

Liquid Smoke Production Quality from Raw Materials Variation and Different Pyrolysis Temperature

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Abstract— This research is intended to know liquid smoke chemical and physical characteristics quality acquired from pyrolysis from various raw materials with different temperature level. The making of liquid smoke is done through pyrolysis process with temperature level $100\pm 10^{\circ}\text{C}$; $200\pm 10^{\circ}\text{C}$; $300\pm 10^{\circ}\text{C}$; and $400\pm 10^{\circ}\text{C}$ for 5 hour. This research is done experimentally by using complete random design on factorial pattern 3×4 with 3 repetition. Factor A is raw materials type that consists of coconut fibre, coconut shell and cinnamon while factor B is temperature level. The observed parameter consists of liquid smoke physical characteristic that consists of equipment performance, density, rendement, degree of acidity and chemical characteristic that consists of water-content, titrated acid total, phenol-content, carbonyl-content and benzo(a)pyren-content. The result of research shows that there is a very actual interaction ($P < 0,05$) between using raw materials type with pyrolysis temperature level toward equipment performance, density, rendement, degree of acidity, water-content, titrated acid total, phenol-content, carbonyl-content and benzo(a)pyren-content. The best liquid smoke production quality can be found in raw materials cinnamon treatment on temperature level $400\pm 10^{\circ}\text{C}$, which is pyrolysis equipment performance 16,29 ml/hour.meter, density 1,017 g/ml, rendement 38,78%, pH 3,503, titrated acid total 0,72 %, phenol-content 0,57 %, carbonyl-content 4,13 %, benzo(a)pyren-content 0,04 ppm. Based on the result of this research, it can be concluded that using cinnamon with level pyrolysis temperature $400\pm 10^{\circ}\text{C}$ is better used rather than coconut fibre and coconut shell because of its lowest benzo(a)pyren-content.

Keywords— raw materials type; temperature; liquid smoke; physical characteristic; chemical characteristic.

I. INTRODUCTION

An easy and simple technology that can be used is needed in fulfilling the need of natural preservatives such as liquid smoke. That technology later can be used as an alternative to replace chemical preservatives that comes from environmental raw materials such as coconut fibre, coconut shell and cinnamon. Liquid smoke is acetic acid that is acquired through pyrolysis process from material that consists of cellulose, hemicellulose and lignin components [1].

Various raw materials have been used in the making of liquid smoke among others by using cashew nut shell and coconut fibre [2], coconut shell [3], organic waste [4], coconut shell and coconut fibre [5]. That raw materials consist of enough cellulose and lignin contents. That result of research shows relationship between raw materials type

and compound composition on liquid smoke product. The result of research is that teak wood dusts pyrolysis result consist of p-guaiakol, 2 methoxy 4 propenil phenol, 2 methoxy 4 methyl phenol, 3,4,5 trimethoxy toluene and 1,3 dimethoxy siringol which is the basic components of biofuel [6]. While dominant compound from miswak wood and acacia wood pyrolysis result consists of acetic acid and vanillin [7], and from organic waste is γ -butyrolactone and 2-hydroxy 3 methyl 2 cyclopentene-1-on [4].

The diversity of raw materials and liquid smoke production method result in complex chemical component with various structure, reactivity and sensory activity [8]. While on pyrolysis, cellulose will experience hydrolysis that produces glucose and further reaction produces acetic acid, water and a little bit of phenol [9]. Lignin in pyrolysis produces phenol compound and its derivatives and pyrolysis on high temperature will produces tar. While on pyrolysis,

hemicellulose will produces furfuran, furan and carboxylic acid. Various compounds with diverse compositions can be acquired from pyrolysis process, apart from that raw materials type along with pyrolysis operation condition is estimated to have influence on chemical and physical characteristics of liquid smoke. The purpose from this research is to know the quality of liquid smoke production from chemical and physical characteristics with pyrolysis on various raw materials combined with different pyrolysis temperature level.

II. MATERIAL AND METHOD

Material and Equipment. Raw material such coconut fibre waste, coconut shell come from Pasar Raya Padang and cinnamon with its bark condition taken in dry condition is acquired from cinnamon farmer in Tanah Datar. Chemical material that is used to analysis between 0,1 N NaOH, distilled water, phenolphthalein, Na_2CO_3 , Folin Ciocalteu this, DMSO, petri dish for nutrients. To analysis smoke component, it uses methanol, helium gas, Whatman No. 42 filter paper. Equipment that is used in pyrolysis consists of one set laboratory scale liquid smoke maker that consists of stainless steel kiln with 3 kg capacity, condenser unit 14 meter length, cooling pipe by using glass pipe, distillate bottle (container liquid smoke), gas stove, water pump, water drum, water hose for condenser cooler, ball valve, PVC pipe, pole, clamp, thermometer, stopwatch, measuring cup, 10 ml microliters syringe, luer key syringe with diameter 25 mM from 0,45 mm PVDF, lock syringe 10 ml vial B x 40 mm and GC MS. and other equipments that are used, such as 1 (one) aquarium set, weights, thermos bottle, patrialis cup, electric stove, filter paper, oven, distillation equipment, analytic scales, oven, porcelain bowl, desiccator, filter, pH meter, 125 ml Erlenmeyer and 500 ml glass, filter paper, Soxhlet, tube, centrifuge tube, micro buret, pipette, volumetric flask pipette 250 ml, centrifuge, spectrophotometer, pycnometer

This research uses factorial experiment 3 x 4 with three repetition in complete random design (RAL). The first factor consists of 3 (three) level of liquid smoke raw materials source like cinnamon, coconut shell, cinnamon, and the second factor consists of 4 (four) level pyrolysis temperature $100 \pm 10^\circ\text{C}$; $200 \pm 10^\circ\text{C}$; $300 \pm 10^\circ\text{C}$; from $400 \pm 10^\circ\text{C}$ for 5 hour. The data is analyzed by using variant analysis (ANOVA), if it is different significantly, then it will be tested the difference between the treatment with Tukey Test (BNJ) on significance level 5 percent [10]. The entire calculation is done with Statistic 8 program.

III. RESULT AND DISCUSSION

A. Pyrolysis Equipment Performance

Liquid smoke maker equipment performance basically is based on distillation volume accumulated every one hour that is produced by condenser, like the formulation used by Hanendyo (11) shown on Table 1 below.

Based on the analysis result of liquid smoke maker pyrolysis equipment performance ranges from 5,79% to 16,29%. The result of variance investigation shows that combination of raw materials factor with pyrolysis

temperature difference shows the interaction and actual influence to equipment performance.

TABLE I
THE AVERAGE OF LIQUID SMOKE EQUIPMENT PERFORMANCE ON RAW MATERIALS WITH DIFFERENT PYROLYSIS TEMPERATURE FOR 5 HOURS

No	Liquid smoke type	Pyrolysis temperature	total distillation	Condenser length	Duration of pyrolysis	Equipment performance
			(ml)	(meter)	(hour)	(ml/hour.meter)
1	Coconut fiber	Temperature $100 \pm 10^\circ\text{C}$	405	14	5	$5.79 \pm 0,075^b$
		Temperature $200 \pm 10^\circ\text{C}$	600	14	5	$8.57 \pm 0,07^e$
		Temperature $300 \pm 10^\circ\text{C}$	660	14	5	$9.43 \pm 0,07^f$
		Temperature $400 \pm 10^\circ\text{C}$	900	14	5	$12.86 \pm 0,07^d$
2	Coconut shell	Temperature $100 \pm 10^\circ\text{C}$	750	14	5	$10.71 \pm 0,75^e$
		Temperature $200 \pm 10^\circ\text{C}$	900	14	5	$12.86 \pm 0,07^d$
		Temperature $300 \pm 10^\circ\text{C}$	935	14	5	$13.36 \pm 0,007^c$
		Temperature $400 \pm 10^\circ\text{C}$	1050	14	5	$15.00 \pm 0,07^b$
3	Cinnamon	Temperature $100 \pm 10^\circ\text{C}$	750	14	5	$10.71 \pm 0,075^e$
		Temperature $200 \pm 10^\circ\text{C}$	1050	14	5	$15.00 \pm 0,07^b$
		Temperature $300 \pm 10^\circ\text{C}$	1050	14	5	$15.00 \pm 0,07^b$
		Temperature $400 \pm 10^\circ\text{C}$	1140	14	5	$16.29 \pm 0,0751^a$

Note : Superscript letter that is different on average column shows actual difference ($P < 0,05$)

Further test with Tukey on $\alpha = 0,5$ shows the actual difference between the combination of raw materials treatment with pyrolysis temperature. The higher pyrolysis temperature then the tendency the tendency of equipment performance will become higher, it is caused by liquid smoke raw materials components decomposition. If equipment performance is reviewed from raw materials, it turns out that coconut fibre raw materials caused the result of lowest equipment performance like in the figure 1 below.

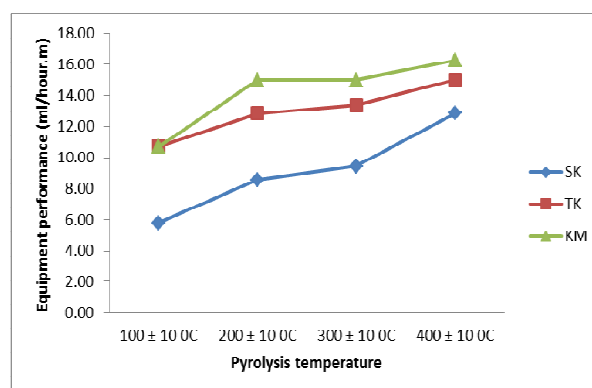


Fig. 1 Liquid smoke maker equipment performance based on treatment combination of raw materials difference and different pyrolysis temperature

Based on figure 1, generally equipment performance will be different with different pyrolysis temperature, the tendency shows with the higher pyrolysis temperature then the acquired volume distillation will cause higher equipment performance. The result in this research is not actually different with the result of research by Tranggono et al. [12], which is 52,85 %. They uses raw materials of various wood types and coconut shell also it is done on burning temperature $350 - 400^\circ\text{C}$.

Condensate that is newly produced has dark brown color specifically to coconut fibre liquid smoke, while coconut shell and cinnamon liquid smoke color is light brown like in figure 2. Dark brown color from liquid smoke is caused by its carbonyl-content. The higher its carbonyl-content, the higher its brown color potency [13], [14]. Coconut fibre liquid smoke has the most brown color so that it can be concluded that in that liquid smoke consists lots of carbonyl.

Cinnamon coconut shell liquid smoke has pungent smell like the smell from liquid smoke produced by burning cinnamon. This pungent smell comes from phenol compound in the liquid smoke [15]. The result of liquid smoke from coconut fibre, coconut shell and cinnamon can be seen on figure 2 below.



Fig. 2 Liquid smoke color of coconut fibre, coconut shell and Cinnamon

B. Physical characteristic from liquid smoke

Physical characteristic on the result of liquid smoke production research from several raw materials that consists of coconut fibre, coconut shell and cinnamon with different pyrolysis temperature can be seen on table 2 below.

TABLE II
THE AVERAGE OF LIQUID SMOKE PHYSICAL CHARACTERISTIC FROM DIFFERENT TREATMENT COMBINATION OF RAW MATERIALS WITH DIFFERENT PYROLYSIS TEMPERATURE

Sample code	Density	Rendement (%)	Color		
			L	a	b
SKT1	1.0267 ± 0,0153 b	24.263 ± 0,6807 d	3.73 ± 0,01	-0.27 ± 0,00de	1.2433 ± 0,0115g
SKT2	1.0100 ± 0,01 c	24.510 ± 0,8996 d	3.92 ± 0,021	-0.31 ± 0,00f	1.0300 ± 0,02j
SKT3	0.9967 ± 0,035d	25.063 ± 0,6807 cd	3.23 ± 0,01	-0.20 ± 0,00b	1.0300 ± 0,02j
SKT4	1.0167 ± 0,0153 c	25.510 ± 0,8996 cd	3.9 ± 0,01	-0.19 ± 0,00b	1.0300 ± 0,02j
TKT1	1.0167 ± 0,0154 c	30.270 ± 0,2646 bc	4.74 ± 0,021	0.43 ± 0,02a	2.8100 ± 0,02a
TKT2	1.0467 ± 0,0153 a	32.477 ± 1,9531 b	5.01 ± 0,021	-0.25 ± 0,00cd	2.1100 ± 0,02b
TKT3	1.0267 ± 0,0153 b	32.590 ± 2,0172 b	2.92 ± 0,01	-0.24 ± 0,00c	1.0800 ± 0,02i
TKT4	1.0267 ± 0,0153 b	34.423 ± 1,0092 ab	2.58 ± 0,01	0.42 ± 0,02a	1.6700 ± 0,02c
KMT1	1.0467 ± 0,0153 a	34.077 ± 0,6120 ab	3.51 ± 0,01	-0.28 ± 0,00e	1.2200 ± 0,02h
KMT2	1.0267 ± 0,0153 b	34.213 ± 0,5733 ab	3.05 ± 0,01	-0.23 ± 0,00c	1.5400 ± 0,02e
KMT3	1.0267 ± 0,0153 b	34.733 ± 1,0551 ab	2.96 ± 0,021	-0.18 ± 0,00b	1.3900 ± 0,02f
KMT4	1.0167 ± 0,0153 c	38.780 ± 6,2155 a	2.95 ± 0,01	-0.18 ± 0,00b	1.6300 ± 0,02d
Japan standard **)	>1,005		Yellow brown and transparent		

Note : SK = Coconut fibre, TK = Coconut shell, KM=cinnamon
T1 = 100±10°C, T2 = 200±10°C, T3 = 300±10°C, T4 = 400±10°C
* Superscript letter that different on average column shows actual difference (P<0,05),
**Quoted from Yatagai (2002)

1) Density (g/ml): Liquid smoke density is a comparison between the liquid smoke weights to raw materials weight. Liquid smoke density shows the level of liquid smoke density, it means that the higher liquid smoke then its density will become higher. Based on table 2 above that statistically the average of coconut fibre raw materials treatment combination with pyrolysis temperature 100 ± 10°C with 200 °C ± 10°C has actual difference, it also has actual difference with the temperature of 3000C ± 10°C and the temperature of 4000C ± 10°C. Similar thing also happens to raw materials coconut shell treatment combination with pyrolysis temperature of 100 ± 10°C with 2000C ± 10°C has shown actual difference, it also has actual difference with temperature 3000C ± 10°C and temperature 4000C ± 10°C. For cinnamon treatment combination with pyrolysis temperature of 100 ± 10°C with 2000C ± 10°C has shown actual difference, it also shows actual difference with temperature 3000C ± 10°C and temperature 4000C ± 10°C. This difference is suspected that pyrolysis temperature has influence to liquid smoke component decomposition so that it has difference statistically.

Liquid smoke density on Table 2 shows that there is actual interaction (P <0.05) between factor A (raw materials type) with factor B (pyrolysis level temperature) on liquid smoke after distillation. Figure of liquid smoke density on raw materials treatment combination with pyrolysis temperature like figure 3 below.

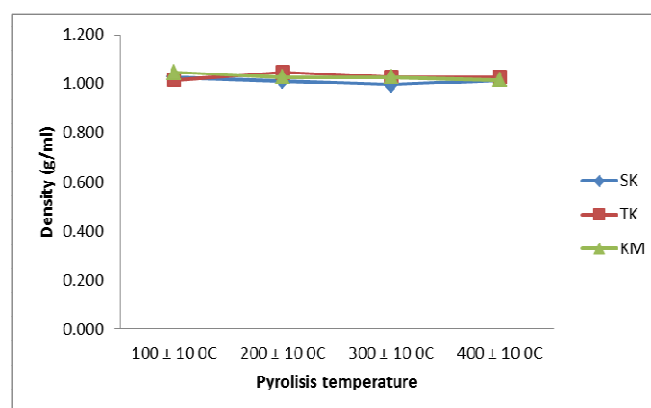


Fig. 3 Liquid Smoke Density Average from Various Raw Materials Type with Different Pyrolysis Temperature Level

On figure 3 it can be seen that liquid smoke density from coconut shell from raw materials on pyrolysis treatment 200°C and cinnamon on pyrolysis treatment 100°C has similar density at 1.0467 mg/ml is higher compared with other treatments. This result is suspected to have close relationship with liquid smoke rendement-content. Pyrolysis on temperature 400°C has lignine decomposition that when a material consists too many lignine factions then it will increase the produced rendement. Temperature 300°C is selected as the burning temperature, because on the temperature of 300°C, decomposed cellulose component produces organic acids and some phenol compound [9], [23]. Burning temperature of 300°C produces better liquid smoke quality compared to temperature 500°C because it causes fewer undesired ter [5].

Liquid smoke density value is greater from the result of research done by Nurhayati [18], ranged between 1,004-

1.018 and Komarayati research [20] with range between 0.97-0.99. However, this research result on density value is smaller compared with the result of research done by Nurhayati and Adelina [19] that is ranged between 1.085 to 1.095 (Table 4). Measurement result of liquid smoke density as the result of different raw materials treatment combination with pyrolysis temperature produces greater average density from 1.005. It means that the result has closely approach the result of Yatagai's research [22].

2) *Rendement (%)*: Liquid smoke rendement is a percentage of the liquid smoke amount that is produced by a raw material through pyrolysis process to the weight of raw materials before the beginning of pyrolysis. Total liquid smoke rendement that is produced on pyrolysis process really depends on raw materials type that is used. Rendement percentage that is acquired also relies heavily on pyrolysis temperature that is used. The result of research on Table 2 shows that cinnamon raw materials combination with pyrolysis temperature 400°C statistically does not have actual difference with treatment combination of pyrolysis temperature 300°C, 200°C and 100°C and produce the highest liquid smoke rendement compared to the other two materials. The high rendement value is supposedly that on pyrolysis temperature of 400°C, lignine decomposition happens so that if the material consists lots of the lignin fraction, then it will increase the produced rendement. The difference of rendement percentage that is acquired is caused by the diverse lignin and cellulose contents of each material among others 38.98 - 63.09% to cellulose and 19.35 - 50.44% to lignine [12]. The high and low of liquid smoke rendement on pyrolysis process is influenced by several factors, such as climate, season, age of the plant, type of the plant, raw materials and way of burning. Figure of liquid smoke rendement average (%) on raw materials treatment combination with different pyrolysis temperature like figure 4 below.

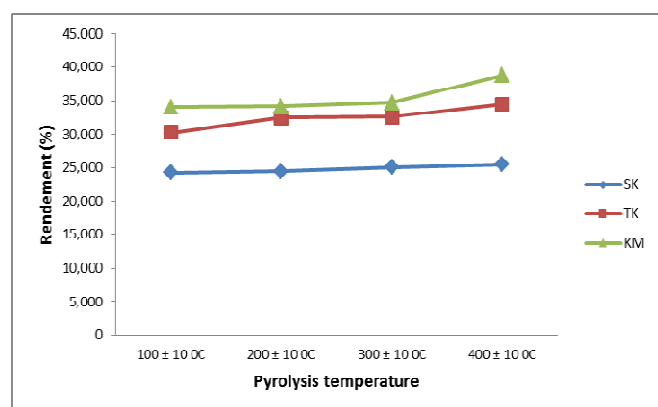


Fig. 4 Rendement Average (%) from Several Type Liquid Smoke Maker Raw Materials with Different Level of Pyrolysis Temperature

Liquid smoke production result on pyrolysis with temperatures of 400°C on cinnamon raw materials has the highest rendement value (38.78%) while the lowest value on coconut fibre raw materials on pyrolysis temperature of 100°C by 24.26%. However, coconut fibre liquid smoke rendement on pyrolysis temperature of 100°C does not have actual difference with pyrolysis temperature on 200°C, 300°C and 400°C and all treatment show that the result is

relatively the same. This condition is usually closely associated with the volume of initial raw materials water-content. Pyrolysis on temperature that is too high and the time that is too long will decrease the formation of liquid smoke. This is in accordance with the statement of Demirbas [24] that wood material pyrolysis process liquid smoke result can be produced maximally if the condensation process happens perfectly. The difference of smoke distillation rendement amount is caused by the fact that the higher content of water in raw materials then the higher the produced smoke distillation rendement amount and the longer the condenser then the chances to condition the imperfect burning result in process of smoke distillation extraction will be more optimal [16].

Liquid smoke rendement that is produced on this research is lower compared to the result of liquid smoke acquired by Tranggono et al. [12] on pyrolysis of several wood types with temperature range of 350-400°C produce liquid smoke with average rendement of 49.1%. The total liquid smoke rendement produced on pyrolysis process really depends on raw materials type that is used. The acquired rendement percentage also really depends on condensation system that is used. This condition is in accordance with Tranggono et al [12], that for the formation of liquid smoke, water is used as a cooling medium so that the heat exchange process can occur fastly. Pyrolysis on temperature that is too high and the time that is too long will reduce the formation of liquid smoke. This is in accordance with the statement of Demirbas [24] that liquid smoke by wood material pyrolysis process can be produced maximally if the condensation process happens perfectly.

3) *Analysis of Color (L, a, b) (HunterLab ColorFlex EZ spectrophotometer)*: Liquid smoke raw materials treatment combination liquid smoke color with pyrolysis temperature was measured by using equipment HunterLab ColorFlex EZ that produces value L, a* (-), and b* (+). L value shows the colour brightness, a* (-); greenish, b* (+); yellow, the higher the value of L (Lightning) shows that liquid smoke becomes brighter, the higher the value of b* (+) the colour of liquid smoke becomes more yellow, the higher the value of a* (-) the colour of liquid smoke becomes more green. The result of research shows that the brightness (L) from coconut shell liquid smoke on different pyrolysis temperature of (+) 3.81 with value a*: (+) 0:09 and b* (+) +1.91, then brightness (L) on coconut fibre liquid smoke on different temperature pyrolysis of (+) 3.65 with the value of a* (-) 0:24 and b* (+) 1:52 and also brightness (L) of cinnamon liquid smoke on different pyrolysis temperature (+) 3,11 with the value of a* (-) 0.06 and b* (+) 1:38 after being converted according to Figure information, the colour is around brownish-yellow. The brightness of liquid smoke is less white. Liquid smoke as the result of pyrolysis with different temperature on average has reddish-brown colour. This is caused by the content of tar compound that basically has black colour and components that have high molecular weight, therefore there will be further research for purification process with intent to produce liquid smoke with clearer colour and no poison, so when applied on food material, it will produce interesting, not dark smoked product colour. The difference of liquid smoke colour from each faction is influenced by the presence of tar. Coconut fibre liquid smoke has the darkest

colour because on that temperature, there is possibility of tar entrainment, while somewhat reddish-brown is from cinnamon liquid smoke. The analysis result of this liquid smoke colour is rather different with liquid smoke colour from coconut shell that on the fraction I light yellow and fraction II whitish-yellow [25]. This is because liquid smoke consists of 1.45% phenol compound. The emergence of brownish yellow colour might be due to the fact that liquid smoke consists of phenol compound [26]. In Judd-Hunter, the colour can be represented by L (light or dark, 0 = perfect black to 100 = perfect white), a (-a = greenish, + a = reddish) and b (-b = blueish, + b = yellowish).

4) To analysis the brightness (L) value diversity of liquid smoke maker raw materials combination with different pyrolysis temperature shows that there is no interaction with the value of $P < 0.05$. Figure of average colour (L) liquid smoke on raw materials treatment combination with pyrolysis temperature shown in figure 5 below.

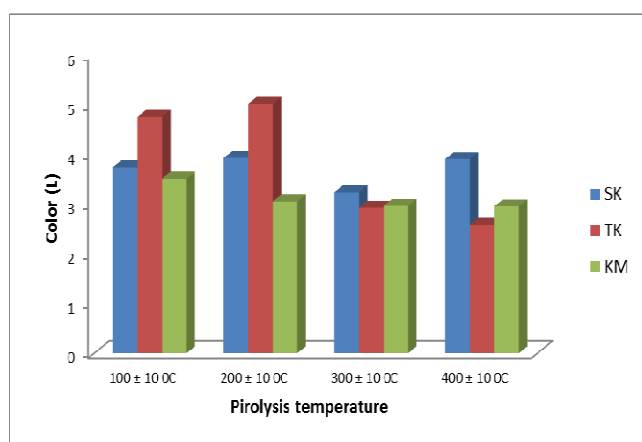


Fig. 5 The Average of Colour (L) from Several Liquid Smoke Maker Raw Materials Type with Different Pyrolysis Temperature Level

For colour a (Table 2) has range from value + 0.43 to - 0.31, it means from greenish until approaching reddish colour, and statistically treatment combination of liquid smoke raw materials with pyrolysis temperature shows that there is real interaction (appendix 5e), further on test HSD 5% on table 2 above shows liquid smoke raw materials treatment combination from coconut shell on pyrolysis temperature of 100°C does not have actual difference with pyrolysis temperature of 400°C because from the colour, both show equally approaching greenish colour, it has actual difference with pyrolysis temperature of 200°C and 300°C and from terms of colour, both are similarly leads to reddish colour. Figure of average colour (a) liquid smoke on treatment combination with raw materials with pyrolysis temperature like figure 6 below.

For colour b (Table 2) ranges from values +1.03 to +2.81, it begins to approach yellowish colour, and statistically treatment combination of liquid smoke raw materials with pyrolysis temperature shows that there is actual interaction, further on test Tukey 5% on Table 2 shows treatment combination of liquid smoke raw materials from coconut shell on pyrolysis temperature of 100°C has no actual difference with pyrolysis temperature 200°C, there is actual difference with pyrolysis temperature to 300°C and 400°C and from the terms of colour both are equally approach

yellowish colour. The figure of average colour (b) liquid smoke on raw materials treatment combination with pyrolysis temperature can be seen in Figure 7 below.

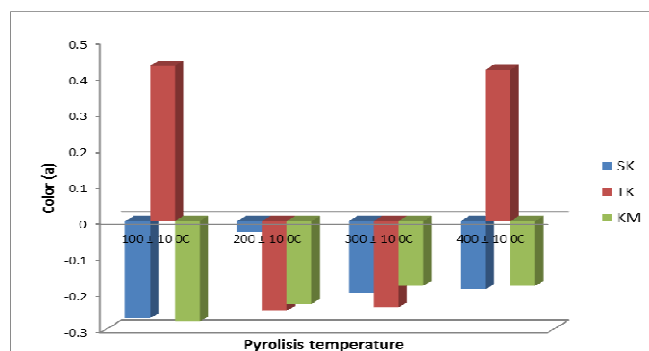


Fig. 6 The Average of colour (a) from Several Liquid Smoke Maker Raw Materials Type with Different Pyrolysis Temperature Level

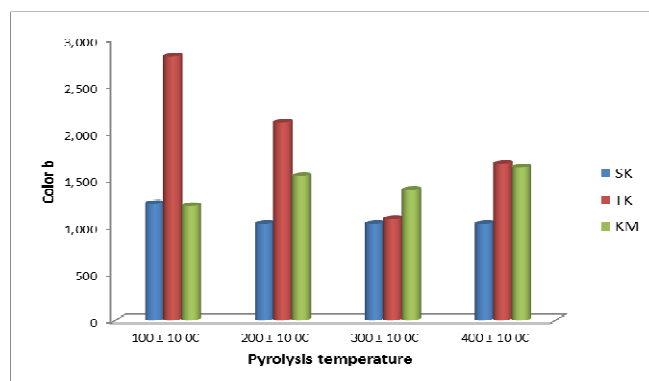


Fig. 7 The Average of Colour (b) from Liquid Smoke Maker Different Raw Materials Type with Different Pyrolysis Temperature Level

Liquid smoke colour is affected by pyrolysis temperature that causes degradation (cellulose, hemicellulose and lignin). It is reinforced by the result of research from Wijaya [27] that temperature change cause the colour change on liquid smoke. The higher pyrolysis temperature then the darker the colour of produced liquid smoke. In the similar aspect, wood liquid smoke is the brownish-yellow/darkish-brown colour liquid acquired from side-result of making charcoal [20]. It was confirmed by Girard [9] that on temperature 200-450°C there is hemicellulose, cellulose and lignin degradation in wood. Liquid smoke colour in the result of this research is almost the same with the result of research by Komarayati, [20] with brownish-yellow colour, but it will be different from the result of research done by Nurhayati [18] and Nurhayati and Adalina [19], which is darkish-brown colour. Liquid smoke colour with bright colour have better quality compared with liquid smoke with dark colour. Liquid smoke colour in the result of this research fulfils the quality of Japanese liquid smoke specification. Transparency of cinnamon liquid smoke on pyrolysis temperature 400°C is transparent so that it meets the quality standards of Japanese liquid smoke specification (Table 2). The result of this research is similar with the result of research done Komarayati [20] that the transparency of wood liquid smoke is not cloudy or transparent. The more transparent liquid smoke, the better the quality of that liquid smoke.

C. Chemical Characteristic of Liquid Smoke

Chemical characteristics from result of research of liquid smoke production from several raw materials that consists of coconut fibre, coconut shell and cinnamon with different pyrolysis temperature as in Table 3 below.

TABLE III
THE AVERAGE OF CHEMICAL CHARACTERISTIC ON DIFFERENT RAW MATERIALS TREATMENT COMBINATION LIQUID SMOKE DIFFERENCE WITH DIFFERENT PYROLYSIS TEMPERATURE

Sample code	pH	Total acid (%)	Phenol-content (%)	Carbonyl (%)	Benzoepypren (ppm)
SKT1	4.8200 ± 0,1706 a	0.2± 0,035	0.1700± 0,00 i	1.1467± 0,0153 k	5.2300± 0,1520 e
SKT2	4.7700 ± 0,1706 a	0.12± 0,035	0.2133± 0,053 h	1.0567± 0,0153 l	5.5700± 0,1620 de
SKT3	2.5733 ± 0,0874 g	0.12± 0,035	0.2000± 0,000 hi	1.9067± 0,0153 i	5.8900± 0,1720 cd
SKT4	4.5733 ± 0,1595 b	0.12± 0,035	0.1767± 0,053 i	1.4267± 0,0153 j	6.4200± 0,1870 bc
TKT1	3.6667 ± 0,1290 de	1.4± 0,035	1.1000± 0,044 d	3.0167± 0,0252 g	4.5633± 0,6866 f
TKT2	3.6967 ± 0,1290 de	0.62± 0,035	1.4600 ± 0,0495 e	3.5867± 0,0351 e	5.7800 ± 0,1680 de
TKT3	3.6267± 0,1290 e	1.62± 0,035	1.6067± 0,031 b	2.7867± 0,0252 h	6.9700± 0,2030 b
TKT4	3.5033 ± 0,1234 f	1.15± 0,035	1.7300± 0,049 a	4.1767± 0,0451 c	8.6200± 0,2510 a
KMT1	4.0100 ± 0,1442 c	1.12± 0,035	0.6500± 0,041 f	3.2367± 0,0351 f	1.0100 ± 0,0290g
KMT2	3.7067 ± 0,1290 d	1.19± 0,035	0.7767± 0,042 e	4.6767± 0,0451a	0.0900 ± 0,033 h
KMT3	3.4700 ± 0,1179 f	0.85± 0,035	0.6567± 0,042 f	4.4067± 0,0451b	0.0600 ± 0,033 h
KMT4	3.5033 ± 0,1234 f	0.72± 0,035	0.5700± 0,041 g	4.1267± 0,0451d	0.0400 ± 0,000 h
Japan standard **)	1,5 - 3,7				

Notes : SK = Coconut fibre, TK = Coconut shell, KM=cinnamon
T1 = 100±10°C, T2 = 200±10°C, T3 = 300±10°C, T4 = 400±10°C
* The different superscript letter on average column shows the actual difference P<0,05)
**Quoted from Yatagai (2002)

1) pH: The principle in testing degree of acidity (pH) by using pH meter equipment is a pH measurement method based on hydrogen ion activity measurement potentiometrically/electrometrically by using pH-meter. Before using, pH-meter equipment is calibrated with buffer solution according to equipment instruction on every measurement.

Result of further test on HSD 5% in tabel 2 shows that coconut fibre liquid smoke produces the lowest pH on pyrolysis temperature level 300°C± 10°C compared to liquid smoke pH from two other raw materials with different pyrolysis temperature. Statistically liquid smoke pH with treatment combination of coconut fibre with pyrolysis temperature 100 ± 10°C does not have actual difference with pyrolysis temperature 200°C ± 10°C, actual difference with temperature 300°C± 10°C and temperature 400°C ±10°C. While on coconut shell liquid smoke pH on treatment combination with pyrolysis temperature 100± 10°C does not have actual difference with 200±10°C, then actual difference with temperature 300± 10°C and 400± 10°C. Next on cinnamon liquid smoke on treatment combination with pyrolysis temperature 100±10°C has actual difference with 200± 10°C, then it also has actual difference with temperature 300± 10°C and 400± 10°C. Statistically, the difference on coconut fibre liquid smoke with different temperature pyrolysis supposedly on pyrolysis temperature above 100°C the component decomposition of liquid smoke

has started, and the higher pyrolysis temperature, liquid smoke pH has tendency to decrease.

On pH values on average variance analysis shows the result of actual interaction (P <0.05) between treatment A and treatment B. Figure of liquid smoke pH condition on treatment combination of raw materials with pyrolysis temperature like in figure 8 below.

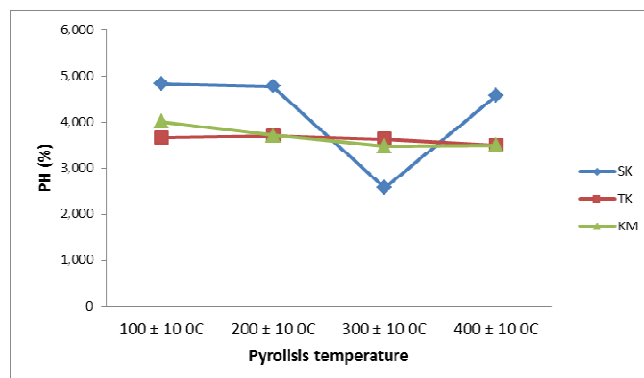


Fig. 8 Graphic of Liquid Smoke pH Average Value from Several Raw Materials Type with Different Temperature Level

The average of pH values in figure 8 shows that in pyrolysis temperature of 100°C-300°C for raw materials of coconut shell and cinnamon is lower than the result from coconut fibre raw materials. It is suspected that pH value is closely related with hemicellulose fractions decomposition that produce acetic acid, in accordance with Sutin research [16] mentions that coconut shell has hemicellulose component and cellulose bigger than coconut fibre so that the amount of acid is produced bigger. Hemicellulose and cellulose are wood components that if it is decomposed, it will produce organic acid compounds such as acetic acid. Besides it is also caused by the liquid smoke density level produced where liquid smoke from coconut fibre raw materials is the lowest compared to the other two raw materials shown by liquid smoke density is successively 1.00; 1.02; and 1.01. This result is in accordance with the research of Haji, et. al. [17] that the average of pH value on three treatment combination shows the result of average pH values in liquid smoke produced on organic waste pyrolysis process has tendency to diverse according to the variation of water-content of each used raw materials.

In liquid smoke pH value, the lower is better because its antibacterial force also becomes higher. The result of research shows that liquid smoke pH from pyrolysis of three raw materials on four different temperature ranges 2,57- 4.82. If pH value is low, it means that produced smoke has high quality, especially in terms of using as food preservatives [18]. The low pH value can entirely influence to the value of product smoke or organoleptic characteristic durability and storability. Because of that low pH on microbes or bacteria as the obstruction in preservatives process tend not to survive and breed. In accordance with the explanation Haji et al, [17], measurement of pH values in liquid smoke is intended to know the level of raw materials decomposition process by pyrolysis. The low pH value can have overall influence to the value of durability and storability of a product. The liquid smoke pH value in this research result, the average pH is almost similar if compared with the result

of research by Nurhayati [18], about 4.3-4.7, Nurhayati and Adalina [19] about 3.2-3.8 then Komarayati, [20] about 3.2-6.80. But it is bigger from the result of research done by Amperawati [21] with pH value ranges from 2.91. For the range of liquid smoke PH measurement result on different raw materials treatment combination with pyrolysis temperature is associated with liquid smoke quality standards from Yatagai [22] on average it is in the range of 1.5-3.7. Especially to coconut fibre with different pyrolysis temperature that produces greater pH than result of research from Yatagai [22].

2) *Total acid (%)*: Liquid smoke total acid variance analysis can be seen in attachments 6a, shows that there is no interaction between treatment combinations of liquid smoke raw materials with pyrolysis temperature. In this conditions, there will be no further tests. The figure of titrated acid-content average can be seen on figure 9 below.

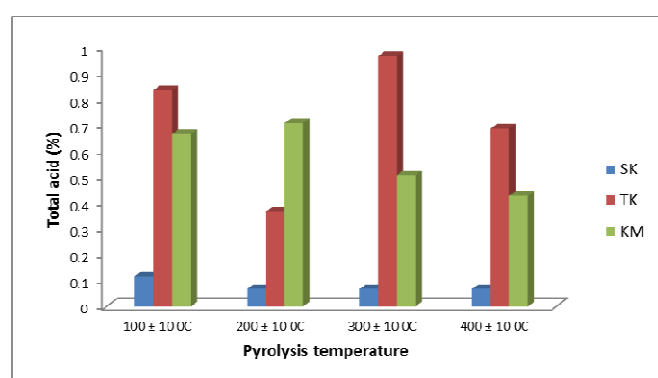


Fig. 9 Graphic of Liquid Smoke Titrated Acid Total from Several Type of Raw Material with Different Pyrolysis Temperature Level

From the observation result on table 3 and figure 9 above, the average of titrated result acid ranges between 0.06 to 0.97%, for the average of total acid, coconut fibre is the lowest and the average of total acid for coconut shell is the highest. The diversity analysis result shows that there is no real interaction ($P > 0.05$) between liquid smoke raw materials treatment with pyrolysis temperature level to titrated acid total, it can be seen that the total value of titrated acid is higher on the treatment combination of coconut shell on pyrolysis temperature 300°C at 0,97% compared with coconut fibre on pyrolysis temperature 300°C at 0.03% and cinnamon on pyrolysis temperature 300°C of 0.51%. The tendency of acid total value on combination of coconut fibre raw materials with the increasing of pyrolysis temperature becomes lower. The same thing also happens on cinnamon raw materials, it shows that the more pyrolysis temperature increases then the tendency of total acid value decreases, this is different with combination of coconut shell raw materials with different temperature, it does not shows such phenomenon.

This difference is because the different raw materials types is used, difference pyrolysis temperature and water-content on raw materials. This is in accordance with the opinion from Darmadji [15], he mentions that pyrolysis process on wood constituents such as cellulose, hemicellulose and lignine will produce organic acid, phenol, carbonyl as the compounds that have important role in food preservatives. Those compounds have different proportions,

such as depending on the type of raw materials, wood water-content, and pyrolysis temperature. Water-content of coconut shell after pyrolysis on temperature 400°C is 3.28%, coconut fibre with temperature 400°C is 9.00% and cinnamon with temperatures 300°C is 6.61%.

On Figure 9, it also can be seen that on pyrolysis temperature 300°C on coconut shell raw materials shows an increase in the titrated acid value. This is due to the temperature 300°C, cellulose and hemicellulose components is decomposed to form acid compounds. This is in accordance with the research by Luditama [5] that tries to compare the best burning process condition to produce liquid smoke from coconut shell and coconut fibre by using temperatures of 300°C and 500°C. Based on that research, it can be seen that temperatures of 400°C can produce liquid smoke with better quality. Besides, on temperatures of 400°C, it also produced lower tar compound where tar is the compound that must be removed to produce liquid smoke with better quality. The quality from liquid smoke is also determined by the purity from the compounds. Liquid smoke consists of compound acid group and its derivatives, alcohols, aldehydes, hydrocarbons, ketones, phenols and pyridine [28]. Acids do not have specific influence but it has general effect on the overall organoleptic quality [9]. The acid products and its derivatives have potential as natural preservatives [27]. Furthermore, acetic acid and furan begin to form on pyrolysis temperature of 260°C as the result from hemicellulose degradation [23]. Wood pyrolysis experiences gradual decomposition, that is the hemicellulose is degraded on temperature 200-260°C [29]. The range of acidity values that is acquired (0.12%-1.62%) is lower to the result of research of Darmadji [15], Kadir, (30), Amperawati, [21] and Apituley and Darmadji [31]. The result of research meet the quality standards of Japanese liquid smoke specifications.

3) *Phenol-Content (%)*: Result of variance analysis shows that liquid smoke raw materials difference treatment combination with pyrolysis temperature gives influence of significant actual difference to phenol-content ($P < 0.01$). Liquid smoke phenol-content data (Table 3) shows treatment combination of coconut shell material with pyrolysis temperatures of 400°C shows the highest phenol-content compared to both of others raw materials. Statistically ($P < 0.05$) treatment combination of coconut shell with temperatures of 400°C has actual difference with treatment combination of temperature to 300°C, 200°C and 100°C. The high value of phenol-content on coconut shell depends on raw materials condition. This is in accordance with the opinion by Girard [9] who mentions that the composition of chemical substances that are contained in liquid smoke can be influenced by several factors, such as: type of wood or raw materials that is used, water-content from material, burning temperature, burning duration, condensation temperature and purification type that is done. Phenols-content on liquid smoke usually has range between 0.2 - 2.9% [23]. For the analysis result of phenol-content from the three raw materials above almost all of them range between 0.2 - 2.9% except for coconut fibre material that is pyrolyzed on temperature of 100°C and 400°C. Phenol is the result of wood component disintegration, which is lignin. The more of lignin content in the wood, then the greater of phenols-content on liquid smoke. The quantity and quality of phenol

compound that can be found on liquid smoke is related with lignin-content and pyrolysis temperature [9]. Furthermore, lignin degradation occurs on temperature 310-500°C, when that temperature has not been reached then it will influence lignin degradation and phenol content from liquid smoke that is produced [23]. The range of values from the result of research that is acquired (0.17% - 1.73%) is lower than the result of research done by Kadir [30], that phenol-content acquired on hybrid coconut shell liquid smoke of 4,71%. The value of phenol-content of this research is smaller than the result of research done by Darmadji [15] with the range of phenol-content from 2.11 - 3.13. But smaller than the result of research by Amperawati [21] with phenol-content of 12.29%. Table 4 is provided with some researches related with liquid smoke quality. The result of this research is to compare the result of research on liquid smoke quality from coconut fibre, coconut shell and cinnamon raw materials below.

TABLE IV
THE COMPARISON OF LIQUID SMOKE QUALITY FROM DIFFERENT RAW MATERIALS TREATMENT COMBINATION WITH PYROLYSIS TEMPERATURE WITH SEVERAL RESULT OF RESEARCH

Parameter (Parameters)	Alvian et al., 2014	Darmadji, 1996	Nurbayati et al., (2006)	Nurbayati dan Adhena, 2009 (Narkayu and Adhena 2009)	Kadir et al., (2010)	Komarajati et al., (2011)	Amperawati et al., (2012)	Apituley dan Darmadji, 2013 (Apituley and Darmadji, 2013)
Warna/Color	Kuning coklat kemerahan (Yellow reddish)		Coklat kehitaman (Blackish brown)	Coklat kehitaman (Blackish brown)		Kuning kecoklatan (Yellow brownish)		
Bahan tercampur (Material of flour)	Tidak ada bahantercampur (No flour mixed)							
Transparansi (Transparency)	Transparan (Transparent)		Ada suspensi (Suspension)	seikit keruh (opaque)		Transparan (Transparent)		
Berat jenis (Specific gravity)/cm	1,008-1,058		1,004-1,018	1,083-1,105		0,97-0,99		
pH	3,105-3,240	3,105-3,195	4,3-4,7	3,2-3,8		3,20-6,80	2,9	
Kesamaan (Aidity)/%	6,722-12,903	5,167-12,903			12,57		8,67	11,97
Karbonil (Carbonyl)/%	10,850-28,307	11,250-30,300			13,19		12,53	10,05
Fenol (Phenol)/%	2,562-9,231	2,429-9,231			4,71		12,20	2,73
Bahan perendahan (Material source)	Batang gelam (Saw of gelam)	Limbah pertanian (Agricultural waste)	Limbah serbuk gergaji kayu campuran (Waste sawdust of mixed wood)	Sabutan kayu campuran (Shells of mixed wood)	Tempurung kelapa hibrida (Shell of coconut hybrid)	Limbah mahoni, nangka dan sengon (Waste of mahoe, jackfruit and sengo)	Tempurung kelapa hibrida (Shell of coconut hybrid)	Kulit batang sago (Bark of sago)

For figure of raw materials treatment combination with pyrolysis temperature can be seen on figure 10 below.

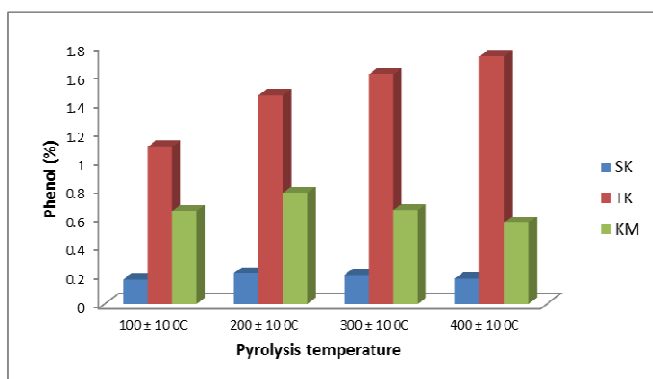


Fig. 10 The Average of Phenol-Content Value (%) from Several Raw Materials Type with Different Pyrolysis Temperature Level

4) Carbonyl-Content (%): The result of variance analysis shows that liquid smoke of different raw materials combination treatment with pyrolysis temperature shows

influence of significant actual difference to carbonyl-content (P<0:05).

Liquid smoke carbonyl-content like on table 3 shows that cinnamon treatment combination with pyrolysis temperature of 200°C gives the result of highest carbonyl-content compared to both of other raw materials. Statistically (P>0.05) cinnamon treatment combination with temperature of 200°C has actual difference with the treatment combination of temperature 300°C, 400°C and 100°C. The high value of carbonyl-content on cinnamon depends on raw materials. This is in accordance with the opinion of Girard [9], he mentions that composition of chemical substances that are contained in liquid smoke can be influenced by several factors such as: type of wood or raw materials that is used, water-content from material, burning temperature, burning duration, temperature condensation and purification type that is done. On carbonyl content of liquid smoke usually has range between 2.6 - 4.6% [23]. For the analysis result of carbonyl-content from the three raw materials above almost all range between 2.6 - 4.6%, except for coconut fibre material that is pyrolyzed on temperature of 100°C, 200°C, 300°C and 400°C. For figure of raw materials treatment combination with pyrolysis temperature can be seen in figure 11 below.

The average of carbonyl result on figure 11 shows carbonyl-content of cinnamon liquid smoke is greater compared to coconut shell and coconut fibre raw materials. Cellulose degradation process occur on temperatures 280-320°C, Cellulose degradation produces acetic acid compound and carbonyl compound [9]. This is also clarified by Sjostrom [29] that wood cellulose experiences decomposition on temperature of 240-350°C. The range of carbonyl-content value that is acquired (1.056% - 4.6767%) is smaller than the range of value from several results of researches, Darmadji [15] has range between 9.30 - 17.14%, Kadir [30] with 13.19% and Amperawati [21] with 12.29%, it is also smaller from the result of research by Apituley and Darmadji [31], with carbonyl-content of liquid smoke from sago cortex by 10.05% (Table 4).

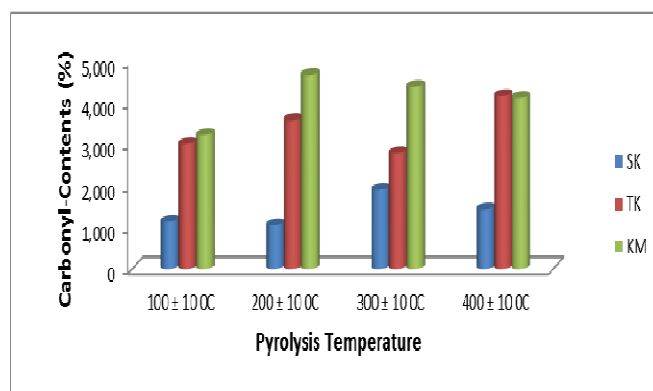


Fig. 11 The Average of Carbonyl-Content Value (%) from Several Raw Materials Type with Different Pyrolysis Temperature Level

5) Benzo(a)pyren-Content from Liquid Smoke: The result of variance analysis shows that treatment combination of different liquid smoke raw materials with different pyrolysis temperature shows the real influence to benzo(a)pyren-content from liquid smoke (P<0:01). Further on Table 3

above shows that the result analysis of benzo(a)pyren-content from coconut fibre, coconut shell and cinnamon raw materials show that liquid smoke from pyrolysis temperature of 400°C on cinnamon has the lowest value (<0.04) compared with liquid smoke from the other materials. This result indicates that cinnamon consists lower benzo(a)pyren-content in the process of pyrolysis. The higher of pyrolysis temperature on cinnamon raw materials, the tendency of benzo(a)pyren-content will decrease. It is suspected that the higher temperature, it will decompose the compounds in cinnamon. For liquid smoke security as food preservatives material, the effort needs to be taken so that benzo(a)pyren-content as small as possible because this compound is very dangerous for humans with its carcinogenic nature. Figure of benzo(a)pyren-content on raw materials treatment combination with pyrolysis temperature can be seen in figure 12 below.

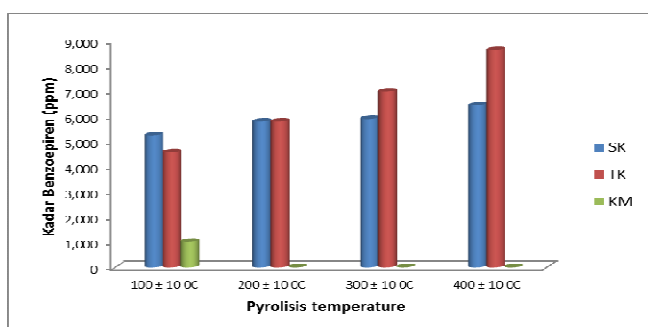


Fig. 12 The Average of Benzo(a)pyren-Content Value from Several Raw Materials Type with Different Pyrolysis Temperature Level

Graphics on figure 12 shows that cinnamon raw materials produces better quality liquid smoke than the other two raw materials, which has the lowest value of benzo(a)pyren-content at 0.04 ppm. Benzo(a)pyren levels of coconut fibre and coconut shell with the higher pyrolysis temperature, it has the tendency to increase. It is different with cinnamon liquid smoke that the higher pyrolysis temperature, benzo(a)pyren-content tends to decline. According to FAO and WHO [32] that permit the maximum of Benzo(a)pyren of 10 µg/kg on food (Joint FAO/WHO Expert Committee on Food Additives, 1987). According to the regulations of Head of National Agency of Drug and Food of the Republic of Indonesia number HK.00.06.1.52.4011 [33], mentions that the maximum limit of the chemical contamination of benzo(a)pyren on smoked fish is at 5 ppb/mcg/kg.

IV. CONCLUSIONS

The result this research on the production of liquid smoke that is produced by various raw materials in various level of temperature pyrolysis shows: Liquid smoke maker raw materials from cinnamon is very potential to be developed to produce liquid smoke, that can produce better chemical and physical qualities better because it can meet the standard in Japan rather than the other raw materials of coconut fibre and coconut shell. Various raw materials with different combination of temperature level has influence on the performance of pyrolysis equipment, liquid smoke chemical and physical qualities. Benzo(a)pyren toxic content compound in cinnamon liquid smoke has the lowest compared to

coconut shell and coconut fibre materials on pyrolysis temperature of 400°C

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