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OPTIMAL EXTRACTION AND EVALUATION ON THE OIL CONTENT OF CITRONELLA OIL EXTRACTED FROM *CYMBOPOGON NARDUS*

(Pengekstrakan Optimum Dan Penilaian Kandungan Minyak Sitronela Daripada *Cymbopogon Nardus*)

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Abstract

An investigation on the extraction of citronella oil from *Cymbopogon nardus* (*C. nardus*) using a custom made medium scale steam distillation apparatus has been conducted. The Clevenger apparatus was used to generate a continuous flow of the hydrosol, creating an efficient distillation system. In the case for *C. nardus* leave samples orientation; the sleeping/standing and close/loose packing in steam distillator was found to have significant effects on the yield of extraction. The 3.5 hours distillation process produced an extraction yield of 0.64% in sleeping position of the leaves as compared to the standing position (0.43%), while the loose packing (0.70%) has almost two fold of yield percentage as compared to the close-packing (0.40%) of the leaves. Therefore, the steam distillation of *C. nardus* leaves using a custom made medium scale steam distillation apparatus was found to be most effective in the combination of sleeping position with a loose packing. Furthermore, the age of *C. nardus* leaves also affect the physical and chemical quality of the citronella oil extracted. The younger leaves produced citronella oil that consists mainly of citronellal, citronellol and geraniol while the older leaves contained a high composition of citral.

Keywords: steam distillation, citronella oil, *cymbopogon nardus*, extraction, essential oil

Abstrak

Penyiasatan terhadap pengekstrakan minyak sitronela daripada *C. nardus* telah dilakukan dengan menggunakan radas penyulingan stim skala sederhana yang khas disediakan. Alat radas Clevenger yang digunakan dapat menghasilkan aliran kitaran hidrosol berterusan yang meningkatkan kecekapan proses pengekstrakan minyak. Didapati orientasi (tidur/menegak) dan kepadatan (rapat/longgar) daun *C. nardus* dalam proses pengekstrakan memberi kesan ketara kepada hasil pengekstrakan minyak. Penyulingan stim yang dilakukan selama 3 jam memberikan hasil pengekstrakan 0.64% bagi daun dalam kedudukan tidur berbanding dengan 0.43% bagi daun dalam kedudukan menegak; pengekstrakan sebanyak 0.70% didapati bagi daun dalam kepadatan longgar berbanding dengan 0.40% bagi daun dalam kepadatan rapat. Oleh itu, hasil pengekstrakan minyak *C. nardus* paling berkesan adalah gabungan kedudukan daun dalam keadaan tidur dan kepadatan daun yang longgar. Tambahan pula, usia daun *C. nardus* didapati dapat memberikan kualiti fizikal dan kimia yang berbeza kepada minyak sitronela yang diekstrak. Daun muda memberikan sebatian utama iaitu sitronelal, sitronelol dan geraniol manakala daun yang lebih matang memberikan komposisi sitral yang tinggi.

Kata Kunci: penyulingan stim, minyak sitronela, *cymbopogon nardus*, pengekstrakan, minyak pati

Introduction

This paper is a continuation of the previous work on the optimal production of the citronella oil from *Cymbopogon nardus* (*C. nardus*) [1]. The preliminary work on the effect of leaves orientation (Figure 1(a)) on the extraction yield of *C. nardus* (Figure 1(b)) was confirmed with further trials and shown in table 1. Besides, the steam distillator was custom-made to have a Clevenger apparatus connected to the opening of the distillation vessel rather than using the conventional apparatus, separatory funnel, at the end of the distillatory as the collector. This modification provides an efficient and continuous flow of the hydrosol in the extraction cycle. In this paper, we also aimed to analyze the composition and the general properties of the extracted oil of different ages of *C. nardus* leaves. The harvest time for the leaves of genus *Cymbopogon* plant was done widely on species such as *Cymbopogon citratus* [2], *Cymbopogon flexuosus* [3] and *Cymbopogon winterianus* [4-6]. There were significant differences shown in composition to ages. Recent studies on the evaluation of the content and composition of *C. nardus* essential oil from different harvest times was done by Castro et al. [7]. The research shows that leaves in Tocantins State have 3 main compounds which are citronellol, geraniol and elemol. Thus, *C. nardus* leaves from different ages grown in Malaysia were taken for the identification of composition and physical properties. Result shows that the composition of the essential oil extracted has main compounds such as citronellol, citronellal, geraniol, nerol and citral.

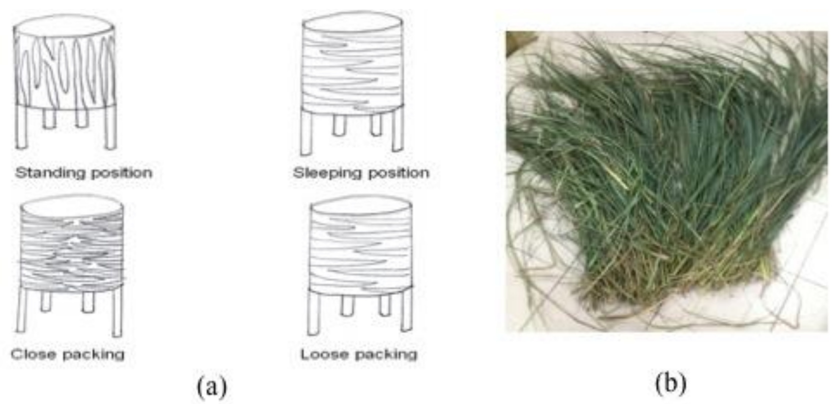

Figure 1. (a) Positions and packing of leaves and (b) *Cymbopogon nardus* leaves

Materials and Methods

Steam distillation set up

The steam distillation process was done 3 times in each category and the average yield was obtained in Tables 1 and 2. Leaves were dried for an hour before the distillation process to evaporate most of the moisture. Experiments were conducted on a 1.5-4.5 kg scale. The steam distillation set-up is custom-made to a medium scale vessel that can fit an approximate maximum 5 kg worth of *C. nardus* leaves. A butanol gas tank was used as fuel to boil up the water. Clevenger apparatus was used in the distillation process.

Steam distillation of *Cymbopogon nardus*

C. nardus leaves were collected at Section 16 Bandar Baru Bangi, Selangor, Malaysia and dried under the sun for one hour. The dried leaves were then chopped to 2-3 portions with an equivalent length each and placed into the distillation vessel. Leaves for standing position were chopped to match the height of the distillation vessel. The condenser with a Clevenger apparatus was connected to the distillation vessel at the top. The distillation process was run for 3.5 hours from the first drop of condensed water collected. The citronella oil obtained was dried with anhydrous sodium sulfate (Na_2SO_4) and then was analyzed using gas chromatography mass spectrometry (GC-MS) analysis. Experiment on close versus loose packing was done in sleeping position as result of citronella oil extracted was more effective in sleeping position.

Gas Chromatography-Mass Spectrometry (GC-MS) analysis

The extracted citronella oil was analyzed by GC-MS performed on an Agilent 7890A gas chromatograph coupled with a mass spectrometer system of an Agilent 5975C inert Mass Selective Detector (MSD) with a triple-axis detector. The gas chromatograph was equipped with a fused silica capillary column AB-5MS (5% phenylmethylpolysiloxane, 30m x 0.25mm, film thickness 0.25µm). The injector and interface were operated at 250°C and 300°C, respectively. The oven temperature was raised from 50°C to 300°C at a heating rate of 8°C/min and then isothermally held for 5 minutes. The constant head pressure was 0.6 bar. As a carrier gas helium at

1.0mL/min was used. The samples were injected in a pulsed split mode. Mass selective detector was operated at ionization energy of 70eV, in the 35-500amu range with a scanning speed of 0.34s.

Results and Discussion

Extraction of citronella oil

Table 1 shows the results of the experiment where the sleeping position has a higher yield in the extraction of *C. nardus* compared to the standing position. This may be due to the direct contact of the stream of steam from the bottom of the distillation vessel with the leaves. The sleeping position closes almost all of the gaps horizontally forcing steam created to boil the leaves whereas standing position allows the steam to pass through without fully contacted with the leaves. Therefore, only part of the steam was in contact to boil the leaves and part of it escaped upward without carrying any oil resulting in a lower yield of extraction. Percentage yield of the extraction was based on the simple calculation in equation (1) below.

$$\text{Percentage yield (\%)} = \frac{\text{Weight of extracted citronella oil}}{\text{Weight of } C.nardus \text{ leaves used}} \text{ x } 100 \tag{1}$$

Table 1. Data of extraction yield (%) of *C. nardus* leaves (kg) by steam distillation under different positions

	1st	**2nd**	**3rd**	**Average**
Sleeping	1.52 kg	1.50 kg	1.52 kg	1.51 kg
	0.68%	0.62%	0.61%	0.64%
Standing	1.52 kg	1.56 kg	1.53 kg	1.54 kg
	0.45%	0.42%	0.41%	0.43%

Table 2 shows the results where loose packing has a higher yield in the extraction of *C. nardus* compared to the close packing. This may be due to the lower resistance level of leaves blocking the stream of steam to proceed upward. Therefore, closer packing gives lower yield.

Table 2. Data of extraction yield (%) of *C. nardus* leaves (kg) by steam distillation under different packing

	1st	**2nd**	**3rd**	**Average**
Close	4.00 kg	4.10 kg	4.07 kg	4.06 kg
	0.40%	0.41%	0.40%	0.40%
Loose	1.70 kg	1.61 kg	1.64 kg	1.65 kg
	0.78%	0.65%	0.68%	0.70%

Modification of steam distillation set up

The application of the Clevenger apparatus in the steam distillation of *C. nardus* had further enhanced the efficiency of the citronella oil production. Lesser monitoring attention was needed while using the Clevenger apparatus compared to the conventional oil collector, separatory funnel, during the distillation processes. The hydrosol produced was recycled back into the distillation vessel to be evaporated again. Hydrosol contains approximately 2-3% of essential oil [8-10]. Therefore, the usage of the Clevenger apparatus can enhance the production of citronella oil by steam distillation.

Quality of citronella oil extracted

Similar results in the form of physical and chemical properties were categorized together in Table 3. Since the *C. nardus* leaves were collected from the same source, the extracted citronella oil was expected to have the same physical or chemically quality but the expectation was proven false. By varying the age of the *C. nardus* leaves collected, the results showed significant differences in both aspects. The appearance and the smell of the extracted citronella oil had clear differences as shown in Table 3. The density of the citronella oil was calculated by the simple calculation formula in equation (2).

$$\text{Density}, p = \frac{m}{V}\text{, where } m \text{ is mass and } V \text{ is volume} \tag{2}$$

Table 3. Physical and chemical properties of the extracted citronella oil

Age of leaves	1-5 months	6-10 months
Appearance		
Smell	Fragrant lemon	Pungent lemon
Refractive index	1.466 at 25°C	1.475 at 25°C
Density	0.854 g/ml at 25°C	0.873 g/ml at 25°C
Chemical composition (GC-MS)	Figure 2	Figure 2
Main composition (%)	Citronellal (39.66) Citronellol (12.98) Geraniol (18.83)	Citronellal (4.80) Citronellol (6.69) Geraniol (46.10) Citral (26.61) Nerol (4.23)

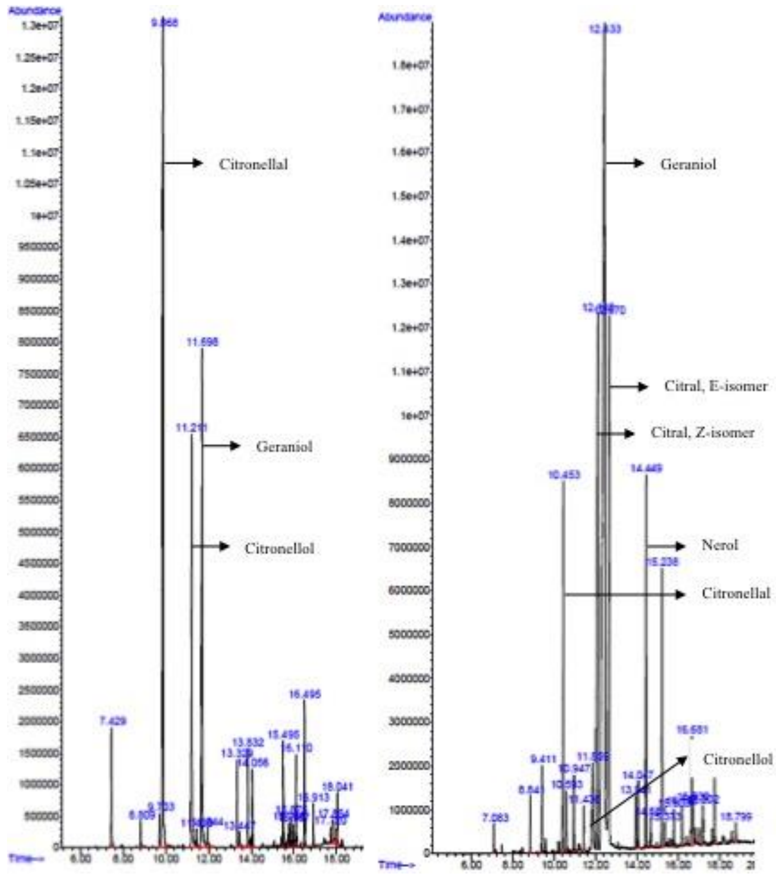

Figure 2. Chemical composition of citronella oil extracted from *C. nardus* leaves at age range between 1-5 months (left) and 6-10 months (right)

The composition of the oils shown by each category differed significantly. The younger *C. nardus* leaves were found to have a higher content of citronellal and citronellol while the older leaves were found to have a higher content in geraniol. Citral was found in high percentage in the older *C. nardus* leaves and this could be the main compound that contributed to the differences in smell and appearance.

As compared to the work done by Castro *et al*. (2010), the oil content of the extracted citronella oil from *C. nardus* in Tocantins States is different from the ones extracted in Malaysia. Elemol, which is one of the main compound found in the citronella oil extracted from *C. nardus* leaves in Tocantins State was not found in the extracted citronella oil from *C. nardus* leaves grown in Malaysia; while citral, which forms as one of the main composition in the oil content of the older leaves was not found in the citronella oil extracted in Tocantins State. This might be affected by several factors such as climate, age of plantation and efficiency of distillation method [5].

Citral is a valuable compound which has a strong lemon odor and serves as an aroma compound and used in perfumery. It is also used widely in flavoring, antimicrobial and the synthesis of vitamin A [11,12]. Refractive index was measured uing the ATAGO-NAR-1T LIQUID. The room temperature for measurement was 25°C and the light source used was sodium vapor lamp (589.4nm).

Like most of the citronella oil produced commercially, they have a range of refractive index specification. For example, Sigma-Aldrich produces citronella oil that has a refractive index of 1.466 to 1.475 at 20°C [13]. This can be explained by the results of this experiment that the range of the refractive index of the oil may be resulted by the age of the *C. nardus* leaves taken for extraction, therefore varying the quality of the oil produced.

Conclusion

In conclusion, the leaves orientation of *C. nardus* in the custom made steam distillation apparatus has significant effect on the oil production yield. It was shown that the sleeping position had much higher yield compared to the standing position while the loose packing had almost twice the yield of the close packing in steam distillation of *C. nardus*. Therefore, a combination of sleeping position and loose packing would be an effective orientation for the steam distillation of *C. nardus*. Addition to that, the Clevenger apparatus was found to be an efficient tool to be used in the distillation process as it provides a continuous flow of the hydrosol back into the distillation vessel, preventing the wastage of the small percentage of essential oil present in the hydrosol. Besides, this experiment had shown that the age of leaves affected the physical and chemical quality significantly in the production of citronella oil. The main compounds in the citronella oil extracted from the *C. nardus* leaves grown in Malaysia in 1 to 5 months old has main composition of citronellol, citronellal and geraniol while the leaves from the 6 to 10 months old age of *C. nardus* leaves has a significant composition of citral.

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