel in water. In a measuring glass, the solutions poured and covered with aluminium foil. The foam shaken and the foam height was

measured at 0 and 5 minutes (Syurgana *et al.*, 2017).

#### Stability Test

Stability test was carried out by storing hydroxyapatite (HAp) toothpaste from mackerel bones at different temperatures at 4 °C, room temperature, and 40 °C for 21 days. Then it keeps observed seen whether organoleptic, pH, homogeneity, scatter power and foam changes formed on the 7<sup>th</sup> day, 14<sup>th</sup> day and 21<sup>st</sup> day (Nagajyothi *et al.*, 2014).

#### Hedonic Test

Hedonic test was carried out to determine the respondent's level of preference for hydroxyapatite toothpaste (HAp) preparations from the bones of the mackerel made. Respondents as many as 20 people with the following criteria: male or female sex, age 17-25 years. Aspects assessed include parameters of color, aroma, taste, and texture The product then being assessed by the respondent to observe the response whether the respondents like the product. (Dian *et al.*, 2014).

## 3. RESULTS AND DISCUSSION

### The Formation of Calcium Oxide (CaO)

The formation of mackerel bone powder (CaO) begins with the boiling process for 45 minutes, which will facilitate the cleansing of the bones from the remaining meat that was still attached, after which the fish bones were soaked with NaOH 0.1% for 7 hours then soaked with acetone 50% for 8 hours to reduce the fat levels contained in the bones of mackerel. The calcination of the sample done by using a furnace with a temperature of 800 °C for 3 hours to remove carbonate that can inhibit crystal formation and eliminate all organic elements contained in the bones of mackerel. (Royani *et al.*, 2016) mentioned the decomposition of  $\text{CaCO}_3$  into CaO ideally occurs at a temperature of 800 °C. At this stage, there was a decomposition reaction of calcium carbonate ( $\text{CaCO}_3$ ) to calcium oxide (CaO) Reactions that occurred according to eq. 1 :



XRF analysis is conducted to determine the chemical composition in the bones of tenggiri (Manggara, 2018). Table 2 showed

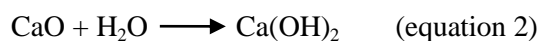
that calcium oxide contained in the bones of mackerel fish by 49.846% therefore it can be used as a precursor of calcium in the formation of hydroxyapatite compounds. The minimum condition of calcium concentration as an abrasive in making toothpaste was 35% (Judge & Patil, 2008). (Mutmainnah *et al.*, 2017) investigated CaO in Tuna Bones which contained 62.31% CaO content and (Anggresani *et al.*, 2018) investigated CaO in Tenggiri Bones which contained 50.814% CaO content. The results obtained are smaller compared to previous studies since the sample was still black after calcination which indicated that there was a process of degradation of organic material that occurred (Mutmainnah *et al.*, 2017).

**Table 2.** XRF analysis result of Tenggiri Bone

Oxide Compound	Concentration (%)
CaO	49.846
P <sub>2</sub> O <sub>5</sub>	26.195
MgO	2.074
Al <sub>2</sub> O <sub>3</sub>	0.712
SiO <sub>2</sub>	0.234

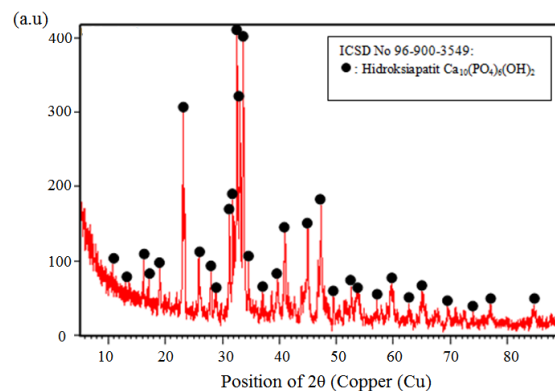
### Hydroxyapatite Synthesis (HAp)

In this study, the hydroxyapatite was made using precipitation methods. CaO powder which is the source of calcium in the fabrication of hydroxyapatite (HAp) was dissolved with water to get calcium hydroxide (Ca(OH)<sub>2</sub>). Reactions that occurred according to eq. 2 :



To synthesize hydroxyapatite (HAp), a phosphate precursor was needed, where in this study the precursor used was diammonium hydrogen phosphate (NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub> with 1.67 mole ratio of Ca/P. Hydroxyapatite formation (HAp) can occurred at pH 8-12 in order to obtained high purity and stability (Tua *et al.*, 2016). In this study set, the basic condition of pH 12 was controlled using NaOH 1 M. Samples was calcined at a temperature of 900 °C for 5 hours aimed to increasing the degree of crystallization (Haris *et al.*, 2016), eliminate

carbonate which can maximized the formation process of hydroxyapatite (HAp) (Ningsih *et al.*, 2014). Heating at 900 °C indicated that the samples used from the bones of the mackerel have a good thermal stability characterized by the formation of hydroxyapatite (HAp) (Royani *et al.*, 2016). The reaction was in eq. 3.

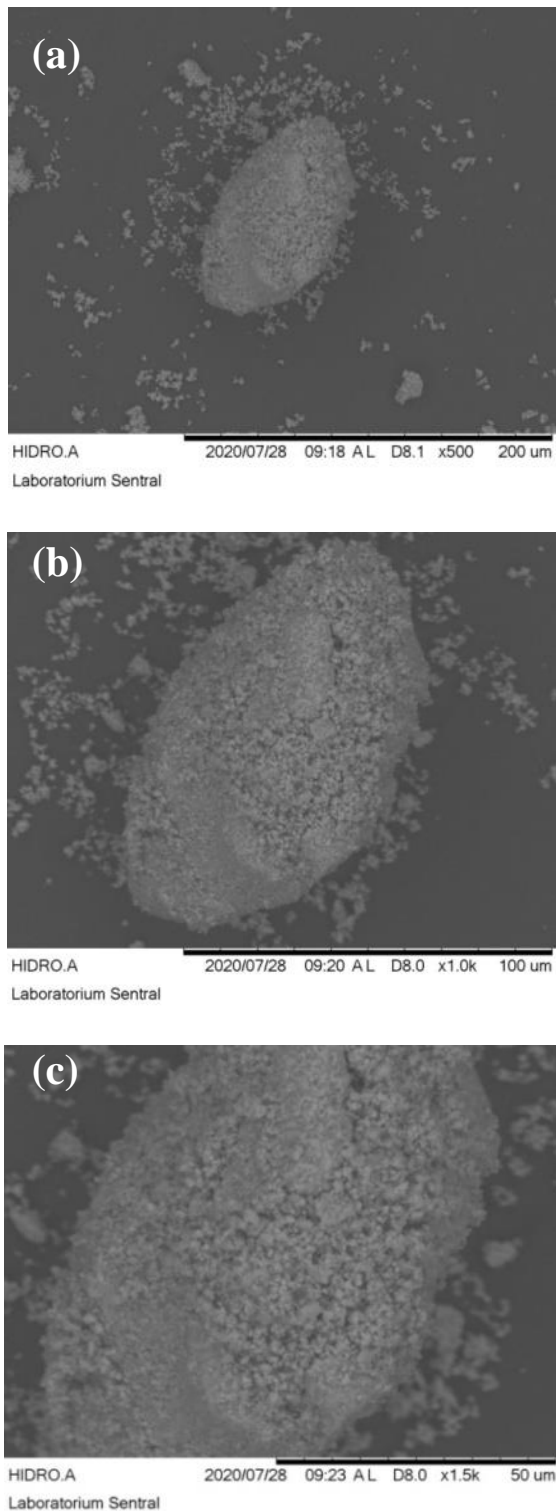


**Figure 1.** XRD analysis of hydroxyapatite

Fig. 1 shows the XRD pattern of hydroxyapatite compound of tenggiri bones. XRD analysis aims to determine the structure, orientation, and size of crystals (Munasir *et al.*, 2012). XRD indicates the presence of hydroxyapatite (HAp) formed from calcium precursors derived from decomposition of mackerel bones and phosphate precursors derived from diammonium hydrogen phosphate. XRD X'PERT POWDER PW 30/40 analysis shows hydroxyapatite in accordance with ICSD standard 96-900-3549 which shows hydroxyapatite with chemical formula Ca<sub>10</sub>(PO<sub>4</sub>)<sub>6</sub>(OH)<sub>2</sub> and crystal form in the form of hexagonal. Sharp peaks with high intensity can be seen at 2θ = 32.58° and other peaks with lower intensity at 2θ = 33.71°; 33.14°; and 23.13°. In previous research, (Anggresani *et al.*, 2018) shows hydroxyapatite with ICSD standard 01-074-9780 where the high intensity was located at 2θ = 32.84°. Hydroxyapatite has many standards, this can be seen from some previous research on hydroxyapatite. However, a high intensity 2θ peak will be obtained at 32°.







**Figure 2.** SEM images of obtained HAp with magnification (a) 500x ; (b) 1000x ; (c) 1500x

Fig. 2 shows the SEM image of hydroxyapatite compound of tenggiri fish bone with the magnification of 500, 1000, and 1500 times which consisted of agglomerates. Based on (Hui *et al.*, 2010) finding, the SEM images were the agglomerates with almost the same

shapes. Previous investigation reported that the result of the surface morphology of hydroxyapatite in the form of agglomerates (Ichsan & Helwani, 2015).

### Toothpaste Evaluation

In toothpaste making, hydroxyapatite was used as an abrasive materials. Formula 0 (F0) used  $\text{CaCO}_3$  abrasive materials for about 55%, while F1, F2, and F3 used hydroxyapatite as an abrasive materials as much as 45%, 50%, and 55%, respectively. In the previous research, the addition of hydroxyapatite to toothpaste preparations was 15% (Sadiasa *et al.*, 2013).

Organoleptic observations show all formulas smell of mint. (Afni & Said, 2015) was investigated the smell of oleum menthol which can mask smell of other ingredients so that there were not difference in the aroma of hydroxyapatite toothpaste (HAp). The formula was white, because the ingredients used to formulate toothpaste were generally white. F2 was more solid than F1 and F3 was denser than F1 and F2, this was due to the concentration of hydroxyapatite powder (HAp) added more and more (Ningsih *et al.*, 2015).

The results of the scatter power test of each formula (F0, F1, F2, and F3) of hydroxyapatite toothpaste (HAp) showed the ability to spread preparations getting more significant as the loading increased, F1 had a large dispersal power followed by F2, F0, and F3 respectively. (Andriana *et al.*, 2011) studied the area of spread was directly proportional to the increase in loading addition, the greater the loading added the wider the scatter power generated. The viscosity of F0 and F3 was higher then followed by F2 then F1, respectively. Viscosity was inversely proportional to dispersability, the greater the viscosity, the smaller the scatter power (Numberi *et al.*, 2020).

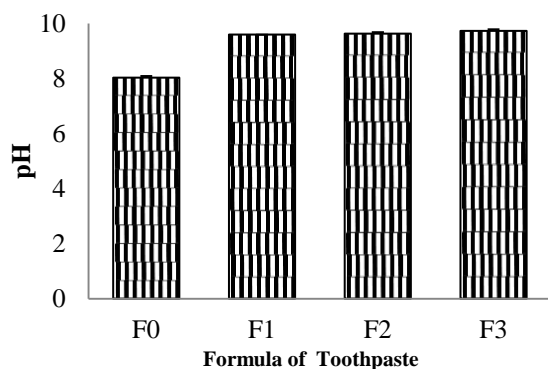
The stability test in 21 days showed F0 had discoloration and smell on the 14th day and the 21st day at each temperature. F1 experienced a change in the smell on the 14th day at 4°C and 40°C and on the 21st day at room temperature. The toothpaste formula F0, F1, F2, and F3 undergone homogeneity changes on the 21<sup>st</sup> day at 40°C. This can be caused by extreme storage temperatures causing unstable preparation.

**Table 3.** Organoleptic test results and homogeneity of hydroxyapatite toothpaste (HAp)

Observations	Formula			
	F0	F1	F2	F3
Color	White	White	White	White
Shape	Semi solid	Semi solid	Semi solid	Semi solid
Odor	Mint	Mint	Mint	Mint
Homogeneity	Homogeneous	Homogeneous	Homogeneous	Homogeneous

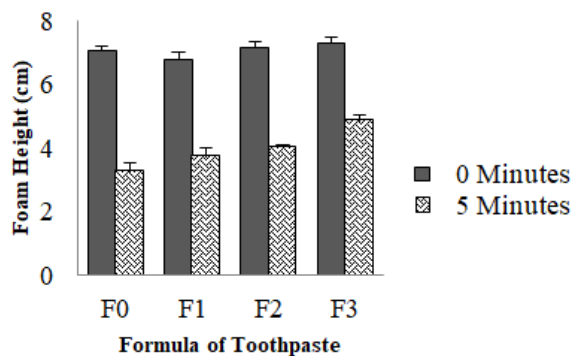
Description :

- F0 : Toothpaste formulation without hydroxyapatite
- F1 : Toothpaste formulation with 45% hydroxyapatite
- F2 : Toothpaste formulation with 50% hydroxyapatite
- F3 : Toothpaste formulation with 55% hydroxyapatite



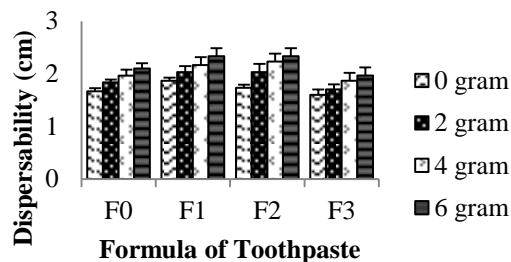
**Figure 3.** HAp toothpaste pH test result

Fig. 3 showed pH test results of each formula (F1, F2 and F3) of hydroxyapatite toothpaste (HAp) which have a range of 9.6-9.8. As for formula F0 has a range of pH 8-8.1. This happened due to the ingredients used in the formulation were alkaline, such as hydroxyapatite (HAp), calcium carbonate, Na-CMC, and sodium lauryl sulfate, yet the preparation of hydroxyapatite toothpaste (HAp) is still within the permissible pH with the range of 4.5-10.5 (Mahdali *et al.*, 2017). pH toothpaste that meets SNI (12-3524-1995) requirements is not expected to irritate the mucosa of the mouth.



**Figure 4.** HAp toothpaste from height test results

The results of foam height testing showed the ability of a detergent to produce foam. There was no high foam requirement for a toothpaste product. It is attributed to the aesthetic value that consumers like (Daud *et al.*, 2016). Based on high observations of foam in each formula (F0, F1, F2 and F3), they have almost the same height of foam. This was due to the sodium lauryl sulfate detergent agent being added in the same percentage. SLS was an anionic surfactant with characteristics as a good foam shaper and has high cleaning power (Syurgana *et al.*, 2017).



**Figure 5.** HAp toothpaste spread test result

After stored each formula (F1, F2 and F3) for 21 days, the preparation undergoes a change in pH in each temperature on a weekly date in the range of 9.1-9.7, while F0 in the range of 7.4-8. Changes in pH values in each formula are caused by environmental factors such as extreme temperature, humidity and storage containers (Afni & Said, 2015). It is suspected that there was a reaction between hydroxyapatite (HAp) and sodium that triggered the formation of alkaline, but the preparation of hydroxyapatite toothpaste (HAp) was still within the permissible pH

requirement range of 4.5-10.5 (Badan Standar Nasional (SNI), 1995).

The height of each formula of hydroxyapatite toothpaste (HAp) (F1, F2 and F3) after 21 days of storage experienced no significant difference, but F0 had a lower foam height on the 7th to 21st day of each storage temperature. However, hydroxyapatite toothpaste (HAp) preparations still qualified for foam ability that lasts after 5 minutes which is 60%-70% (Benediktus, 2017).

The increase in the number of hydroxyapatite (HAp) increases the diffusion power of the preparations in each formula. This happened because the more hydroxyapatite (HAp) that was added, the more dense the preparation (Ningsih *et al.*, 2015). Based on the results of spread tests showed that hydroxyapatite toothpaste (HAp) stored at 4 °C had a greater spread than stored at room temperature. It was because storage at 4 °C had higher humidity and low viscosity than storage at room temperature. Storage at 40 °C had a higher temperature than storage at room temperature thus reducing the amount of water found in toothpaste that can affected viscosity (Lupita & Kadiwijati, 2019). This happened because of the storage temperature and is thought to occurred because the storage container is made of glass so that at 4 °C it can absorb water and at 40 °C it can reduce water. Hydroxyapatite toothpaste preparations (HAp) should be stored at room temperature.

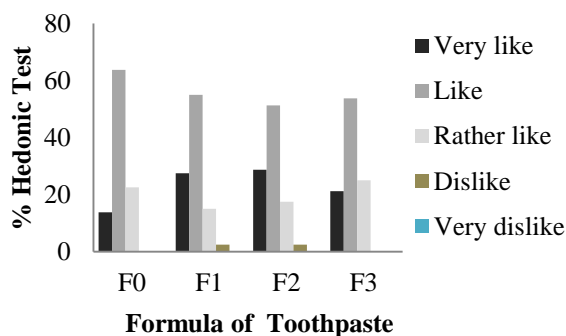


Figure 6. HAp Toothpaste Hedonic Test

Hedonic test of each hydroxyapatite toothpaste (HAp) preparation against 20 volunteers on average liked each formula of toothpaste both in terms of color, aroma, taste and texture, but there were 2 volunteers who did not like the texture of toothpaste from F1 and F2. Hedonic test observations can be

concluded that the most preferred percentage of color, aroma, taste and texture is formula F1.

#### 4. CONCLUSIONS

Hydroxyapatite which was synthesized from tenggiri fish bones can be formulated into toothpaste and the best formula was F1 with a concentration of 50% Hydroxyapatite (HAp).

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