Application of Liquid Smoke from Rubber Wood Clone PB-340 as Latex Coagulant and Preservation of Natural Rubber Coagulum

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ABSTRACT

The utilization of rubber wood waste in the form of liquid smoke has the potential to overcome environmental problems caused by the industry. Therefore, this study aimed to determine the potential of liquid smoke from rubberwood clone PB 340 to be used as a coagulant and preservative. Rubber wood waste was processed into liquid smoke using the pyrolysis method with a temperature of 400°C. Determination of the composition of liquid smoke was carried out using gas chromatography-mass spectrometry (GC-MS) analysis. The results showed that the rubber wood clone PB 340 contained 57.78% cellulose, 12.16% hemicellulose, and 19.01% lignin. Furthermore, volatile analysis with GC-MS showed that liquid smoke from rubber wood clone PB 340 contained 58 organic compounds. Some compounds in liquid smoke were phenols, furans, furfurals, acetic acid, and cyclopentene. The product was then tested for its performance as a latex coagulant at several concentrations of 5%, 10%, 15%, 20%, and 25% v/v, as well as storage time of 1, 7, and 14 days. The treated latex samples that had turned into coagulums were examined for their sheet quality parameters, including initial plasticity (P0), plasticity retention index (PRI), total volatile compounds, impurity, and ash content. The outcomes from all comparisons of pure liquid smoke concentrations and storage time of up to 14 days of the coagulum samples showed that in the initial plasticity value (P0), the plasticity retention index (PRI), volatile matter, ash content, and dirt content had met the applied Standard Indonesian Rubber (SIR).

Keywords: Coagulant; latex; liquid smoke; rubber wood; rubber coagulum

INTRODUCTION

Indonesia has extensive rubber plantations, which is important to the nation's economy. In 2019, the natural rubber production was 3.33 million tons, with a plantation area of one million hectares. North Sumatra is one of Indonesia's provinces with natural rubber plantations covering an area of 369,391 hectares. The province can produce up to 309,973 tons of natural rubber (BPS, 2019). Even as strategic commodities, farmers faced problems regarding rubber wood waste, which should be processed or converted into more useful products with high added value.

Nancy et al, (2013) stated that only about 18% of rubber wood was utilized or reused by farmers and industry in replanting. Rubber wood serves as raw material for the carpentry, fiber board, and plywood industry. Most of the rubber wood waste, the remaining

DOI: http://doi.org/10.22146/agritech.70487 ISSN 0216-0455 (Print), ISSN 2527-3825 (Online) 82%, had not been used optimally. Therefore, the remaining is burned in its place or left to rot and occupy the spaces in the plantation area. Under the circular economy principle, one of the utilization options is to convert rubber wood into liquid smoke through the pyrolysis process. In addition to reducing environmental problems, producing liquid smoke can increase their economic value. Prasetyowati et al. (2014) stated that the smoke produced could be used as a coagulation agent and reduce unwanted odors from resulting products.

Solichin (2007b) and (2007a) stated that pure and commercial liquid smokes, such as Deorub Super (DS) and Deorub K, are alternative coagulants in rubber processing. However, using formic acid, which contains one carboxylic group, is not recommended because it can irritate human skin and the environment. Nonirritant and environmentally friendly rubber coagulants, such as liquid smoke, should be considered.

Prasetyowati et al. (2014) showed that liquid smoke contains high acetic acid and phenolic compounds. Prasertsit et al. (2011) proved that due to the phenolic compounds and acetic acid in the liquid smoke, the quality of the sheet rubber's physical properties could be improved and prevent mold growth. Previous research by Vachlepi (2020) showed that the products from coconut shells, known as deorub super (DS) and deorub K with a dilution concentration of up to 5% and formic acid, could be used as coagulants.

The composition of the originating wood material would affect the resultant liquid smoke and its applications. Therefore, this study investigated the liquid smoke produced from the local rubber tree clone in Deli Serdang, North Sumatra, Indonesia, as a latex coagulant. Meanwhile, no study has evaluated the potency from rubber wood clone PB 340 as a coagulation agent and coagulum preservative.

MATERIALS AND METHODS

Materials

This study used rubber latex from PTPN III Gunung Para Plantation and rubberwood from the PB-340 tree clone of Deli Serdang Smallholder's Plantation, North Sumatra, Indonesia, with formic acid from PT.BASF-YPC, China.

Chemical Component Analysis of Rubber Wood

Chemical components analyzed include water content using the AOAC method (1990), cellulose, hemicellulose, and lignin content using the fractionation procedure according to Cheeson (1978) in Datta (1981).

Liquid Smoke Production

A total of 3 kg of rubber wood pieces with dimensions of 3 cm x 3 cm x 3 cm were placed in a pyrolysis reactor equipped with a refrigerated condenser circuit. The pyrolysis process was carried out for 90 minutes at 400 °C and was stopped when no more liquid smoke dripped into the tank (Darmadji et al., 2000). The total time required was approximately 3 hours, and the smoke produced from this combustion flew through the condenser circuit. It converted the smoke from the gas to the liquid phase by circulating through the condenser pipe equipped with cooling water. The liquid smoke formed was accommodated in an Erlenmeyer flask placed at the bottom of the condenser, and was distilled at a temperature of 120 °°.

Chemical Analysis of Liquid Smoke

The chemical composition of crude liquid smoke includes total phenolic content (Senter et al., 1989), carbonyl (Lappin & Clark, 1951), and acidity (AOAC, 1990) determination by titration method. Furthermore, compounds were identified using gas chromatographymass spectrometry (GC-MS).

Identify The Compound Components

Chemical composition in liquid smoke was analyzed using GC equipped with a mass spectrometer as a detector at LPPT Gadjah Mada University. A total of 0.2 mL of the smoke was dissolved in 1 mL of methanol in a GC vial and vortexed until homogeneous, and the injector temperature was 260°C. The column was operated at a column temperature of 60°C, carrier gas was Helium with a pressure of 12 KPa, total flow 25 mL/m, column flow 0.51 mL/m, linear acceleration 26 cm/s, cleaning flow 3.0 mL/m, ratio split 1:50, column type Agilent DB-1, column length 30 m, diameter 0.25 mm, and ionization type EI (Electron Impact) 70 eV. Compounds in liquid smoke can be determined by comparing the results of GC-MS with standard compounds.

Sample Performance on Latex Coagulation

The liquid smoke obtained from the pyrolysis results was used to coagulate latex with a dry rubber content of 44%. The factor studied was the effect of the concentration of liquid smoke and storage time. Furthermore, 1,000 mL of latex was inserted into the coagulation duct with the treatment concentration of pure liquid smoke at 5%, 10%, 15%, 20%, and 25% (%v/v), while formic acid with a concentration of 5% was used as a control. Coagulum was stored for 1, 7, and 14 days, and each rubber coagulum was sheated

using a creeper to facilitate drying and further analysis. Natural rubber sheets were dried using a hot air oven at a temperature of \pm 105 °C for 4–5 hours until constant weight. Each sample was tested according to ISO for five physical properties, including initial plasticity (ISO 2930), plasticity retention index (ISO 2930), dirt content (ISO 249), ash content (ISO 247), and volatile matter content (ISO 248).

Statistical Analysis

This study was designed using a completely randomized design (CRD) consisting of two factors, namely the comparison of liquid smoke treatments at 5%, 10%, 15%, 20%, and 25%, and control with a storage time of 1, 7, and 14 days. The research data were analyzed using analysis of variance (ANOVA) with IBM Statistical Product and Service Solutions (SPSS) version 19 for Windows. Variations between samples were determined using the smallest significant difference test at p< 0.05. Furthermore, when F count > F table, the Duncan Multiple Range Test (DMRT) is carried out to determine the significant difference between liquid smoke and control ratio treatments.

Chemical Components of Clone PB 340 Rubber Wood

The analysis of the chemical components showed that the lignin content of rubberwood clone PB 340 could be seen in Table 1, which was 19.01%. It was higher than the lignin content of rubberwood clone GT-1 in the study by (Darmadji et al., 2000), which was 16.69%. Meanwhile, the cellulose content of clone PB 340 was 57.77%. It was also higher than the clone of GT-1 at 45.67% (Darmadji et al., 2000). This chemical composition is due to the variation in rubber tree cultivars used, and Razar & Aris (2012) showed that each type of rubberwood's lignin content and cellulose differ.

Lingbeck et al. (2014) suggested that the pyrolysis of lignin will produce phenolic compounds that affect the smoke aroma. The results of the pyrolysis of cellulose in the rubberwood will determine the acid, furan, and phenolic levels. Hemicellulose can

Table 1. Cellulose, hemicellulose, and lignin composition of rubberwood clone PB 340

Components	Amount (%)
Cellulose	57.77 ± 0.06
Hemicellulose	12.15 ± 0.03
Lignin	19.01 ± 0.11

affect the furfural levels of furan, carboxylic acid, and acetic acid. Therefore, the amount in rubberwood is directly proportional to acid, lignin, and phenolic levels in liquid smoke.

Chemical Composition of Rubber Wood Liquid Smoke

The results showed that the liquid smoke from rubber wood clone PB 340, which was pyrolyzed at 400 °C, contained 2.16% phenolic compounds, 5.43% carbonyls, 16.17% organic acids as indicated in Table 2, and a pH of 2.55 as seen in Table 3.

Results of volatile analysis with GC-MS indicated that liquid smoke contains 58 organic compounds, as shown in Table 3. The chemical components, which have the largest percentage were furfural, 2-methoxyphenol, 2-furanethanol, creosol, 2-methyl-2-cyclopenten-1-one, 1-(2-furanyl)-ethanol at 24.53%, 11.81%, 6.37%, 4.01%, 4.61%, and 3.81%, respectively. Based on the table above, the antimicrobial compounds are 2-methoxyphenol, 4-ethyl-2-methoxyphenol, and creosol.

Coagulation Time

The coagulation period is required to convert liquid natural rubber latex into coagulum by adding a coagulation agent, and the results of the average coagulation time are presented in Figure 1.



Figure 1. Coagulation time of the coagulum sample treated with several ratios of liquid smoke.

Table 2. Phenolics, carbonyls, and organic acids composition
of liquid smoke from rubberwood clone PB 340

Components	Amount (%)
Phenolics	2.15 ± 0.37
Carbonyls	5.42 ± 0.15
Organic acids	16.17 ± 3.49

No	% Relative content	Molecular formula	Compound name
1	0.02	C ₆ H ₁₂ O ₅	rhamnose
2	24.53	$C_5H_4O_2$	furfural
3	1.34	$C_5H_9NO_4$	2-acetylamino-3-hydroxy-propionic acid
4	0.61	$C_6H_8O_2$	2-(methoxymethyl)furan,
5	1.88	$C_6H_{10}O$	2-methylcyclopentanone
6	1.18	$C_8H_{18}O_4$	3-methoxy-2,2-bis(methoxymethyl)- 1-propanol,
7	1.20	$C_{11}H_{20}O_{2}$	2-hexyl cyclopropane acetic acid
8	0.54	$C_{14}H_{28}O_{3}$	2-hydroxytetradecanoic acid
9	0.06	$C_{6}H_{14}O_{3}$	3-methyl-1,3,5-pentanetriol
10	0.05	$C_7 H_{12} O_2$	2-propenyl butanoate
11	1.69	C8H18O3	3,5-dimethylhexane-1,3,4-triol
12	0.15	C ₁₇ H ₃₂ O ₂	10-methyl-E-11-tridecen-1-ol propionate
13	0.27	$C_{8}H_{14}O_{2}$	9-oxabicyclo[6.1.0]nonan-4-ol
14	2.08	$C_5H_8O_3$	1-(acetyloxy)- 2-propanone
15	0.05	$C_9H_{16}O_2$	4-methylcyclohexanol acetate
16	3.62	$C_{6}H_{12}O_{3}$	tetrahydro-2,5-dimethoxyfuran
17	0.19	$C_{6}H_{10}O_{5}$	6-deoxy-D-mannono-4-lactone
18	0.46	$C_{6}H_{10}O_{2}$	3-hydroxycyclohexanone
19	0.08	$C_{10}H_{21}N_3 S$	10-azido-1-decanethiol
20	0.15	$C_{5}H_{10}O_{5}$	DL-arabinose
21	0.10	$C_9H_{16}O_3$	7-hydroxy-6-methyl-oct-3-enoic acid
22	4.61	C ₆ H ₈ O	2-methyl-2-cyclopenten-1-one
23	3.81	$C_6H_6O_2$	1-(2-furanyl)- ethanone
24	0.04	$C_{10}H_{18}O_{3}$	1,2-cis-1,5-trans-2,5-dihydroxy-4-methyl-1-(1-hydroxy-1-isopropyl)
25	0.10	$C_8H_{14}O_7$	6-acetyl-B-d-mannose
26	0.29	$C_{16}H_{30}O_{2}$	(E)-9-tetradecen-1-ol acetate
27	0.03	$C_{11}H_{14}N_4 O_5$	2-methyl-9-B-d-ribofuranosyl hypoxanthine
28	1.86	$C_7H_{10}O$	3,4-dimethyl-2-cyclopenten-1-one,
29	0.19	$C_{16}H_{36}OSi$	2-dimethyl silyloxy tetradecane
30	9.04	$C_7 H_{10} O_3$	ß-methoxy-(S)- 2-furanethanol
31	6.37	$C_{11}H_{14} N_4O_5$	5-methyl-2-furancarboxaldehyde
32	0.05	$C_7H_{10}O$	2-methyl-2,3-divinyloxirane
33	0.74	$C_6H_6O_3$	methyl 2-furoate
34	0.41	$C_{18}H_{32}O_{2}$	17-octadecynoic acid
35	2.75	$C_7H_{10}O$	2,3-dimethyl-2-cyclopenten-1-one
36	0.42	C_8H_{13} NO ₄	2-(1-buten-3-yl)-2-nitro- acetate
37	0.82	C ₇ H ₁₂ O	1,6-heptadien-4-ol

Table 3. Identification of compounds in liquid smoke by GC-MS

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38	0.80	$C_7H_{13} NO_3$	2-nitrohept-2-en-1-ol
39	0.43	$C_{23}H_{45} N_5O_{14}$	paromomycin
40	0.45	$C_{11}H_{18}O_{2}$	Bicyclo[2.2.1]heptan-2-ol, 7,7-dimethyl-, acetate
41	0.30	$C_{11}H_{20}O_{4}$	4-Hexenal, 6-hydroxy-4-methyl-, dimethyl acetal, acetate, (Z)-
42	2.78	$C_{8}H_{18}O_{2}$	hexanal dimethyl acetal
43	0.29	$C_8H_{10}O_4$	dimethylmuconic acid
44	0.38	$C_{9}H_{16}$	2,2-dimethyl-3-heptyne
45	0.55	$C_{10}H_{16}O_{2}$	5-isopropenyl-2-methyl-7-oxabicyclo[4.1.0]heptan-2-ol
46	0.19	$C_{_{14}}H_{_{19}}\;NO_{_4}$	N-benzyloxycarbonyl-dl-norleucine
47	0.42	C_9H_{16}	(1-methylethylidene)-cyclohexane,
48	11.81	$C_7H_8O_2$	2-methoxyphenol,
49	0.16	$C_8H_8O_3$	4-methoxy phenyl formate
50	1.63	$C_{11}H_{18}O_{2}$	2,6,8-trimethylbicyclo[4.2.0]oct-2-ene-1,8-diol
51	0.65	$C_8 H_{10} O_2$	2-methoxy-6-methylphenol
52	0.29	C ₁₃ H ₂₂	4-(2,6,6-trimethyl-2-cyclohexen-1-yl)-trans-3-buten-2-ol
53	4.01	$C_8 H_{10} O_2$	creosol
54	0.12	$C_{11}H_{19}$ NO	2-(2-isopropenyl-5-methyl-cyclopentyl)-acetamide
55	0.23	$C_{10}H_{16}O$	2,6,6-trimethyl-1-cyclohexene-1-carboxaldehyde
56	0.09	$C_{_{11}}H_{_{19}}$ NO	4-(2,5-dihydro-3-methoxyphenyl)butylamine
57	2.48	$C_9H_{12}O_2$	4-ethyl-2-methoxyphenol
58	0.05	$C_{11}H_{16}O_{2}$	4,4a,5,6,7,8-hexahydro-1-methoxy-2(3H)-naphthalenone

Figure 1 shows that by comparing the short agglomeration period between the 25% liquid smoke dosage and the control, there was a significant difference at 41.33 and 31.33 minutes. At a ratio of 5% of liquid smoke, the time required to coagulate latex is 100.67 minutes. The concentration of formic acid used in this research was 5%. Furthermore, when the concentration of formic acid is used without dilution (97%), the rubber will be difficult to grind, resulting in damaged rubber quality. Chemical compounds in coagulants, especially organic acids, greatly determine the speed of latex clumping (Muthawali, 2016). The added acid compounds can trigger this damage while accelerating coagulate latex.

The higher the concentration of the acid compound, the lower pH, thus, the process of coagulation will be faster. The volume of the coagulant added is directly proportional to the contact area, causing the latex to clot quickly (Ulfah et al., 2017). Formic acid had a shorter coagulation time due to high acid content and low pH



Notes: Different letters indicate statistically significantly different based on Duncan Multiple Range Test (p<0.05)

Figure 2. P_0 of sheeted rubber with various liquid smoke dosages during several storage periods

value (2.6), hence the freezing process would be fast (Sucahyo, 2010), while liquid smoke had a pH of 2.6.

Initial Plasticity (P₀)

Initial plasticity (P_0) predicts rubber polymer chain length or molecular weight (MW). Furthermore, P_0 and

PRI measurements are very important in evaluating the elasticity quality of rubber (Achmadi et al., 2015). The P_0 of the sample treated with various amounts of liquid smoke during several storage days can be seen in Figure 2.

Figure 2 shows that the initial plasticity of the rubber sample treated with liquid smoke was significantly different from the control using formic acid as the coagulant. The P_0 value produced from the control tends to be lower than the sample treated with coagulant from rubber wood liquid smoke. The lowest P_0 value was obtained from rubber coagulated using formic acid with 1-day and 14 days storage at P_0 37 and P_0 40. The lowest and highest values were achieved at a ratio of 15% and 25% of liquid smoke for 1-day and 14 days storage, which was 35 and 42.

The increase in P_0 value was faster under dry storage conditions than wet storage conditions due to the pre-drying treatment. Pre-drying can increase the P_0 value because a storage-hardening process occurs during pre-drying. This causes cross-linking between the aldehyde groups on the rubber polymer chain (Wicaksono, 2001).

Plasticity Retention Index (PRI)

Figure 3 shows that the plasticity retention index (PRI) of the rubber sheet coagulated with liquid smoke of rubber wood significantly differed from the control during the storage period of up to 14 days. The lowest and highest PRI value was obtained at a ratio of 5% and 10%, which was 75 and 90. Treatment using liquid smoke could maintain the PRI value during storage which was still under the SIR category, at a minimum value of 70.



Notes: Different letters indicate statistically significantly different based on Duncan Multiple Range Test (p<0.05)

Figure 3. PRI Sheet from several liquid smoke ratios and storage days

The PRI value is important because it determines

the resistance of rubber molecules to oxidative degradation, such as in the heating process (Aguele et al., 2015). The results indicate that the amount of liquid smoke in the coagulant and storage time affects the PRI value. The liquid smoke ratio is directly proportional to PRI value, which indicates the quality of the rubber sheet produced. Suwardin and Purbaya (2015) explained that the amount of liquid smoke is proportional to the antioxidant content, especially phenolic compounds, and organic acids. Therefore, it can inhibit the activity of spoilage bacteria, act as an antioxidant, protect the rubber molecules at high temperatures and maintain a constant PRI value. It inhibits the growth of mold on rubber products and the oxidation damage of rubber sheets tends to be low.

Several factors affect the PRI value, including the processing of raw materials, such as suitable coagulation materials and storage methods. PRI is affected by nitrogen or protein content in natural rubber (Nun-anan et al., 2018). According to Handayani (2014), various factors that can reduce the PRI value include drying rubber, heating rubber at very high temperatures, and excessive grinding process. The PRI value can be increased by pre-drying the sheet-shaped rubber processing material. The sheet is already halfdried and does not require high-temperature drying (Wicaksono, 2001).

Ash Content

The lowest ash content value was obtained in the comparison treatment of 5% liquid smoke with a storage time of 1 day at 0.21%. In contrast, the highest value is in the ratio of 5%, 15%, and 25%, with a storage period of 14 days at 0.48%. The high ash content meets the crumb rubber quality requirements in SNI 06-1903-2011, namely the ash content of SIR 5 at 0.50%.



Notes: Different letters indicate statistically significantly different based on Duncan Multiple Range Test (p<0.05)

Figure 4. Ash content sheet from various amounts of liquid smoke and storage days

Zulyanti et al. (2017) showed that contamination of foreign materials, such as sand, mud, and the type of coagulant material used, affected the high ash content. Purnomo et al. (2015) indicated that seasons and latex processing techniques also affect latex ash content.

According to Draudt (1963 in Darmadji et al., 1998), liquid smoke contains phenol, carbonyl, tar, resin, and benzopyrone crystals. These components will increase the dirt and ash content in rubber, and the high levels of these impurities will reduce the purity level of polyisoprene in rubber.

Volatile Matter

Volatile matter analysis indicates the content of the materials, especially water, in the rubber. Residues of these materials can cause a foul odor in rubber and allow fungal growth (Falaah et al., 2016). The volatile matter of the sample treated with several ratios of liquid smoke and days of storage can be seen in Figure 5.



Notes: Different letters indicate significantly different based on Duncan Multiple Range Test (p<0.05)

Figure 5. Volatile matter from samples treated with various amounts of liquid smoke during several storage days

The volatile matter is the percentage of water content in rubber, and factors affecting volatile substances' content are temperature and drying time, milling creeper conditions, and latex added with high doses of ammonia. The level of volatile substances increases during storage due to the conditions of the storage room.

Figure 5 shows that the lowest volatile matter value is found in the ratio of 10% with a storage time of 1 day at 0.11%. For comparison, the highest volatile matter was found in control, and the amount was 5% with a storage period of 14 days at 0.51%. The results

of volatile matter in Figure 5 showed a significant difference between the coagulant liquid smoke and the control ratio.

According to Sakdapipanich and Rojruthai (2012), about 50 volatile organic components from various rubber classes can be classified into (1) compounds with low polarity, aliphatic and aromatic hydrocarbons, (2) compounds of medium polarity, aldehydes, ketones, (3) high polarity compounds, volatile fatty acids, (4) nitrogen or sulfur-containing derivatives. The components contained in the liquid smoke of clone PB 340 were ethylamine, benzylhydrazine, and low molecular weight short-chain fatty acids, namely acetic, propionic, isobutyric, butyric, isovaleric, and valeric acid. Factors affecting volatile matter include drying conditions, creeper conditions, and ammonia content.

Dirt Content

Foreign insoluble materials that cannot pass through the 325 mesh filter are impurities. The level of impurities in the base and specifications of rubber processing materials greatly affect the elasticity and flexibility of rubber products. The impurity content of samples treated with several ratios of liquid smoke can be seen in Figure 6.



Notes: Different letters indicate statistically significantly different based on Duncan Multiple Range Test (p<0.05)

Figure 6. Dirt content of sheets treated with various amounts of liquid smoke during several storage periods

Figure 6 shows that the lowest levels of impurities are found in the ratio of 5% and 10% with a storage period of 14 days at 0.007%. According to (Draudt, 1963) (1963 in Darmadji et al., 1998), liquid smoke contains phenol, carbonyl, tar, resin, and benzopyrone crystals. These components will increase the dirt and ash content in rubber. The high levels of these impurities can reduce the purity level of polyisoprene in rubber.

Handayani (2014) stated that agglomerates

causes high levels of impurities in rubber, and foreign objects such as stones, sand, twigs, and wood chips. The relatively high impurities can reduce the superior dynamic properties of natural rubber vulcanization, such as heat generation and flexural crack resistance.

CONCLUSION

Based on the research and discussion above, it can be concluded that liquid smoke from the pyrolysis of rubber wood clone PB 340 can be used as a coagulant, with a ratio of adding 5% to 25% of liquid smoke, with storage up to 14 days. In addition, the guality of sheet rubber can be maintained, such as the initial plasticity value (P_o), plasticity retention index (PRI), volatile matter, ash content, and impurity content. In all comparisons, with a storage life of 1, 7, and 14 days, sheet rubber can be produced with quality that meets the requirements of SIR 5 (SNI No. 1903:2011), namely the overall P_o value (> 30), PRI (> 70), volatile matter (< 0.80), ash content (< 0.50), and dirt content (< 0.04). The dose of liquid smoke that is good to be added to latex is 10% (v/v) of liquid smoke. The disadvantage of liquid smoke is that there are additional treatments in producing liquid smoke, such as distillation to remove tar content. Furthermore, the color of the rubber produced will not be good when distillation is not carried out.

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CONFLICT OF INTEREST

There is no conflict of interest concerning the study.

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