Improvement of Agarwood Oil Extraction by Heat Transfer Control in Multiple Hydrodistillation Systems

Saiful Anwar Che Ghani[a,b*], Mohd. Nasri Muda[c], Timo Michael Mueller[d], Wan Sharuzi Wan Harun[a], Saiful Nizam Tajuddin[b], Mahadzir Ishak[a], Anwaruddin Hisham[b]

[a]*Faculty of Mechanical Engineering, Universiti Malaysia Pahang, 26600 Pekan, Pahang, MALAYSIA*
[b]*Bioaromatic Centre, Universiti Malaysia Pahang, 25200 Gambang, Pahang, MALAYSIA*
[c]*I Gawa Resources, Lot 2888, Kampung Padang Chekering, Wakaf Che Kapur, 20050 Kuala Terengganu, Terengganu, MALAYSIA*
[d]*Centre for Modern Languages and Human Sciences, Universiti Malaysia Pahang, 26600 Pekan, Pahang, MALAYSIA*
*email: anwarcg@ump.edu.my

Abstract

Quantity of oil, and chemical composition of the Agarwood essential oil should be evaluated to determine the performance of an extraction system. The aim of this work was to investigate the effects of heat transfer control (HTC), applied at three hydrodistillation systems simultaneously, on the quantitative and qualitative characteristics of extracted essential oil from inoculated Agarwood, compared to a conventional hydrodistillation (HD). The extractions by conventional and HTC-ed HD procedures were done by supplying heat from liquefied petroleum gas (LPG); the ratio of the raw material to be extracted and the solvent was 0.1 $g \cdot mL^{-1}$ and the extraction time was 72 hours. The compositions of the extracted essential oils (using HTC-ed HD and conventional HD) were assessed using gas chromatography with flame ionization detector (GC–FID). The results of the extraction processes show that the extraction of inoculated Agarwood essential oil by HTC-ed HD is faster and produces higher yields compared to the extraction by conventional HD method. Further, the testing of the chemical properties of the Agarwood oil shows that essential oil obtained by HTC-ed HD has better quality compared to the oil obtained by conventional HD. The implementation of optimum thermal management in HTC-ed HD technology in Agarwood essential oil production industry is therefore of great importance.

Keywords: Thermal management; Hydrodistillation; Heat transfer; GC-FID; Agarwood

1. Introduction

Agarwood (Gaharu in Malay, Oudh in Arabic or Jinkoh in Japanese) is a resinous heartwood, abundantly used, prominent incense of the Orient [1]. Essential oil extracted from aromatic plants is a valuable commodity for domestic and export which is in high demand by various industries for their primary and secondary products, such as the cosmetics and perfume industries, the food and beverages industries, as well as in manufacturing of medicine/ pharmaceuticals [2].

Agarwood gains economic interest has always been due to its pathological heavy and dense resin-impregnated wood, which is generated in the stem tissues as a response to wound [3]. Over half-century ago, Indian chemists separated a very complex mixture of agarwood and characterised two major sesquiterpenes, agarol [4] and agarospirol [5] from an agarwood originating from *Aquilaria agallocha* family [6]. The identification of the components of complex mixtures of terpene compounds in the oil determines the quality grade of the extracted oil for use as medicinal, aromatic, cultural and religious purposes [7, 8].

. It is reported by Kusuma and Mahfud [9] the consumption of essential oil or fly oil increases annually in the range of 8–10% worldwide. Such a trend is driven by the demands for essential oil used for the perfume industry, cosmetics, and healthcare [10]. Furthermore, the essential oil based products cannot be simply substituted by synthetic materials [11]. The good prospect of essential oil in Malaysia should be governed by the optimisation of refining technology especially by heat transfer control (HTC), and the cultivation techniques of plants in order for Agarwood industry in Malaysia can be sustainable to compete in global market [12].

To the best of our knowledge, no research work has been conducted to improve heat transfer in hydrodistillation (HD) by thermal management for the extraction of essential oil from agarwood plant. The objective of this study was to evaluate the impact of heat transfer control on the quantitative and qualitative characteristics of essential oil from inoculated Agarwood. The extraction yields, and the composition the extracted essential oils (by HD and HTC-ed HD) were compared.

2. Methodology

Raw material dried agarwood *Aquilaria agallocha* (cultivated, 72 % moisture content, dry basis, sun-dried (30–40) °C for three days) was obtained from Selangor, Malaysia in July 2017. Ground plant materials were soaked in water for 14 days before use. Dewatering process as Fig. 1 would be applied to the soaked chips until no water drip is visible before the chips are transferred into the HD system.

Figure 1. Soaked chip dewatering before transferred in still.

Three boilers have been used in this project. In every boiler soaked raw material (30 kg) was suspended in 300 L distilled water (ratio raw material to water solvent 0.1 g·mL^{-1}) then hydrodistilled during 72 h in an all-stainless steel heat exchanger apparatus (Fig. 2), as previously described. Gas burner using liquefied petroleum gas (LPG) was utilised to supply heat for evaporating the mixture and transporting the essential oil together to the condenser.

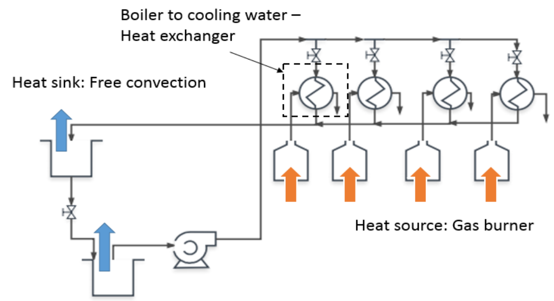

Figure 2. A HD system consists of four boilers (only three used in this project).

The improvement of extraction of essential oil from Agarwood was performed by modeling heat transfer in the conventional HD (CHD). Heat transfer control strategy involves the improvement of heat supply unit (Fig. 3a), heat removal unit (Fig. 3b), and cooling water supplying unit (Fig. 3d). Fig. 3c depicts the three HD used in the project configured in series.

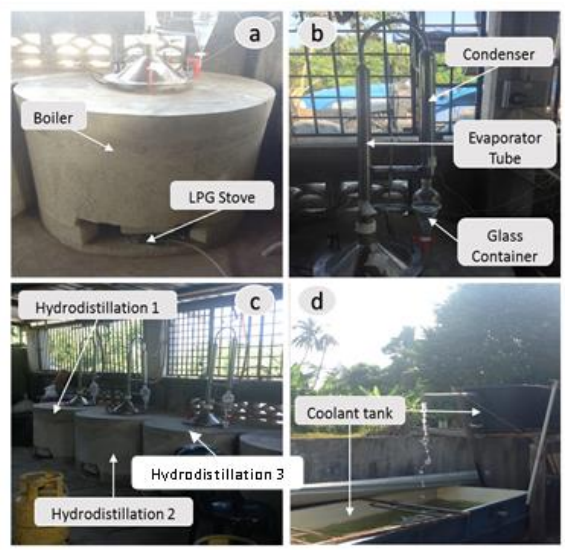

Figure 3. Heat transfer control strategy for improving HD performance (a) Boiler and heat source, (b) Evaporator tube and condenser, (c) Multi HD serial configuration, and (d) Cooling tanks.

The extractor unit included a boiler in cylindrical form (300 L capacity), and was equipped with an evaporator tube made of stainless steel with internal diameter of 100 mm and a condenser that is cooled by water (Fig. 3b). The cooling fluid (i.e. water) is supplied to the condenser through an inlet of 2 mm diameter by high mass flowrate immersible pump from Lifetech and an automatic self priming pump from OKAZAWA for CHD and HTC-ed, respectively. The specifications of the pumps are as listed in Table 1.

Table 1. Specification of the water pump

Specification	Pump CHD	Pump HTC-ed
Pump Model	SP608	P150BZ
Max. Flowrate	125 L/min	34 L/min
Suction Height		9 m
Pump Head (m)	4.2	33
Speed		2850 r.p.m.
Voltage (V)	220-240	240
Frequency	50	50 Hz
Power	100 W	0.37 kW/0.5 HP
Size (mm^3)	308×128×148	90×60×50

The burner has 4 independent rings which allow the supplied heat to be reconfigured in 16 combinations. The CHD procedure by conventional practice was strongly dependent on operators' skills and experience. The heat supplied beneath the boiler is as intuitively set by the operator to minimum amount to avoid the mixture from overcooked. The cooling water from the reservoir was pumped to the condenser at highest flowrate using SP608 submersible pump but the accurate mass flowrate had never been quantified.

The HTC-ed HD procedure commenced with determination of required heat for HD process by raising the temperature of the solution, up to the boiling point, which was achieved by varying heat (500–5000) W and applying constant mass flow rate of cooling water 20 L/min by using self priming pump. The temperatures variation was observed by attaching 8 k-type thermocouples in real time for throughout the whole 72-hours process. After the collection of the extracted essential oil, 2 µL extracted oil has been taken from each distiller for chemical composition analysis using gas chromatography flame ionization detector (GC-FID). Every process with CHD and HTC-ed HD was triplicated.

3. Results and Discussion

The HTC-ed HD in this project was developed based on the numerical analysis using Gnielinski equation to compute the heat transfer coefficient in the multi HD. The correlation between boiler heat flow, cooling water massflow and the temperature difference of the cooling liquid between the inlet and outlet in the condenser is shown in Fig.4. It can be seen for supplied heat in the range 500 to 5000 W the temperature difference of the cooling liquid at inlet and outlet for this HD configuration supposedly not higher than 3 °C. However to achieve this optimal working condition the pump must have the ability to produce cooling liquid higher than 500 kg/h. The massflow rate experiment resulted that the pump model SP608 could only produce 480 kg/h coolant whilst the pump model P150BZ could produce about 1200 kg/h of cooling massflow rate.

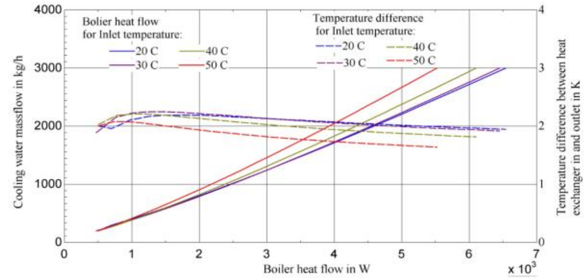

Figure 4. Result from numerical analysis

The output obtained when both CHD and HTC-ed HD were used to extract 30 kg of soaked aqualaria agalocha wood using 500 W heat is shown in Table 2. Average extracted oil yield has increased from 8.27 mL by CHD to 30.5 mL using HTC-ed. The standard deviation in HTC-ed HD reduced significantly to 2.7 % from 12.8% to indicate better reproducibility process by HTC-ed HD. Both processes, nonetheless, produce green extracted oil but the oil should be analysed chemically to quantify the quality of the extraction. Interestingly, the average steam temperature measured at the end of evaporator tube demonstrated higher reading recorded by CHD than HTC-ed. This might be due to partial steam disposed to the environment observed in CHD and heat up the evaporator tube. On the other hand, the measurements of temperature difference of the cooling liquid at the inlet and outlet of the condenser also show almost double the difference obtained by CHD than HTC-ed HD.

Table 2. Yield obtained by CHD and HTC-ed HD procedures.

Output	CHD	HTC-ed
Average Yield (mL)	8.27	30.5
Standard deviation (%)	12.8	2.7
Colour	Green	Green
Average Steam Temp. (°C)	109.7	103.4
Max. Cooling Liquid Temp. Difference (°C)	8	4.3

Gas chromatography was carried out on Agilent Technologies capillary Gas Chromatograph column equipped with flame ionization detector (FID) and head space analyzer using a fused silica capillary column (30 m×0.25 mm, film thickness 0.25 µm) coated with dimethyl polysiloxane (RTX-5). Oven temperature was programmed from 80 to 250 °C at

3 °C/min, with injector temperature 250 °C and detector temperature 250 °C. Injection volume1 μL per sample, helium was used as a carrier gas (1.18 ml/min and pressure 15.33 PSI). The sample GC result for essential oil extracted from HTC-ed in Fig.5 shows that the oil is dominated by the presence of Agarospirol/ epoxybulnesene.

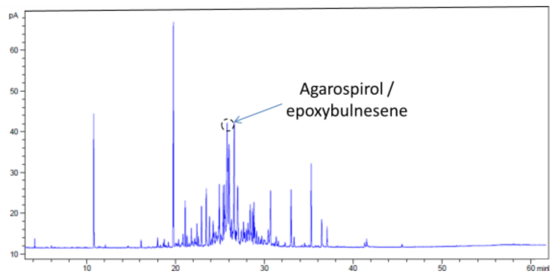

Figure 5. GC result for chemical composition analysis of oil extracted HTC-ed HD.

4. Conclusion

In conclusion, our results reveal that the essential oil obtained from HD procedure was improved significantly by optimizing heat transfer in the process. The extracted essential oils from HTC-ed HD yielded 300% more than CHD and qualitative similarity of the oil quality to the commercial oil. This project demonstrates that HD with heat transfer control can be used by local agarwood farmers in terms of producing essential oil with high productivity, commercial standard quality but at minimum modification to current HD procedure. The proposed HTC-ed HD can be directly implemented in local agarwood plants by retrofitting of automated temperature controller and computed optimum process parameters. More work, including an independent industrial validation, a parametric heat exchanger study, a sensitivity analysis, and an optimization of heat transfer in the distiller, are planned for future studies.

Acknowledgement

This project was supported by grants from Malaysia Ministry of Higher Education (grant number RDU151007 and RDU151313).The authors thank Mr. Haslizam Abdullah, Mr. Mohd. Hafizu Zakaria, Miss Siti Rohaida and Mr Vorathin, the graduate interns under the KTP Grant RDU 151007 for their assistance in carrying out the improvement process at I Gawa Resourcse. The authors also thank Mr. Che Aizal Mohd for his expertise in GC analysis, and Pak Musa, from I Gawa Resources for his assistance in extracting Agarwood oil.

References

1. Abdullah, M.H., et al., *μltraxtor-Low Resources High Yield Benchtop Agarwood Essential Oil Extractor.*
2. Hashim, Y.Z.H.-Y., et al., *Aquilaria spp. (agarwood) as source of health beneficial compounds: A review of traditional use, phytochemistry and pharmacology.* Journal of Ethnopharmacology, 2016. **189**: p. 331-360.
3. Xu, Y., et al., *Identification of genes related to agarwood formation: transcriptome analysis of healthy and wounded tissues of Aquilaria sinensis.* BMC genomics, 2013. **14**(1): p. 227.
4. Pant, P. and R.P. Rastogi, *Agarol, a new sesquiterpene from Aquilaria agallocha.* Phytochemistry, 1980. **19**(8): p. 1869-1870.
5. Tajuddin, S.N., et al., *Characterization of the Chemical Constituents of Agarwood Oils from Malaysia by Comprehensive Two-Dimensional Gas Chromatography–Time-of-Flight Mass Spectrometry.* Mendeleev Communications, 2013. **23**(1): p. 51-52.
6. Jain, T.C. and S.C. Bhattacharyya, *Structure, stereochemistry and absolute configuration of agarol, a new sesquiterpene alcohol from agarwood oil.* Tetrahedron Letters, 1959. **1**(9): p. 13-17.
7. Wang, H.-N., et al., *Three new sesquiterpenoids from agarwood of Aquilaria crassna.* Fitoterapia, 2016. **114**: p. 7-11.
8. Chua, L.S.L., *AGARWOOD (AQUILARIA MALACCENSIS) IN MALAYSIA*, in *International Expert Workshop on CITES Non Detriment*

Findings. 2008, International Expert Workshop on CITES Non Detriment Findings: Cancun, Mexico.
9. Kusuma, H.S. and M. Mahfud, *Kinetic studies on extraction of essential oil from sandalwood (Santalum album) by microwave air-hydrodistillation method.* Alexandria Engineering Journal, 2017.
10. Jok, V.A., N.C. Radzi, and K.H.K. Hamid, *Agarwood Oil Yield As A Result of Changes in Cell Morphology Due To Soaking Process.* Procedia-Social and Behavioral Sciences, 2015. **195**: p. 2443-2450.
11. Cheah, L.C.E., *Study of extraction processes and their impact on bioactivity of botanicals*, in *Department of Pharmacy*. 2009, National University Singapore: Singapore.
12. Ahmad Fadzli, Z., *Extraction of gaharu essential oil using spinning band distillation*. 2006, Universiti Malaysia Pahang.