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AN ENVIRONMENTAL FRIENDLY PESTICIDE FROM BINTARO (*Cerbera odollam* GAERTN) LIQUID SMOKE FOR PINE WOOD PRESERVATION AGAINST A SUBTERRANEAN TERMITE *Captotermes curvignathus* HOLMGREN ATTACK

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ABSTRACT

This study investigated the possibility of using liquid smoke from the waste of pyrolysis *Cerbera odollam* Gaertn fruit as an environmental friendly pesticide to prevent pine wood from a subterranean termite *Captotermes curvignathus* Holmgren attack. The liquid smoke was made from three kinds methods of pyrolysis temperature, 300ºC, 400ºC, 500ºC. Liquid smoke characterized by determining the yield, specific gravity, pH, acidy and phenol content. Compounds of liquid smoke were identified using Gas Chromatography and Mass Spectrometry. Material durability test against subterranean termites *C. curvignathus* Holmgren used Japan International Standard K-1571-2004. The results showed that the higher the temperature pyrolysis, yield, specific gravity, ph. Acid, and phenol content from liquid smoke also increases and liquid smoke with pyrolysis temperature 500ºC caused 100% mortality of termites. Most compounds in liquid smoke are phenol, phenol 2 methoxy, phenol 2-methoxy, mequinol, phenol 2 methyl, phenol 4 methyl, phenol 2 methoxy 4 methyl,9-octadecenoid acid (2), 2,3 dihydroxypropyl ester. The liquid smoke from waste of pyrolysis *Cerbera odollam* Gaert fruit is potential as environmentally friendly pesticide

Keywords: *Cerbera odollam* Gaert fruit; liquid smoke; *Captotermes curvignathus* Holmgren; environmentally friendly pesticide.

INTRODUCTION

Supply solid wood with high quality and durable is so limited that people started to switch using durable low-grade wood such as pine (Pinus) as a raw material. However, because the pine wood has a low level of durable, wood susceptible to termite attack, especially from families Rhinotermitidae types *Captotermes curvignathus* Holmgren[1]. This time to overcome the termite attack, people using artificial chemical containing toxic substance such as fipronil. The use of plant extracts such as *Nootkaton*[2], *Protiumplant javanicum Burm*[3], has been reported to have repellent and toxic effects against termites and be a promising alternative for pest control in the future. Therefore, researchers are looking for anti-termite which is more environmentally friendly, safe, reliable application, to prevent the destruction of wood and economical in terms of cost[4].

Cerbera odollam Gaertn or bintaro derived from poisonous family Apocynaceae found in coastal swamps, rivers and streams in many Asian countries[5].Studies of Gailard *et al*[5] and Ahmed[6] showed that bintaro contain toxic compounds that are used as an insecticide. Its seeds contain a toxic and lethal potent compound called glycosides cerberin.

Bintaro fruit pyrolysis produces charcoal and smoke. Liquid smoke condensation products of combustion smoke containing lignocellulosic feedstock to produce a variety of complex chemical compounds. This chemical compound consisting of phenolic, carbonyl derivatives and acetic acid[7]. Some scientists are

already researching the bintaro fruit seeds as diesel oil[8] and forget about the waste in the form of layers mesocarp and fruit epicarp bintaro. The aim of this study is to investigate the characteristics of liquid smoke fruit bintaro pyrolysis temperature to 300ºC, 400ºC and 500ºC. The second objective, identify the compounds contained in the liquid smoke at different pyrolysis temperatures and the third determines the effectiveness of liquid smoke as a preservative pine wood from termite attack and thus potentially as environmentally friendly pesticides.

EXPERIMENTAL

Before being burned, separated parts epicarp and mesocarp *C.odollam* Gaertn from seed. Parts of *C.odollam* Gaertn cut to a size of about 5-6cm and dried in the sun. These pieces, then put in the kiln reactor. The reactor is heated at high temperatures (100-500ºC). Pyrolysis temperature is set at 300ºC, 400ºC, and 500ºC. After reaching the desired pyrolysis temperature, charcoal was issued. The smoke that came out during the pyrolysis process is condensed and collected in a flask. Furthermore, liquid smoke is purified with distillation, and then chemical characteristic is measured as pH, specific gravity (spgr), acidity, phenol content and yield calculated as % w/w. The chemical compounds of liquid smoke are identified by using Gas Chromatography and Mass Spectrometry (GC-MS). For anti-termite test, pine wood dipped in liquid smoke made at a temperature of 300ºC, 400ºC and 500ºC. Each week termite activity in the bottle is observed, recorded and stored in the dark at a temperature of 28 ± 2°C for 3 weeks. After 3 weeks of the test, containers are opened and mortality calculation of termites. Furthermore, after the observation on termites, wood samples were washed, oven for 48 hours at a temperature of 60 ± 2°C and weighed to determine the weight loss of wood. In addition there is the observation of durable grade wood and termite mortality[9].

RESULTS AND DISCUSSION

Yield, from liquid smoke of the pyrolysis temperature at 300ºC, 400°C, and 500°C are shown in figure 1. The results of the liquid smoke with a temperature pyrolysis 300ºC, 400ºC, and 500°C respectively are 40.143%; 40.857% and 41.429%. This means that the result of lower liquid smoke obtained at a lower temperature pyrolysis. Oramahi and Diba[10]say that the result of lower liquid smoke at lower temperatures due to the temperature rise is not enough for the complete pyrolysis to occur so that not all of the celluloses convert into solution pirolignat and gas CO, CH_4 and H_2. Conversely, at higher temperatures secondary decomposition reactions may occur. At a temperature of 500ºC hemicellulose, cellulose and lignin decomposes thoroughly. Lignin at a temperature of 500ºC to decompose completely and form a layer of aromatic and tar[11] which will be mixed with a liquid smoke resulting greater yield at temperatures 500ºC than temperature of 300ºC and 400ºC.

Charcoal burning also affects yield results. This happened because the process does not occur due to imperfect pyrolysis kiln, so that pyrolysis cannot be optimally maintain the pyrolysis process. The amount of oxygen is still abundant in the burning drum that causes yield results smaller at lower temperatures[12]. According Guillen and Ibargoita[13]the decrease yield of liquid smoke because there is charcoal, tar and compounds that are not condensed well. Based on figure 1, it is known that the pH value will increase with increasing temperature. pH value of liquid smoke is about 2.28 to 2.78. Sung *et al.*[14] said that the acidity of the liquid smoke depends on the source of the wood, the steps of processing and refining parameters. The acidity of the liquid smoke also affected by temperature, because at temperatures higher than 280°C, acetic acid production is well done.

The pH of the liquid smoke or wood vinegar bintaro fruit of 2.28-2.78 indicates that wood vinegar bintaro fruit is still in a state of acid in the standard range of Japanese wood vinegar (1.5-3.5) [15]. The lowest pH value was achieved in liquid smoke at temperatures of 300ºC because decomposition cellulose compound occurs into acetic acid and homologues, which causes the pH at a temperature of 300°C is very low. Similarly, at a temperature of 400ºC, the pH value is also still acid because at this temperature, decomposition lignin into phenol occurs, homologues and derivatives that are acidic. At the temperature of 500ºC, the pH value is also acidic because partial decomposition lignin of liquid smoke at this temperature is still phenol and derivatives. Various phenolic compounds present in the liquid smoke with low pH[15].

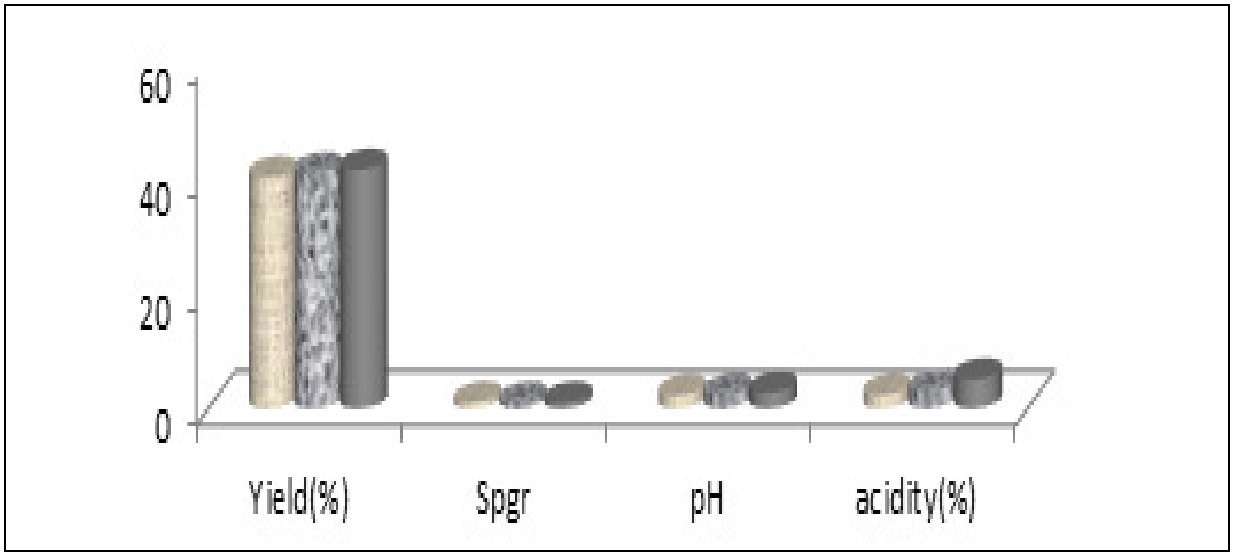

Fig.-1: The Results of the Analysis of Liquid Smoke at a Different Temperature Pyrolysis

Based on the analysis, the level of acidity of the liquid smoke bintaro fruit is in the range of 2.35 to 5.062%, from figure 1, it appears that the higher the pyrolysis temperature of bintaro fruit the higher the percentage of acidity in liquid smoke. The increasing in the percentage of acidity of wood vinegar is caused by the removal and leaching of organic acids from wood[16], beside that pyrolysis will cause more and more unraveling hemicelluloses and cellulose into chemicals that are acidic primarily acetic acid and homologues[17].

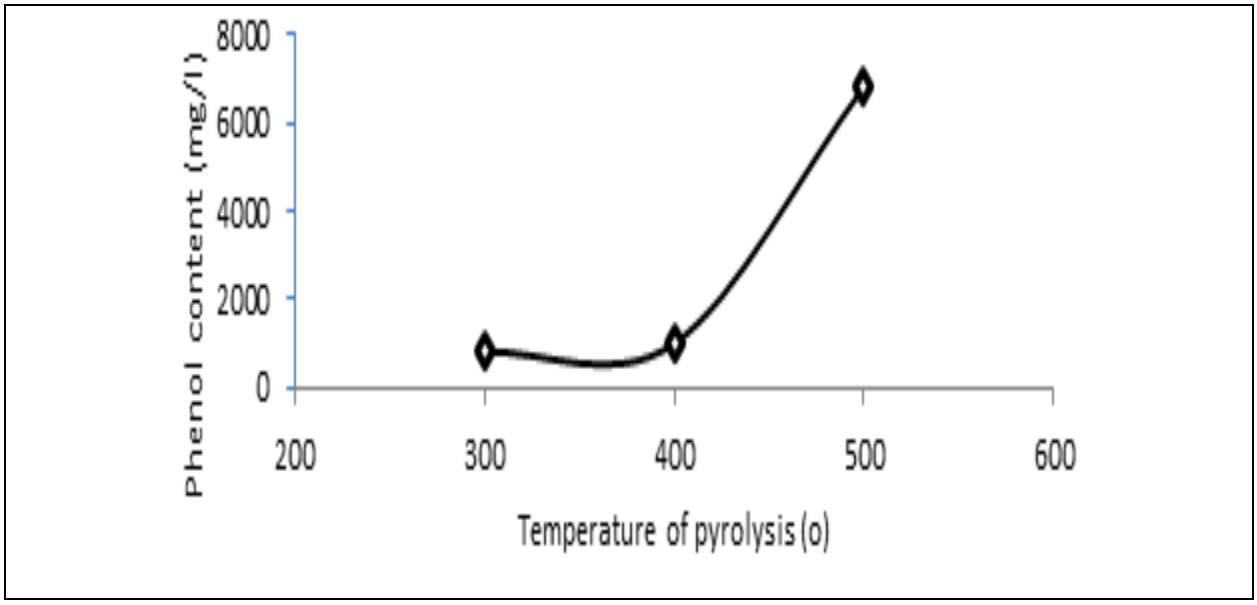

Fig.-2: Phenol content of liquid smoke at different temperature of pyrolysis

Figure-2 and table-1 show that the increase in temperature will increase the yield of pyrolysis liquid smoke and produce higher contents of phenols. Phenol content of liquid smoke with a temperature

pyrolysis 300ºC, 400ºC and 500ºC is 813.0081 mg / L; 1016 mg / L and 6,833.33 mg / L respectively, with a dilution factor of 2500 times.

Table -1: Chemical compound of liquid smoke in different temperature of pyrolysis

S. No.	Compound	Area (%)		
		300ºC	400ºC	500ºC
1.	2-Propanone, 1 hydroxyl	4.66	-	-
2.	Furfural; pyrazole, 1,4- dimethyl	4.71	-	-
3.	8 – Azabicyclo[3.2.1] octan -3-one	0.16	-	-
4.	1.6;2,3-Dianhydro-4-O-acetyl	0.72	-	-
5.	4,4-Dimethyl-2-cyclopenten-1-one	2.32	-	-
6.	Pyridine-3-carboxamide, Oxime, N-(2-trifluoromethylphenyl)	1.35	-	-
7.	Butyrolactone	0.64	-	-
8.	2-Furan Carboxaldehyde, 5-methyl	0.54	-	-
9.	2 Cyclopenten-1-one, 3 methyl	0.24	-	-
10.	2 Cyclopenten-1-one, 2,3 dimethyl	0.96	-	-
11.	2 Cyclopenten-1-one-2-methyl	-	2.17	-
12.	2-(1-methyl vinyl) thiophene	0.44	-	-
13.	Cyclohexan-1,4,5-triol-3-one-1-carboxylic acid	0.1	-	-
14.	3-Allyl-6-methoxyphenol	0.28	0.47	-
15.	Benzene, 1-methyl-4-(phenylmethyl)	0.1	-	-
16.	13-Tetradecen-1-ol-acetate	0.51	-	-
17.	9-Octadecenoic acid	0.56	-	-
18.	Propanoic acid	1.49	-	-
19.	Oleic Acid	2.22	-	-
20.	Acetic Acid	46.09	3.30	13.09
21.	Butanedioic acid, 2,3-bis(acetyloxy)	-	0.85	-
22.	2,4-Hexadien-1-ol; Cyanamide, dimethyl	-	0.63	-
23.	Furan, 2,4 - dimethyl	-	2.17	0.92
24.	Phenol	2.65	5.28	4.08
25.	Phenol, 2-methoxy;	12.43	15.19	18.18
26.	Phenol, 2,6-dimethoxy	0.65	-	-
27.	Mequinol	-	15.19	-
28.	Phenol, 3-methyl; Phenol, 4-methyl	-	5.06	3.05
29.	Phenol, 2-methoxy-4 methyl	2.73	7.28	9.36
30.	Phenol, 2-methoxy-4- (1-Propenyl);	-	0.47	1.15
31.	Phenol, 2,3 dimethyl	-	2,47	-
32.	Phenol, 4-ethyl-2-methoxy	1.23	4.25	4.30
33.	Phenol, 2,4,6-trimethyl	-	0.83	0.53
34.	3 Fluorobenzaldehydesemicarbazone	-	1.6	-
35.	5-tert-Butylpyrogallol	-	0.45	-
36.	9-octadecenoic acid (Z), 2,3 dihydroxypropyl ester	-	42.57	23.76
37.	1-H-Imidazole, 1,5-dimethyl; 1-H-imidazole, 1,4 dimethyl	-	-	0.28
38.	1-H-pyrazole, 3,5-dimethyl	-	-	0.31
39.	Hept-3-yn-2-one	-	-	1.28
40.	Napthalene, 1,1-ethylidene bis(decahydro); 2,4-Dihydroxypropiohenone	-	-	1.15

This happens because the chemical compound of liquid smoke or vinegar is influenced by the type of feedstock, pyrolysis time and temperature pyrolysis. Based on Guillen and Ibargoita[13], pyrolysis

temperature is one of the important parameters to achieve the best quality of liquid smoke. Pyrolysis process involves a variety of reactions, namely decomposition, oxidation, polymerization, and condensation. Yaman[18] says that the process of thermal degradation of cellulose to produce glucose then the first phase of the second stage produces acetic acid and homologues, water and small amounts of furan and phenolic compounds. This causes the phenol contents are not too big from pyrolysis temperature of 300ºC.

From table-1, we can see that guanicol (2 methoxy phenol) is the most abundant compound found in this liquid smoke which is consider when a soft wood is used as a source wood. Therefore syringol(2, 6-dimethoxy phenol) is only found about 0.15% at temperature pyrolysis of 300ºC. Guillén and Manzanos[19] said that when soft wood used as source, syringol is barely found in liquid smoke. At a temperature of 400ºC found chemical compounds such as phenol and homologues, furan and a little carboxylic acid while at a temperature of 500ºC detected most of the phenolic compounds and polycyclic aromatic hydrocarbon. Liquid smoke contain acid compounds show antibacterial activity of phenolic compounds[20]. Result of identification GC-MS gives similar results with the measured phenol contents of liquid smoke, wherein at pyrolysis temperature of 400ºC, phenol content less than that at 500ºC. The naphthalene compound at a temperature of 500°C which include in liquid smoke signifies the PAH group is already dangerous for the environment and humans[21].The data from table 1 also indicate that the volatile organic compounds such as acetic acid, 9-octadecenoic acid phenolic compounds and carboxylic formed by pyrolysis in the carbonization process.

Wood weight loss and mortality at different temperatures of pyrolysis and liquid smoke concentration are summarized in table 2. At temperature of 300ºC termite death occurred due to the temperature of liquid smoke contains large acetic acid (46.09%), which serves as an agent termicidal[21], while at temperature of 400ºC which act as antibacterial are phenolic compounds and derivatives that caused the termite becomes dead, phenolic components like phenol, 2-methoxy phenol, 4-ethyl-2-methoxy considered to act as a biocidal agent may be responsible for the antifungal properties, termicidalof liquid smoke [15,21].

Table-2: Termites Mortality and Wood Weight Loss of Efficacy Test of liquid smoke

Treatment	Mortality (%)	Wood Weight loss (%)	Grade durable wood
Control	16.2233333	11.61	IV
300ºC	97.5566667	2.56333	I
400ºC	96.4433333	2.21667	I
500ºC	100	2.33	I

Pinewood weight loss on termite test was 11% for the control, it indicates that the control has the highest percentage of weight loss of wood, meaning that the resistance class pine wood without dipped in liquid smoke is lowest so that pine is very vulnerable to attack by termites. but different conditions found in pine wooddipped in liquid smoke, which have wood weight loss percentage of (2.21 to 2.56%). This means that durability of wood can change if the pine wood dipped into liquid smoke.

CONCLUSION

The higher the temperature pyrolysis liquid smoke causes more yield generated, the greater levels of specific gravity, pH, acidity and phenol content. GC-MS analysis of the liquid smoke from pine wood showed that the components of liquid smoke predominat acetic acid; Phenol, 2-methoxy; Phenol, 2-methoxy-4 methyl; 9-Octadecenoid acid(Z), 2.3dihydroxyprophyl ester.

Based on the anti-termite test results the death of termites, liquid smoke from the pyrolysis temperature of 300ºC, 400ºC and 500ºC cause termites death are 96.44%; 97.56% and 100% respectively. Among the pyrolysis temperatures of liquid smoke, no significant difference as anti-termite but with controls(pine) there is significant difference. This means that liquid smoke from three temperature pyrolysis can be used as an anti-termite. Based on the wood weight loss can be concluded that the pine wood dipped in a solution of liquid smoke can increase the grade durable wood of class IV(control) to the class I.

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