

## Characterization of Liquid Smoke from Coconut Shell to Be Applied as Safe Food Preservatives for Human Health

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### ABSTRACT

Rubber industries' major problem is the unpleasant smell of raw materials deterioration that would trigger the growth of antioxidant decomposer bacterias since it was brought from the plantation. Liquid smoke, is a condensed smoke from pirolized coconut shell, had been used as smell neutralizer in these industries to overcome the problem. This research is aimed to characterize the liquid smoke properties that is resulted from pirolisis of 30 kilograms of coconut shell waste using pirolisis reactor under various operational temperatures, which are 300, 350, 400, 450 and 500°C for 5 hours. Resulted liquid smoke is analyzed using Gas Chromatography -Mass Spectrometry (GCMS). Based on its analysis, it was proceeded the yields and pH values for each temperature variable are, respectively, 7.15% and 1.64, 6.87% and 2.13, 11.83% and 1.23, 5.24% and 1.46, 4.33% and 1.27 whereas the phenol contents are, respectively, 3.48%, 2.92%, 4.63%, 3.67% and 4.62%. Nevertheless, liquid smoke that is gained from the pirolized coconut shell waste has not food grade yet so it can not be applicated on food due its harmness to human health but it is used in wood and plantation industries.

**KEYWORDS:** Coconut shell, pirolisis, liquid smoke, antioxidant.

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### INTRODUCTION

Nutritional food as energy source for human body is significantly required in performing daily physical activities. Nonetheless, the widespread use of formalin in food preservation is posing a serious threat to human health. Liquid smoke, a condensation product of coconut shell pyrolysis, could be suggested for being an alternative solution to overcome the problem. Pyrolysis is a thermochemical decomposition of high-content-carbon materials at elevated temperatures in the absence of oxygen which occurs in a closed-stainless steel reactor and generally lasts within 4 until 7 hour-process (Paris, et.al., 2005). In addition, electricity power and organic waste incineration generate heat energy into the process.

The liquefied smoke has been developed as food preservative agent, food antioxidant and biopesticide (Nurhayati, 2000). It consists of dispersed gas-phase in water system (Darmaji, et.al., 1995). Furthermore, composition and organoleptic properties of the liquefied smoke fundamentally depends on pyrolysis temperature, quantity of oxygen, humidity, particle size, apparatus type and raw materials content such as wood characteristics (Girard, 1992). In conclusion, temperature is the most valuable affecting factor of pyrolysis.

The condensed smoke delivers an ability in preserving food due to the presence of acids, carbonil and phenolic compounds. Maximum capacity of those compounds is deliberated at 600°C, satisfactory organoleptic property is pursued at 400°C.

### MATERIALS AND METHODS

This research begins with the pyrolysis process with coconut shell material. Pyrolysis process is the process of separating the material by heating without direct fire, at first 30 kilograms of crushed coconut shell into pyrolysis reactor (capacity 50 kilograms) then heated under various temperature that were 300, 350, 400, 450 and 500°C for 5 hours. The pyrolysis process was performed in order to separate components by using indirect heating (no direct contact with fire) and held until it produced 3 fractions that were charcoal (solid-fraction), tar (liquid-fraction) and smoke and methane gas (gas-fraction or light-fraction).

Smoke, the light-fraction of pyrolysis, was then condensed into liquid, the methane gas was kept remain in gas phase. The liquefied smoke as the condensation yield was considered to be third-grade which was still harmful for food preservative uses due its toxic content.

#### Materials

Coconut shell waste obtained from several places near Malang, served as raw materials of this research after being cut into smaller sizes.

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**Experimental Apparatus**

The stainless steel reactor that has completed with condenser and tar-collector was specifically designed to conduct the experiment and to manufacture pyrolyzed product in three-kind of different phase which were solid, liquid and gas through burning process of coconut shell.

**Characteristic**

Standard methods involves yield determination, pH measurement and total phenol content calculation was conducted to characterize the obtained liquid smoke.

**Yield**

Dark glass vessel filled with liquid smoke was accurately weighed. Yield was then determined based on %wt equation.

$$\text{Yield}(\% \text{ w / w}) = \frac{\text{weight of liquid smoke}}{\text{weight of raw materials}} \times 100\%$$

**pH values**

pH value had measured using "Hanna" digital pH meter at which the electrode needed to be submerged first in distilled water before measuring continued with wiping for removing water and finally, the monitor screen would display the result.

**Total Phenol Content**

Major component that had found in liquid smoke is phenolic compound, hence, it was certainly required an analysis of total phenol content. The analysis was done, firstly, by vaporizing the liquid smoke then weighing it at 0.5 grams followed by placing it into a 250-mililitres-volumetric-flask and adding 30 mililitres of distilled water. After all the solution had transferred, it was continued with adding 5 mililitres 0.2 N sodium hydroxide solution and distilled water until the bottom of the miniscus reached the line. Secondly, 25 mililitres amount of the solution was drawn up using pipette then being put into 500-mililitres-erlenmeyer. Thirdly, 25 mililitres of 0.2N chromate bromide solution, 50 mililitres of distilled water and 5 mililitres of concentrated hydrochloric acid were added then it was capped and shaken for 1 minute to homogenize the mixture. One-minute-shaking was continued after additional of 5 mililitres of 15% sodium iodide solution. Finally, homogenized mixture was titrated with 0.1N  $\text{Na}_2\text{S}_2\text{O}_3$  using 1% amylum solution as an indicator. Similar procedures had repeated for blank sample.

$$\text{Total of Phenol (\%)} = \frac{\{(BL - C) \times N - \text{tio} \times BMf / 6 \times 1000\}}{0,1 \times \text{weight of sample}}$$

Description:

BL =Volume blanko

C =Volume of sample

N-tio =Normality of thiosulfate

BMF =molecular weight phenol

**Data Analysis**

Chemical components of liquid smoke performed in GC-MS using HP Ultra 2 column and helium as carrier gas with flow rate 0.6 mL/min, injection volume 1ul, oven temperature at 280°C/10 minutes, injection temperature at 250°C and interface temperature at 280°C.

**RESULTS AND DISCUSSION****Process Characterization**

The temperatures indicated an elevating trend almost in each pyrolysis process in the experiment. Furthermore, colorless smoke had resulted at the beginning process of pyrolysis then it gradually started becomes reddish brown.

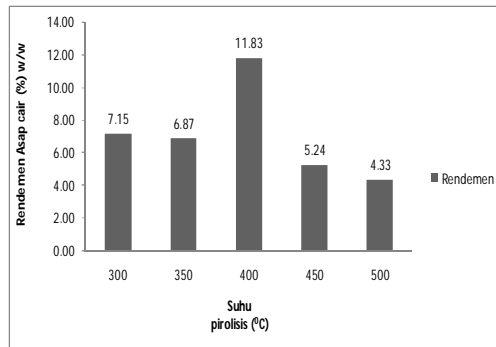
**Yield of Liquid Smoke**

Yield is one of the most important parameters to determine the process outcome. Liquid smoke in this study produced by the condensation of smoke released pyrolysis reactor. During the pyrolysis process that occurred evaporation process of various chemical compounds. Table 1 presents characteristics of liquid smoke as condensation product of coconut shell pyrolysis at temperature process ranged from 300°C - 500°C. In addition, determined yield of liquefied smoke at those temperatures varied from 5.24% -11.83% which is influenced by

components present in raw materials and condensation procedures. Furthermore, similar results were also obtained in the same subject of research that was carried over by Tranggono, et.al., 1996 in which water had needed as cooling medium in the condensation process due its fast exchange of heat.

**Table 1.Characterization of liquid smoke from coconut shell**

| Type of sample | Pirolisis temperature | liquid smoke  |           |
|----------------|-----------------------|---------------|-----------|
|                |                       | Yield (% w/w) | colour    |
| Coconut shell  | 300°C                 | 7,15          | Red brown |
|                | 350°C                 | 6,87          | Red brown |
|                | 400°C                 | 11,83         | Red brown |
|                | 450°C                 | 5,24          | Red brown |
|                | 500° C                | 4,33          | Red brown |



**Picture 1. The relationship between pirolisis temperature and yield of liquid smoke**

The literature studies show the effect of high temperature and long residence time on product yields of coconut shell pyrolysis. Furthermore, the formation of liquid smoke, as the yield of pyrolysis, would decreased due to temperature elevation of water cooling system. Pyrolysis at temperatures that are too high and too long will cause the formation of liquid smoke is reduced as the temperature increases in the cooling water so that the smoke generated is not completely condensed. The process will take optimal condensation if the water in the cooling system flushed continuously, so the temperature in the system is not increased. Nevertheless, the condensation would obtained optimum and perfect level by avoiding temperature increasing through continuous water flow in condenser (Demirbas, 2005).

**Quality of Liquid Smoke**

Chemical components of liquified smoke determine its grade that associate with pyrolysis condition and raw materials used. Chemical compounds contained in liquid smoke heavily dependent on pyrolysis conditions and materials used (Nakai et al., 2006).

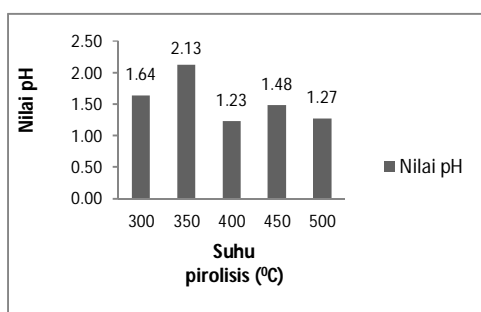
Meanwhile, imperfect pyrolysis delivers evidential impact on incomplete components that build the product. Major compounds of liquid smoke that had shown by identification of the product were dominated with phenolics, carbonils, carboxylic acids, furans, hydrocarbons, alcohols and lactons (Girard, 1992).

**pH Value Measurement**

PH value is one of the quality parameters of the liquid smoke produced. Decomposition process that occurred in pyrolysis had investigated by measuring its pH value. Table 2 highlights the measured pH of liquid smoke at various temperatures.

**Table 2.pH Value of liquid smoke from coconut shell**

| Type of sample | Pirolisis temperature | pH value |
|----------------|-----------------------|----------|
| Coconut shell  | 300°C                 | 1,64     |
|                | 350°C                 | 2,13     |
|                | 400°C                 | 1,23     |
|                | 450°C                 | 1,48     |
|                | 500° C                | 1,27     |



**Picture2. The relationship between pirolysis temperature and pH value**

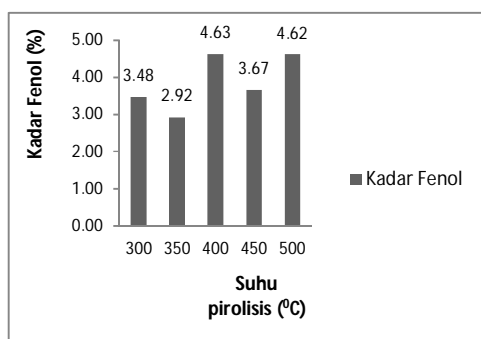
In general, pH of liquid smoke on each temperature are acidic due more complete the decomposition as the temperature increase. Significantly, the lowest pH was reached at 400°C giving value at 1.23. This acidic condition is prominent in liquid smoke and play a major contribution to food preservation activity that affect to its storage time and flavour. Therefore, phenolic compounds and acids in liquid smoke generate an ability of inhibiting the bacteria growth and acting as an antioxidant (Pszczola, D.C., 1995).

#### **Total Phenol Content**

Identification of phenolic components was expected to indicate the quality of the pyrolysis product due its future function. Results were shown on table 3 in which total phenol content ranged from 2.92 - 4.63%. Determined phenol content reached the highest and the lowest level , respectively, at 400°C and 350°C whereas it has no positive correlation with %wt of yield. Furthermore, chemicals compounds of liquid smoke is definitely affected by chemicals content that present in raw material and the temperature of pyrolysis (Djatkiko, 1986).

**Table 3. Quality of phenol total in liquid smoke**

| Kind of sample | Pirolisis temperature | Quality of phenol (%) |
|----------------|-----------------------|-----------------------|
| Coconut shell  | 300°C                 | 3,48                  |
|                | 350°C                 | 2,92                  |
|                | 400°C                 | 4,63                  |
|                | 450°C                 | 3,67                  |
|                | 500° C                | 4,62                  |



**Picture 3. The relationship between pirolysis temperature and quality of phenol**

Pyrolysis occurs in several steps of mechanism at elevated temperatures in which consist of water vaporation at 100°C - 150°C, hemicellulose decomposition at 200°C and followed by cellulose and lignin decomposition into piroglinate, carbon monoxide gas, carbon dioxide gas and tar. In addition, the decomposition of cellulose takes place further at 240°C - 400°C produce greater amount of tar, at above 400°C, converting aromatic layer to piroglinat, carbon monoxide gas, carbon dioxide gas and tar starts to happen (Byrne and Nagle, 1997).

Nevertheless, the experiments provided similar result to Darmadji (1995) specifically for total phenol content measurement. Hence, Demirbas (2005), had successfully identified two different kinds of phenolic compound in liquid smoke which were 2,6-dimethoxyphenol and 3-methyl-2,6-dimethoxyphenol, respectively,

giving %wt of 0.74% and 0.62%. Therefore, Tranggono, et.al. (1997), had also investigated the existence of five kinds of phenolic group that resulted from pyrolysis of wood at 350oC -400oC namely 2-methoxyphenol, 4-methyl-2-methoxyphenol, 4-ethyl-2-methoxyphenol, 2,6-dimethoxyphenol and 2,5-dimethoxyphenol. The increase in total phenol content of pyrolysis product leads to the decrease in pH value which means the liquid smoke, as the product, becomes more acidic.

#### ***Components of Liquid Smoke***

The presence of phenolics in liquid smoke was analyzed using gas chromatography-mass spectrometer (GCMS) by subjecting the samples in methanol solvent. Furthermore, plotted chromatogram and mass spectral data on chemstation data system delivered five until six components in liquid smoke at each varied temperatures with total phenol ranged from 2.48% until 23.40%, as shown on the chromatogram data.

### **CONCLUSION**

Generally, it was resulted the reddish brown liquid smoke with % weight of yield varied from 5.24% until 11.83% whereas the highest was gained at 400°C giving value at 11.83%. In addition, the most acidic liquid smoke was produced at 400°C with pH value 1.23 among pH range of yielded liquid smoke from 1.23 until 2.13 and the highest total phenol content was reached at the same varied temperature, 400°C, which was 4.63%. In conclusion, the higher the total phenol content in liquid smoke the more acidic its product.

### **ACKNOWLEDGEMENT**

The research indeed needs continuous purification with optimum temperature that had investigated using the exact raw materials to convert its grade from third grade to second or first grade.

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