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Improving the quality of liquid smoke by filtration using membranes

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Abstract

Liquid smoke (LS) is getting more attention in many applications, especially food preservation. However, there is still a big problem with the purity of LS. The objective of this study is to improve the quality of LS using membrane filtration. The filtrations were conducted using ultrafiltration or nanofiltration membrane at various pressures. The quality of LS before and after filtration was compared based on their appearance. During the filtration process, LS changed from a dark yellow fluid with a pungent odor to a pale colorless substance with a less smokey odor and a slightly sweet aroma. The color and fragrance parameters of LS before and after filtration using UF membranes were not significantly different. NF membrane filtering had the opposite effect on the appearance of LS. Chemical composition of volatile fraction was studied by GC/MS. Phenols were the predominant aromatic chemicals found in all samples, with LS containing up to 80.8%, 17.53% in treated LS by UF membrane (2 bars), and 48.31% in NF membrane (5 bars). The results proved that the membrane processing can eliminates potentially toxic compounds while still imparting the desired flavors and pleasent aromas of LS.

1. Introduction

In the last decade, liquid smoke (LS) has become one of the main preservatives' choices in food applications. In addition, LS is also applied in protein foods acting as a coloring and flavoring agent (Tuesta-Chavez *et al.*, 2022). This ability comes from various organic compounds in LS, such as acids, phenolics and carbonyl compounds (Lingbeck *et al.*, 2014; Xin, Dell, Udugama *et al.*, 2021). However, LS also contains several chemical components like tar, polycyclic aromatic hydrocarbons (PAH), and their derivatives, which are carcinogenic to humans (Simon *et al.*, 2010). In order to ensure safe food products, we need to make LS that does not exceed applicable regulations.

Liquid smoke characteristics depend on several factors, such as the type of wood and pyrolysis temperature (Tegang *et al.*, 2020; Xin, Dell, Udugama *et al.*, 2021). All these factors, mainly the type of wood used, influence the chemical composition of the smoke that permeates the products. Different woods generate different levels of phenols, carbonyls, and organic acids, which affect their antimicrobial properties (Lingbeck *et al.*, 2014). Hardwood will have a more excellent syringol value than softwood which can boost the flavor properties of the LS (Montazeri *et al.*, 2013). Other compounds like organic acids, have a high odor threshold value and have such a negligible contribution to the overall odor of smoked products (Tegang *et al.*, 2020).

The production of LS can be accomplished in three different ways: 1) smoldering, which is a form of combustion that takes place at a low temperature without producing flames; 2) carbonization, which is a form of slow pyrolysis that takes place in the absence of oxygen using an external heat source; and 3) fast pyrolysis, which is very similar to carbonization except that higher heat temperatures are used (Xin, Bissett, Wang *et al.*, 2021). Most of the industries used smoldering methods to prepare LS. This procedure is inefficient and pollutes the air by emitting harmful CO and NOx. In addition to

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carbonyls, acids, and phenols, pyrolysis of wood often generates unfavorable compounds such as PAH. PAH occurs due to incomplete burning and is typically formed at pyrolysis temperatures between 500°C and 900°C (Akhtar and Amin, 2012)

Liquid smoke can be fractionated to remove unwanted compounds, as it efficiently separates LS and a bio-oil product, including heavy tar and aerosol oil (Papari and Hawboldt, 2018; Xin, Bissett, Wang *et al*., 2021). One way of fractionation that can be conducted is by distillation. Research conducted by Maulina and Karo (2021) showed that the distillation process of LS can remove tar and PAH compounds. However, some studies showed that the purification of LS by distillation method or fractionation is less effective (Rozum, 2014; Tegang *et al*., 2020; Yulistiani *et al*., 2020). During the distillation process, many of the acid fractions contained in the LS are lost due to heat applied (Muhammad *et al*., 2011; Lombok *et al*., 2014). Acid compounds in LS serve to kill bacteria that can spoil food. If the acid content in LS is low, LS is no longer effective as a food preservative. Therefore, it is necessary to develop other methods besides distillation to purify LS without reducing its quality.

One of the purification methods that can be used is membrane filtration, which separates unwanted compounds based on their particle size. The use of membranes has advantages in the process of food filtering, such as operations carried out at room temperature so that energy consumption is lower and prevents the quality from decreasing due to heat treatments (Susanto *et al*., 2016). On the other hand, the limited literature on the use of membranes in the purification of LS is caused by the mindset of most people that membranes are only used effectively in water purification. It is important to carry out the use of membranes to remove unwanted components and to improve the quality of LS. In this context, this present work studies on the use of ultrafiltration and nanofiltration for removing unwanted component from LS.

2. Materials and methods

2.1 Materials

Liquid smoke (LS) (grade 2 from burning coconut shells and has undergone a distillation process, produced by UKM 3 brilliant, Yogyakarta, Indonesia) was used. Two membrane types (ultrafiltration, DSS-GR81PP (purchased from Alfa Laval) and nanofiltration, NF99 (purchased from Alfa Laval)) were used for membrane filtration. Aquadest or pure water used was produced by a homemade integrated membrane unit (conductivity 1.7 µΩ/cm). More detailed membrane and pressure used are presented in Table 1.

2.2 Liquid smoke analysis

The characteristic of liquid smoke was analyzed by the GC-MS method. All LS were injected using a 1:100 inlet split ratio analysis to provided better separation and peak resolution for the majority of detected compounds. The compounds were identified using the Wiley Library (Wiley9.lib). When the similarity index (SI) was >90%, the compound was considered "likely known", when SI between 80% and 90% identified as "tentatively identified" and when SI<80% identified as "unknown". Peak areas were recorded and reported as total ion count (TIC) and as peak percentage (%TIC), while retention times were recorded and reported in minutes. The quality of liquid smoke is also analyzed based on aroma and color parameters.

2.3 Crossflow experiment and analysis

Crossflow experiments were carried out using a self-made laboratory scale for multipurpose filtration. Figure 1 shows a simplified experimental setup diagram. The filtration followed our experimental protocol in previous publication (Istirokhatun *et al*., 2018). The membranes (circle-ocular membrane disks with the diameter and area of 4.2 cm and 13.85 cm^2, respectively) were immersed in distilled water for 30 minutes, and compaction using pure water as feed for 30 min. Thereafter, filtration of LS was conducted at room temperature (25±2°C) and constant transmembrane pressure of (1, 2 and 3 bars for ultrafiltration; 4, 5 and 6 bars for nanofiltration). The permeate flux over filtration time was monitored and gravimetrically measured. A

Table 1. Membrane properties.

Type	Classification	Pressure (bar)	Material	MWCO (Da)
DSS-GR81PP	UF	1	Polysulfone	10,000
DSS-GR81PP	UF	2	Polysulfone	10,000
DSS-GR81PP	UF	3	Polysulfone	10,000
NF99	NF	4	Polyamide	200
NF99	NF	5	Polyamide	200
NF99	NF	6	Polyamide	200

UF: ultrafiltration, NF: nanofiltration, MWCO: molecular weight cut off.

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new circular membrane disk was used in each experiment. To maintain a constant feed concentration, the retentate and permeate were returned to the feed tank.

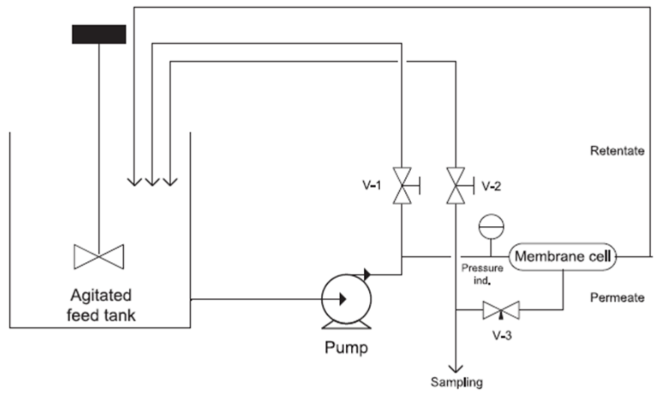

Figure 1. The simplified diagram of filtration experimental set -up (Istirokhatun *et al*., 2018).

In term of quality, CIELAB color space using colorimeter sensor was applied to each sample. The display on colorimeter showed a value containing the brightness (L^*), red/green (a^*), and yellow/blue (b^*). The LS color was determined using the AMT-501 Colorimeter. CIELAB color space results was plotted, and metric chroma (C^*) value was presented. The metric chroma (C^*) is defined as follows (CIE, 2004).

$$C^* = [(a^*)^2 + (b^*)^2]^{1/2} \quad (1)$$

Control and treated LS were compared and scored according to their aroma. Overall acceptance was based on a four-point hedonic scale. The score scale was ranged from “very much dislike” (score 4) to “very much like” (score 1). Twenty-four panelists organoleptically tested the resulting LS. SPSS for windows (version 22, IBM, Inc., NY, USA) was used to apply significant difference among the group with One Way ANOVA test at $p<0.05$. The Tukey–Kramer post hoc test was applied to determine homogenous subsets and their significance between categories.

3. Results and discussion

3.1 Liquid smoke characteristic

Liquid Smoke is generally brownish, as described in Table 2. Based on the literature, compounds that cause color in LS are carbonyl compounds (Rozum, 2014; Lund and Ray, 2017) and phenolic compounds (Silva *et al.*, 2018; Maga, 2018). The color of the smoked product is due to the interaction between the carbonyl group and the amine group (Varlet *et al*., 2007). Other research reported that syringol and its derivatives were important phenolic compounds which could boost the properties and contributed to liquid smoke’s flavoring and coloring (Maga, 2009; Silva *et al*., 2018). With various references, Xin, Dell, Udugama *et al.* (2021) attempted to explain the color components of LS; it was found that carbonyl and organic acids with a molecular weight of 400 to 1000 Da are components in the color of LS.

Liquid smoke has a distinctive smoke smell. Smoke flavor compounds, including phenols, are responsible for the smoke flavor and smoky aroma while carbonyl compounds impart a sweet aroma and color to smoked meat products (Lingbeck *et al*., 2014). Other research reported that the class of phenols and its derivatives have been considered as the main contributors to smoke flavor and aroma (Cardinal *et al*., 2006). Phenols also have antibacterial and antioxidant properties (Lingbeck *et al*., 2014). Like phenols, carbonyl groups (aldehydes, ketones, their derivatives) also have antibacterial properties but provide a background odor and taste described as sweet, caramel, that surrounds the smoky smell in LS (Kostyra and Barylko-Pikielna, 2006).

Volatile compounds identified from LS are represented in Table 3. Phenolic compounds were detected constituted as major portion of LS and accounted for 80.8% TIC. The most abundant phenolic chemical was syringol (2,6-dimethoxyphenol), followed by guaiacol (2-methoxyphenol) and 1,2,3-Trimethoxybenzene. The ratio of syringol derivatives to guaiacol derivatives ranged between roughly 2.1:1 (lower than 3.3:1), which is indicative of the usage of hardwood as a wood source (Baltes *et al*., 1981). These findings are also reinforced by the fact that hardwoods appear to be the industry's favored LS source (Xin, Bissett, Wang *et al*., 2021). Other identified compounds were 4,5-dimethylimidazole compounds. The primary carcinogens in LS (Zachara *et al*., 2017), poly-aromatic hydrocarbons (PAHs) were not found in this study. A relatively low pyrolysis temperature was maintained in burning coconut shells by UKM 3 brilliant and formation of PAHs was induced at higher temperatures between 500°C and 900°C (Akhtar and Amin, 2012). Due to the low similarity index value, many compounds are not well identified. Liquid smoke pyrolysis system produces organic acids (Lombok *et al*., 2014), carbonyl (Tegang *et al*., 2020) and aldehydes (Rozum, 2014) that may also be present in LS.

The flux profile (expressed as relative flux, the ratio of permeate flux to initial flux) measured every 15 minutes for 2 hrs was investigated. Two membranes, i.e. ultrafiltration (UF) and nanofiltration (NF) were used in this experiment. The results are presented in Figure 2.

In all conditions, both UF and NF, the permeate flux showed rapid decline in the beginning of filtration followed by gradual decrease until relatively stable flux obtained. The reason for this behavior is due to concentration polarization and fouling. The rapid decline in the beginning of filtration is mainly due to

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concentration polarization whereas the gradual decline for long period was due to fouling phenomenon (Zuriaga -Agustí *et al*., 2014; Ho *et al*., 2016). No difference in flux behavior was observed for UF and NF membrane.

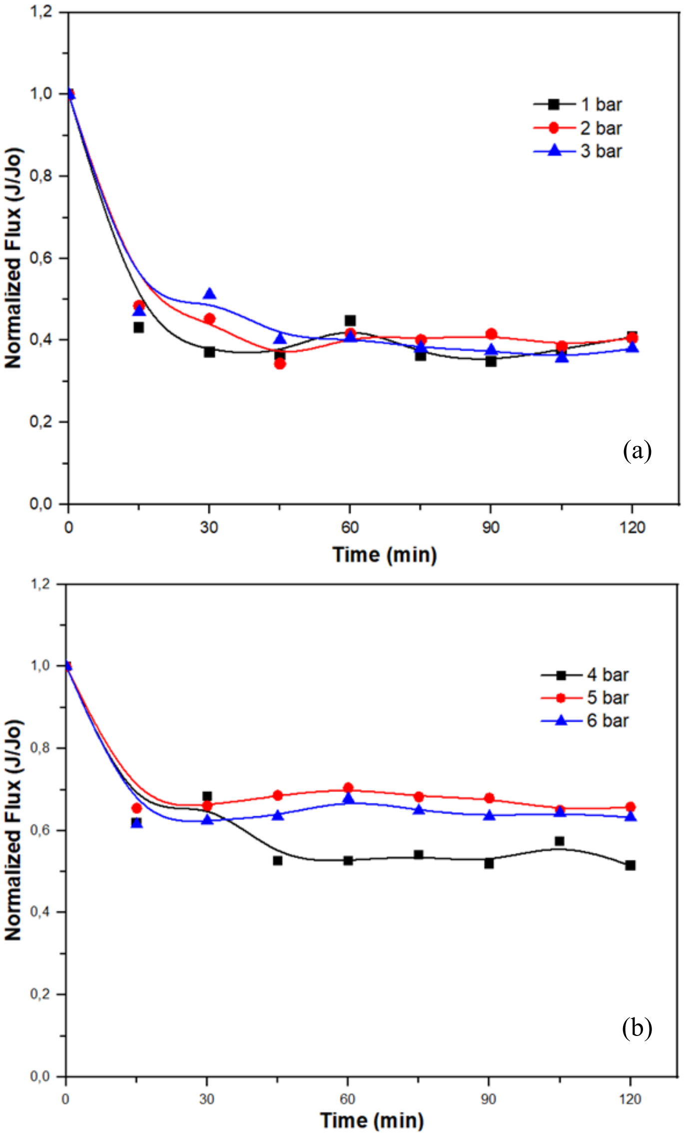

Figure 2. Flux behavior J/Jo during filtration liquid smoke using membrane with various TMP (a) UF and (b) NF.

The effect of transmembrane pressure on permeate flux during ultrafiltration was observed. The increasing pressure from 1 bar to 2 bars increased the permeate flux, however further increase to 3 bars decreased the resulting permeate flux. At first, the increase in TMP increased the driving force of the process leading to increase in permeate flux. Further increase in TMP increased not only the driving force but also the potential to push the solute or non-liquid components in the feed into the membrane surface and pores leading to fouling and flux decrease. Similar phenomenon in the effect of pressure on permeate flux was observed for NF membrane. These phenomena suggest that the dominant separation was due to sieving mechanism. In this context, the best TMP for filtration of LS was 2 bars for UF membrane and 5 bars for NF membrane.

3.2 Treated liquid smoked appearance

Beside the permeate flux as quantity parameter, the quality of the treated LS is also important parameter in determining the membrane performance. The best LS should have clear color, slightly sour taste and neutral aroma (Alçiçek and Balaban, 2015; Himawati *et al*., 2018; Malelak *et al*., 2020). The results are presented in Table 2.

In the CIELAB color space, L^* denotes lightness (brightness–darkness) with values from 100 (white) to 0 (black). Here, a^* and b^* indicated chromaticity corresponding to green (negative value) to red (positive value) for a^*, and blue (negative value) to yellow (positive value) for b^*.

The Δa^* and Δb^* values are calculated as the difference between color parameters of treated LS and those of feed LS. Figure 3 plots Δa^* against Δb^*. Overall treated LS had some negative Δb^* value (decrease in yellow) and negative Δa^* value (increase in green). The Δa^* value of LS and UF 1 bar LS were almost the same. The ΔC^* value indicates the difference between the metric chroma of treated LS and that of feed, and ΔL^* indicates the difference between the brightness of treated LS and that of feed. Figure 4 plots ΔL^* against ΔC^* to show color differences in the treated LS.

The ΔL^* values of all treated LS were positive. However, the value of UF membrane-treated LS was

Table 2. Color properties and aroma response of feed and treated liquid smoke.

	Aroma Response	CIELAB color space		
		L*	a*	b*
LS	3.54±0.58[a]	42.12±1.17[a]	1.96±0.32[a]	28.18±0.66[a]
UF 1 bar	3.42±0.58[a]	43.61±0.99[ab]	1.93±0.16[a]	26.46±0.70[b]
UF 2 bar	3.33±0.63[ab]	45.35±2.23[bc]	0.69±0.47[b]	21.20±1.03[c]
UF 3 bar	3.17±0.76[abc]	46.33±0.21[c]	-0.26±0.17[b]	17.91±0.47[d]
NF 4 bar	2.79±0.77[bc]	59.37±0.40[e]	-4.39±1[c]	2.57±0.52[e]
NF 5 bar	2.79±0.72[bc]	56.93±0.98[d]	-4.79±0.68[c]	1.64±0.63[e]
NF 6 bar	2.58±0.77[c]	59.39±0.82[d]	-5.34±0.51[c]	1.43±0.52[e]

Values are presented as mean±SD. Values with different superscripts within the same column are statistically significantly different ($p<0.05$).

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small (~4), while the values of NF membrane-treated LS were large (up to 17) (Figure 4). The ΔC^* values of UF membrane-treated LS were negative in the range of −1.72 to −10.34, while those of NF membrane-treated LS were negative in the range of −22.72 to −23.19.

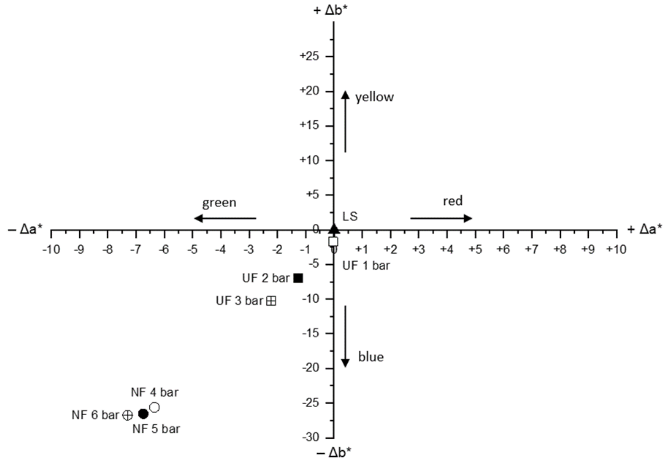

Figure 3. Plots of Δa* versus Δb* for the membrane-processed liquid smoke.

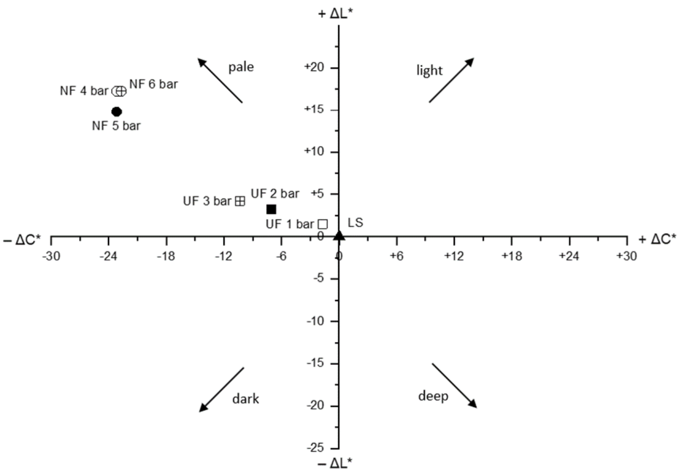

Figure 4. Plots of Δ*C** versus Δ*L** for the membrane-processed liquid smoke.

The treated LS by filtering using membrane has a more transparent color, indicating decolorization has occurred. It is clearly seen that the decolorization of LS was more significant by NF membrane than UF membrane. The difference between membrane color separation performances can be explained as follows. Definitely, this is due to smaller pore size of NF membrane than UF membrane. Many components in LS having low molecular weight (lower than 1000 g/mol) could not be separated by UF membrane but they can be separated by NF membrane. Therefore, the rejection of the color compounds for the UF membrane (10 kDa), with higher decolorization ability for the NF membrane (200 Da), are reasonable.

In the food industry, the aroma is an important aspect that can quickly assess whether a product is acceptable. The customer acceptance rate for aroma sensorial is shown in Table 2. Overall, there is no significant difference in the UF membrane-treated LS's aroma from the feed's aroma. They can be considered members of the same group. Table 2 reveals that group a consists of treated LS by UF membrane (1, 2 and 3 bars), whereas group b consists of treated LS by UF membrane (2 and 3 bars) as well as NF membrane (4 and 5 bars). Group c consists of treated LS by a UF membrane at a pressure of 3 bar, as well as all treated LS by an NF membrane. All groups contain treated LS with UF membrane (3 bars). There are no statistically significant differences amongst aromas sharing a letter. LS and treated LS by NF membrane (6 bars) do not share a letter, indicating that treated LS by NF membrane (6 bars) has a substantially higher aroma value than LS.

Besides being successful in clearing the LS colors, membrane process also tended to soften the heavy smoky aroma associated with phenols. The degradation of aroma was more significant for NF membrane. This observation should be due to the rejection of some components existed in the LS by the membrane via sieving mechanism. Certainly, NF membrane could retain more component than UF membrane. For the NF membrane, in addition to sieving mechanism, Donnan equilibrium and dielectric exclusion may play a role. Based on Donnan equilibrium and dielectric exclusion, phenol has a positive charge while the membrane has a negative charge, thus phenol should be able to be absorbed and pass through the membrane.

Phenols are responsible for the aroma of LS, as well as their antioxidant and antimicrobial properties. Many phenolic compounds are bioactive compounds showing inhibitory effects on mutagenesis and carcinogenesis (Kim *et al*., 2011). Thus, the removal of phenolic compound should be conducted properly, on the one hand it could be increased the quality of LS in term of aroma, on the other hand it could reduce the functional of LS. Thus, further analysis for chemical composition was performed by using GC-MS.

3.3 Chemical composition

The analysis of treated LS by UF and NF membrane performed using GC-MS is presented in Table 3. GC-MS analysis results supported the results obtained on the characterization of color and aroma. There is rejection of various phenolic compounds reducing smoke odor. The carbonyl molecules in LS are weak acids that are slightly positively charged, while membranes have a negative charge. Due to this charge difference the charge repulsion leading to rejection might occur. However, due to the sieving mechanism, many components having negative charge including carbonyl can still be observed in permeate flux. Even though carbonyl rejection occurs,

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Table 3. Retention time (t_R) of identified volatile compound in liquid smoke before and after membrane processed.

Compounds	LS				UF 2 bar				NF 5 bar			
(mass spectra data)	t_R (min)	SI	TIC	%TIC	t_R (min)	SI	TIC	%TIC	t_R (min)	SI	TIC	%TIC
Aldehydes and ketones												
Methyl cyclopentenolone									4.224	97	1,366.40	2.6
Olealdehyde					23.21	82	3,650.00	10.07				
Pentadecyl ketone					24.355	88	1,554.80	4.29				
Organic acids												
Phenyl Carbamate									3.411	95	14,759.40	28.14
Ethyl Oleate					13.828	92	1,304.50	3.6				
Phenolic compounds												
Phenol	3.505	98	7,566.60	57.44	3.527	98	5,423.60	14.96	3.57	97	10,864.70	20.71
Phenol									3.73	98	4,750.20	9.06
2-Methoxyphenol	4.996	94	913.8	6.94					4.929	96	4,822.90	9.19
2-Methoxy-4-methylphenol									6.403	96	843.7	1.61
2,6-Dimethoxyphenol (Syringol)	8.225	93	1,887.60	14.33	8.247	93	930.1	2.57	8.231	93	4,059.30	7.74
1,2,3-Trimethoxybenzene	9.065	83	275.9	2.09								
Miscellaneous												
4,5-Dimethylimidazole	2.081	82	568	4.31								
1-Methoxycyclohexane									2.883	88	2,011.60	3.83
2-Tetradecyloxirane					12.583	85	971.1	2.68				
14-Beta-H-Pregna					17.601	82	1,593.40	4.39				
Total aldehydes and ketones			Un	Un			5,204.80	14.36			1,366.40	2.6
Total organic acids			Un	Un			1,304.50	3.6			14,759.40	28.14
Total phenolic compounds			10,643.90	80.8			6,353.70	17.53			25,340.80	48.31
Total miscellaneous			568	4.31			2,564.50	7.07			2,011.60	3.83
Total unknown compound			1,960.40	14.89			20,828.80	57.44			8,978.90	17.11

Un = unidentified. Similarity index (SI) was identified using the Wiley9.lib. The area of peaks is reported in 10^4 TIC.

the carbonyl content becomes larger, because the other contents, especially phenol, were also reduced more significantly. Because carbonyl has antimicrobial properties, the quality of LS resulting from membrane filtration in food preservation is maintained.

The refining process reduced the content of phenols but decrease aldehydes and ketones in treated LS UF 2 bar and NF 5 bar. The olealdehyde is detectable in treated LS UF 2 bar. Olealdehyde is a food additive used to improve the taste or smell of food . Therefore, it is true that there is an improvement in aroma after membrane UF filtration, though there is not a major difference. On the other hand, methyl cyclopentenolone is available in high proportion in the permeate flux of NF 5 bar. The smell and taste of cyclotene have been described in literature with very pronounced sensory notes similar to that of maple syrup and to a lesser extent that of liquorice (Chartier, 2012). This demonstrates that filtration using a 5 bar NF membrane produces LS with a less smokey odor and a sweet aroma.

Organic acids originated from the pyrolysis of wood carbohydrates play an important role in the flavor, color, texture and microbiological stability of foods impregnated during smoking (Rozum, 2014). The organic acids found in the treated LS by NF membrane were phenyl carbamate (NF 5 bar) and treated LS by UF were ethyl oleate (UF 2 bar) (Table 3). In accordance with the original purpose of LS, these chemicals provide treated LS the capacity to preserve food. This is consistent with Xin, Dell, Udugama *et al*. (2021) assertion that utilizing acid with the antibacterial potential of LS reduces the number of live bacteria in meat samples. The same cytotoxicity assay was previously reported in the studies by Kim *et al*. (2011), The acidic environmental conditions of LS were mainly responsible for reducing cell viability.

Numerous studies (Montazeri *et al*., 2013; Lingbeck *et al*., 2014; Tegang *et al*., 2020; Xin, Dell, Udugama *et al*., 2021) have analyzed the chemical profile of LS to identify the chemical components responsible for the organoleptic qualities and to better comprehend their relationship. Based on the findings of these studies, it can be deduced that the key components of LS include phenols, organic acids, furans, pyrans, aldehydes and ketones. The class of phenols and its derivatives are regarded to be the primary contributors to the flavor and aroma of LS (Cardinal *et al*., 2006). Furans and pyrans provide fruity, caramel-like, and sweet aspects to foods and help reduce the phenolic compounds' highly noticeable smoky flavor (Kostyra and Barylko-Pikielna, 2006). The carbonyl groups of ketones and aldehydes interact with the amine groups of the dietary amino acids, so contributing to the golden brown color of smoked items (Varlet *et al*., 2007). Organic acid contributes to the overall odor of LS by imparting a somewhat pungent smell; it also possesses antibacterial effects against sporulating bacteria and food deterioration fungi (Martinez *et al*., 2011).

On the basis of the findings in this research on the chemical composition of LS and the impact of specific chemicals on the organoleptic features of LS products. It is proposed that filtering LS through an NF membrane with a TMP of 5 bars can improve the quality of LS products.

4. Conclusion

In conclusion, the results obtained in this study reveal that the membrane filtration improves the quality of LS both in terms of its sensorial characteristics and composition. Using a NF membrane, LS was greatly decolored, whereas a UF membrane had a minimal effect. The decolorization performance of the NF membrane was mostly determined by its MWCO. By changing the blend ratio, plots of Δa^* against Δb^* and ΔC^*against ΔL^*draw characteristic curves. By filtering with a NF membrane, a LS color can be tailored to the customer's preferences. As a result, the refined LS had significantly lighter color, and a pleasant smoky aroma.

Preliminary analysis of LS using GC/MS indicates the presence in large quantities of certain phenolics reported in the literature as the most active aromatic compounds, which may help explain these woods' effect on the sensory properties of smoked products. The NF membrane was likely to reject phenolic compounds more than the UF membrane. The membrane processing of LS eliminated phenolic compounds and, to a lesser extent, reduced the contents of carbonyl-containing compounds. Overall, the results from this study are useful to food product developers seeking to determine a suitable quality of liquid smoke products.

Conflict of interest

All authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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