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Research paper

Moisture & Acetaminophen analysis on the *Aquilaria Malaccensis* dried leaves extract

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Abstract

Aquilaria malaccencis is a traditional herbal that has been long used in medicinal remedies due to its various health benefits. Acetaminophen is a synthetic drug often used to treat fever and other illness and this compound can be found in *Aquilaria malaccencis* leaves. In this work, the presence of acetaminophen was evaluated by drying the leaves at various temperatures (30- 70°C) using a fabricated vacuum far-infrared dryer. The dried leaves were then extracted using hydro distillation method to produce essential oil. The presence of acetaminophenin in the extracted sample was confirmed by using FITR. A peak at 1640 cm^{-1} region indicated that carbonyl vibration infrared band in the spectrum of acetaminophen was observed for all three of the extracted samples. This confirmed the presence of acetaminophenin *aquilaria malaccencis* leaves.

Keywords: *acetaminophen; Aquilaria Malaccensis, Moisture effect, vacuum far-infrared dryer*.

1. Introduction

Aquilaria malaccencis, also known as agar wood, is a heavy and fragrant resinous wood formed in Aquilaria tree. Aquilaria has wide range of applications and often used as incense, perfumery, medicine, religious ceremony, and as ornamentals [1]. Its numerous health benefits as a traditional herb have also led to various studies on its medicinal benefits. Studies on Aquilaria leaves have shown that it has various health properties such anti-oxidant, antimicrobial, anti-inflammatory activity and hepatoprotective [2]. Often time, this herb has been widely used to treat pain, relieve fever, rheumatism, and asthma [3]. Despite that, there is still lack of studies on the presence of acetaminophen in aquilaria leaves.

Acetaminophen is the most prescribed drug to treat pain and relieve fever [4]. A drying method of food preservation is usually used to extract the components from Aquilaria leaves. Research done by Afiffudden et.al [5] shows that the presence of 4'-hydroxyacetanilide a scientific name of acetaminophen in three types of aquilaria leaves. Previous studies showed that vacuum far-infrared (VFIR) drying does not only reduce drying time but also energy consumption and yield a better dried product quality [6]. Vacuum drying has also been known to quicken the drying process without diminishing the dried product quality [7]. However, there are little studies conducted on the effect of VFIR on Aquilaria malaccencis leaves.

Various studies have been conducted on the bioactive compounds in Aquilaria trees, which is important in discovering therapeutic agent in plant. Alkaloids, tannins, flavonoids and phenolic compounds are considered as the most significant phytochemical compounds of plants. The bioactive compounds of plants carry a significant potential in application in human healthcare[8].Often time, this herb has been widely used to treat pain, relieve fever, rheumatism, and asthma[3].

Fever is known as pyretic and it can be treated by consuming antipyretic drug. Acetaminophen is the most prescribed drug used as antipyretic and analgesic to treat pain and relieve fever [4]. Despite a decline in the use of medicinal plants in the middle of 20th century due to the rise of synthetic drugs, there is a change in trend in people's favour. Medicinal plants are gaining favour as research are focusing on natural products and these plants have long been used are a source of antipyretic agents to treat fever. Due to this matter, there is an urge for more search on potential antipyretic agents from herbal materials[9]. Despite that, there is still lack of studies on the presence of Acetaminophen in *Aquilaria* leaves.

In food preservation, the dehydration technique is probably one of the oldest methods practiced by mankind. More rapid drying techniques and methods have been created to decrease the substantial amount of energy needed in the drying process[10]. A drying method of food preservation is usually used to extract the components from *Aquilaria* leaves.

One of the commercially used drying techniques in producing dried products is convective drying using hot air. Materials are heated up with hot air to enhance mass diffusion, where water is taken out from the surface. Drying rate is limited based on water from the surface. On hot air drying, heat can influence on the physical, chemical, sensory characteristics which then causes loss in final product quality [11]. Although it involves low costs, this method requires an extended period of time and higher temperatures for completion [12] and thus, results in serious injuries including degradation of taste and colour and reduction in nutrition content, density and water absorbance capacity[13].

Far-infrared radiation (FIR) has gained popularity as a thermal energy source during the past decade for drying of many food and

agricultural materials[14]. Lower pressure level also plays significance factors to ensure faster drying of the sample as evaporation speed of the water is higher at lower pressure [13] Previous studies showed that vacuum far-infrared (VFIR) drying does not only reduce drying time but also energy consumption and yield a better dried product quality[10]. Vacuum drying has also been known to quicken the drying process without diminishing the dried product quality. Addition to that, vacuum drying also help prevent oxidation from occurring when air is present[15], [16]. However, there are little studies conducted on the effect of VFIR on *Aquilaria malaccencis* leaves.

Fourier-transform infrared (FTIR) spectroscopy has risen to be one of the major tools for a wide range of applications including analysis of small molecules or molecular complexes to the analysis of cells or tissues[17].Presently, there are only few works are available reporting the IR spectra of acetaminophen. FTIR is gaining wide interest amongst scientists due to its relatively simple, reproducible, non-destructive to the material, and only require small amounts of materialwith a minimum sample preparation. Direct information about the chemical composition can be provided as the spectral bands in vibrational spectra are not only molecule specific, but also relatively narrow, easy to resolve, and sensitive to molecular structure, conformation, and environment [18]. Therefore, the purpose of this research is to evaluate the drying effects of VFIR at various temperatures and at shorter time on the *Aquilaria malaccencis* leaves. The investigation on the presence of Acetaminophen in the leaves will be discovered.

2. Methodology

2.1. Materials

Fresh, undamaged, and matured *Aquilaria malaccencis* leaves were obtained from Jalan Kebun, Shah Alam, Malaysia. The leaves were then rinsed with tap water toremove impurities and wiped dried with clean tissueto remove the excessive moisture. The cleaned leaves were kept in a plastic container with a good lid seal and stored in a refrigerator at 4°C prior to the experiment.

2.2. Experimental Procedure

Five experimental runs were carried out to investigate the effect of vacuum far-infrared (VFIR) dryer drying on the *Aquilaria malaccencis* leaves. A fabricated VFIRwas used to dry the sample at 5 different temperatures i.e. 30°C, 40°C, 50°C, 60°C, and 70°C. The pressure set for this experiment was 0.5 bar. About 50 pcs of cleaned leaves were used for each run, placed on the tray inside of VFIR and then dried for 120 minutes to ensure the leaves were well-dried in a fixed residence time.

The leaves samples were weighed using weighing balance (Mettler Toledo) prior to the drying and after drying process to determine the moisture content removal. The removal of moisture content was determined using the following equation:

$$MC = \frac{W - W_1}{W_1}$$

$$MC_{dry\ basis} = \frac{W_{initial} - W_{dried}}{W_{dried}} \quad (1)$$

$$MC_{wet\ basis} = \frac{W_{dried} - W_{initial}}{W_{initial}} \quad (2)$$

where MC is Moisture content (g water/g solid), W is sample weight at a specified time.

Three sets of experiments were conducted for 5g, 10g, and 15g of *Aquilaria malaccencis* leaves that were to be extracted. For 5g of dried sample, 5 sets of temperature were run which were at 30°C, 40°C, 50°C, 60°C and 70°C. Meanwhile, only 3 sets of temperature were run for both 10g and 15g which are 40°C, 50°C and 60°C, due to the time constraint to remove excessive moisture at 30°C and lack of remaining fresh leaves samples. The dried samples of *Aquilaria malaccencis* leaves from the temperature of 40°C, 50°C, and 60°C were weighed accurately to different amount of 5g, 10g, and 15g.

The samples were extracted using hydrodistillation method and it was performed for 5 hours until the extraction completed. The 5L-flask containing grounded dried leaves were and distilled water was put into heating mantle and heated up to 150°C for the hydrodistillation process. After the extraction was completed, the extracted samples were collected using vials and stored in a refrigerator at temperature of 4°C. The extracted samples were put into rotary evaporator to remove the excessive water prior to FTIR analysis.

Perkin Elmer Spectrum One FT-IR Spectrometer Serial No. 74630 was used to get the FTIR analysis. The extracted samples from the hydrodistillation were analysed using this equipment to determine its functional group and the infrared band from the spectrum of acetaminophen.

3. Results & Discussion

3.1. Effect of VFIR drying on the moisture content of Aquilaria malaccencis leaves

Vacuum far-infrared drying process caused some weight loss of the leaves, showed by the difference of weight of the leaves before and after drying. The loss of weight was attributed to the loss of moisture inside the leaves after it underwent drying process. The main purpose of drying process is to remove the moisture content in order to preserve the ingredients inside the leaves and thus, retain its quality [19].

Based on the Table 1, Table 2, and Table 3, there is a similar trend observed in terms of difference of weight. The leaves that were dried at temperature of 60°C and 70°C have greater difference of weight compared to the leaves that were dried at temperature of 30°C and 40°C. For batches of 5g extraction sample, the highest percentage of weight loss is run 3 and run 5 using temperature of 50°C and 70°C respectively, with 61.93% and 61.54% loss respectively. Supposedly, the leaves at run 5 should present the highest percentage of weight loss compared to run 3 due to greater moisture removal from the leaves at higher temperature. However, the amount of leaves used for drying at run 3 is more than the other run therefore it is possible that it would have a greater weight loss.

Table 1: Effect of VFIR on *Aquilaria malaccensis* for 5g of extraction sample.

Run	Tempe-rature	Weight of leaves (g)		% Difference	Moisture content	
		Before	After		Dry basis	Wet basis
1	30°C	15.1465	13.9110	8.07	0.0878	0.0807
2	40°C	13.3185	9.2283	30.71	0.4432	0.3071
3	50°C	16.8547	6.4223	61.93	1.6269	0.6193
4	60°C	14.0937	5.6200	60.12	1.5077	0.6012
5	70°C	13.4603	5.1833	61.54	1.6001	0.6154

For batches of 10g of extraction sample, the highest percentage of weight loss is run 3 using temperature of 60°C with 63.93% loss as shown in the Table 2.

Table 3: Effect of VFIR on *Aquilaria malaccensis* for 10g of extraction sample.

Run	Tempe-rature	Weight of leaves (g)		% Difference	Moisture content	
		Before	After		Dry basis	Wet basis
1	40°C	31.8433	20.2604	36.37	0.5717	0.3637
2	50°C	29.0262	11.0717	61.86	1.6217	0.6186
3	60°C	30.4882	10.9986	63.93	1.7720	0.6393

Similarly, in Table 3 for batches of 15g of extraction sample, the highest percentage of weight loss is run 3 using temperature of 60°C with 62.40% loss. At higher temperature, the moisture removal from the leaves during drying process is greater, hence, explains the increasing trend of weight difference from lower temperature to higher temperature.

Table 3: Effect of VFIR on *Aquilaria malaccensis* for 15g of extraction sample.

Run	Tempe-rature	Weight of leaves (g)		% Difference	Moisture content	
		Before	After		Dry basis	Wet basis
1	40°C	49.7302	36.4287	26.75	0.3651	0.3675
2	50°C	46.3732	18.8439	59.36	1.4609	0.5936
3	60°C	47.6510	17.9161	62.40	1.7720	0.6240

Moisture content is the amount of moisture in the sample given as a percentage of the sample's original (wet) weight. The amount of moisture in agricultural materials and food product can affects the physical properties of the material. Moisture content of materials can be expressed in two ways: wet basis and dry basis.moisture content. To perform these calculations, it is assumed that the sample loses only water and that the weight of the dry matter remains constant.

Wet basis moisture content is defined by the percentage equivalent of the ratio of the weight of water to the total weight of the material. It is often used to define the water content of agricultural material and food products. Based on the Table 1, Table 2, and Table 3, an increasing trend can be observed as the temperature used for drying process increases. For batches of 5g extraction sample, run 3 and run 6 have the highest wet basis moisture content, at 0.619329 and 0.615403 respectively using temperature of 50°C and 70°C respectively. As been mentioned before, this outcome may result from the difference in initial weight of leaves used for drying process between these two runs.

A similar increasing trend in wet basis moisture content can be observed for both batches of 10g of extraction sample and 15g of extraction sample. The highest wet basis moisture content for both batches of extraction sample is run 3 using temperature of 60°C with values of 0.639251 for 5g and 0.624036 for 10g. The increasing values indicate that at higher temperature, the amount of water per unit mass of moist (wet) sample is larger.

Dry basis moisture content is defined by the percentage equivalent of the ratio of the weight of water to the weight of dry matter. It is commonly used for describing moisture changes during drying. For batches of 5g extraction sample, the highest dry basis moisture content is run 3 with value of 1.626941 using temperature of 50°C.

Theoretically, dry basis moisture content changes linearly with the weight loss or gain when a sample loses or gains moisture. During drying process, moisture is loss and the amount of moisture removed from sample is higher with increasing temperature. Supposedly for this experiment, run 5 should have the highest dry basis moisture content as it is using the highest drying temperature, 70°C. However, during this experiment, the difference in initial weight of leaves sample might have affected the result for dry basis moisture content. The differences in sizes and shapes, including maturity of the leaves sample might also affected the result obtained.

This consideration may also apply for batches of 5g and 10g of extraction sample since it showed similar trend of increasing dry basis moisture content. From the result tabulated in Table 2 and Table 3, the highest dry basis moisture content for both batches of 5g and 10g are 1.772007 and 1.772007 respectively, using drying temperature of 60°C. As been mentioned before, the differences in initial weight attributed by the differences in sizes and shapes, including the maturity of the leaves; may have affected the results obtained.

3.2. Characterization of Acetaminophen using Fourier-transform Infrared Spectroscopy (FTIR)

Fig. 1 shows the FTIR spectra *Aquilaria malaccencis* leaves extractafter being dried using VFIR at temperature of 40°C, 50°C, and 60°C and extracted using hydrodistillation. From the spectral data, there was a strong peak in 3200-3400 cm^{-1} region which showed the infrared bands of water. Since water has a broad band, it has a very strong absorption in the 3700-3100 cm^{-1} region. The broad band of water was due to the extraction method using water as solvent in the hydrodistillation.

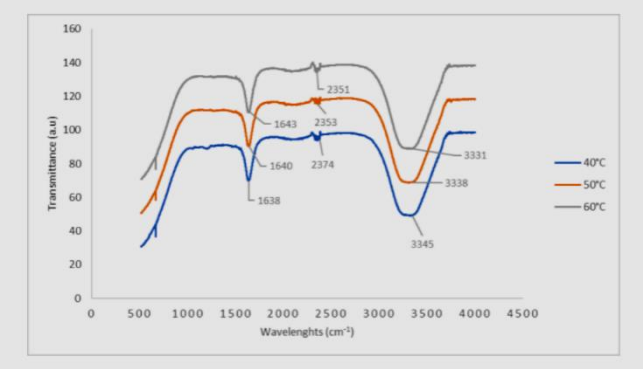

Fig. 1: FTIR Spectrum for *Aquilaria Malaccensis* extracts

Another peak from the spectral data is at 1640 cm^{-1} region which can be observed for all three of the extracted sample. For 5g of extracted sample dried at temperature of 40°C, 50°C, and 60°C, the peak is observed at 1638 cm^{-1}, 1640 cm^{-1}, and 1643 cm^{-1} respectively. This infrared band is estimated and can be identified in the spectrum of acetaminophen which is the carbonyl vibration. Carbonyl vibration infrared band can be identified at 1637.2 cm^{-1} in the spectrum of acetaminophen [20].

Other major bands identified in the spectrum of acetaminophen were the aromatic ring vibration at 1517.5 cm^{-1} and the OH deformation-C-O stretch at 1243.2 cm^{-1}. Other bands that can also be identified in the spectrum of this compound is H3C-C=O stretching at 1378.4 cm^{-1}, NH bending at 1447.9 cm^{-1} and CNH vibration at 1552.2 cm^{-1}[20].

From the Fig. 1, only infrared band of carbonyl vibration can be observed and identified in the spectrum of acetaminophen. The lack of other major bands presence from this spectrum might due to lack of sufficient essential oil extracted from the dried leaves samples. Most of the extracted samples contained water from the hydrodistillation extraction and contain only little of essential oil. The reason for this matter might due to the fact that the capacity used for the hydrodistillation extraction process was too large which was at 5L capacity instead of 2L capacity. Since the capacity for the extraction was too large, the amount of water used for hydrodistillation is larger compared to the amount of dried leaves samples that were to be extracted. Therefore, there were only little amount of oil produced and may not be sufficient enough for characterisation of Acetaminophen using FTIR.

4. Conclusions

From the data tabulated in Table 1, 2, and 3 from Results and Discussion section, it is clear that *Aquilaria malaccencis* leaves dried at higher temperature had greater moisture loss. The leaves samples that are dried at 60°C had the highest weight loss difference, which indicates the most removal of moisture from the leaves. At higher temperature, the dried products should also have the highest value of wet basis moisture content and the lowest value of dry basis moisture content as rate of moisture reduction of the leaves increases.

Extraction method is needed in order to analyse phytochemical content from the leaves samples. The extracted samples then can be analysed using FTIR since it allows investigation of functional groups, bonding types, and provide molecular-level information. Based on the spectral data obtained from using FTIR, presence of acetaminophen in dried extracts of *A. malaccencis* leaves can be determined. After drying, few infrared bands from the spectrum of acetaminophen can be identified from the spectral data. However, FTIR analysis on extract samples of fresh *A. malaccencis* leaves presented more identified peaks that show infrared bands in the spectrum of acetaminophen. The infrared bands that can be identified from the spectral data belonging in the spectrum of acetaminophen are carbonyl vibration, NH bending and C-NH bending. The presence of these infrared bands indicates the presence of acetaminophen in *Aquilaria malaccencis* leaves.

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References

[1] A. Z. Adam, S. Y. Lee, and R. Mohamed, "Pharmacological properties of agarwood tea derived from Aquilaria (Thymelaeaceae) leaves: An emerging contemporary herbal drink," J. Herb. Med., no. November 2016, 2017.

[2] Y. Z. H. Y. Hashim, P. G. Kerr, P. Abbas, and H. Mohd Salleh, "Aquilaria spp. (agarwood) as source of health beneficial compounds: A review of traditional use, phytochemistry and pharmacology," J. Ethnopharmacol., vol. 189, pp. 331–360, 2016.

[3] M. Samadi, Z. Z. Abidin, R. Yunus, D. R. Awang Biak, H. Yoshida, and E. H. Lok, "Assessing the kinetic model of hydrodistillation and chemical composition of Aquilaria malaccensis leaves essential oil," Chinese J. Chem. Eng., vol. 25, no. 2, pp. 216–222, 2017.

[4] G. M. Pacifici and K. Allegaert, "Clinical Pharmacology of Paracetamol in Neonates: A Review," Curr. Ther. Res. - Clin. Exp., vol. 77, pp. 24–30, 2015.

[5] Afiffudden, S. K. N, Alwi, H & Ku Hamid, K. H., (2015) Determination of 4'-Hydroxyacetanilide in Leaves Extract of Aquilaria malaccencis by High Pressure Liquid Chromatograph,Procedia -Social and Behavioral Sciences, Vol 195, pp 2726-2733

[6] S. Mongpraneet, T. Abe, and T. Tsurusaki, "Far Infrared --Vacuum and - Convection Drying of Welsh Onion," Trans. ASAE, vol. 45, no. 5, pp. 1529–1535, 2002.

[7] C. Nimmol, "Vacuum Far-infrared Drying of Foods and Agricultural Materials," J. KMUTNB, vol. 20, no. 1, pp. 37–44, 2010.

[8] K. B. Khalil, A. S.; Rahim, A. A., Taha, K. K., Abdallah,"Characterization of Methanolic Extracts of Agarwood Leaves," *J. Appl. Ind. Sci.*, vol. 1, no. 3, pp. 78–88, 2013.

[9] S. Sultana, H. M. Asif, N. Akhtar, and K. Ahmad, "Medicinal plants with potential antipyretic activity: A review," *Asian Pacific J. Trop. Dis.*, vol. 5, no. S1, pp. S202–S208, 2015.

[10] S. Mongpraneet, T. Abe, and T. Tsurusaki, "Far Infrared --Vacuum and - Convection Drying of Welsh Onion," *Trans. ASAE*, vol. 45, no. 5, pp. 1529–1535, 2002.

[11] Bai-Ngew, S., Therdthai, N., Dhamvithee, P., & Zhou, W. (2015). Effect of microwave vacuum drying and hot air drying on the physicochemical properties of durian flour. *International Journal of Food Science and Technology*, *50*(2), 305–312. https://doi.org/10.1111/ijfs.12651

[12] Wojdyło, A., Figiel, A., Lech, K., Nowicka, P., & Oszmiański, J. (2014). Effect of Convective and Vacuum-Microwave Drying on the Bioactive Compounds, Color, and Antioxidant Capacity of Sour Cherries. *Food and Bioprocess Technology*, *7*(3), 829–841. https://doi.org/10.1007/s11947-013-1130-8

[13] Liu, Y., Miao, S., Wu, J., Liu, J., Yu, H., & Duan, X. (2015). Drying Characteristics and Modeling of Vacuum Far-Infrared Radiation Drying of Flos Lonicerae. *Journal of Food Processing and Preservation*, *39*(4), 338–348. https://doi.org/10.1111/jfpp.1223

[14] X. Ning, J. Lee, and C. Han, "Drying characteristics and quality of red ginseng using far-infrared rays," *J. Ginseng Res.*, vol. 39, no. 4, pp. 371–375, 2015.

[15] C. Nimmol, "Vacuum Far-infrared Drying of Foods and Agricultural Materials," *J. KMUTNB*, vol. 20, no. 1, pp. 37–44, 2010.

[16] T. Swasdisevi, S. Devahastin, P. Sa-Adchom, and S. Soponronnarit, "Mathematical modeling of combined far-infrared and vacuum drying banana slice," *J. Food Eng.*, vol. 92, no. 1, pp. 100–106, 2009.

[17] C. Berthomieu and R. Hienerwadel, "Fourier transform infrared (FTIR) spectroscopy," *Photosynth. Res.*, vol. 101, no. 2–3, pp. 157–170, 2009.

[18] Z. Movasaghi, S. Rehman, and I. U. Rehman, "Fourier transform infrared (FTIR) spectroscopy of biological tissues," *Appl. Spectrosc. Rev.*, vol. 43, no. 2, pp. 134–179, 2008.

[19] H. Alwi, S. A. Ali, K. Halim, K. Hamid, and M. Zaid, "Drying effects of vacuum far-infrared on aquilaria malaccensis leaves Drying effects of vacuum far-infrared on aquilaria malaccensis leaves," 2017.

[20] M. L. Ramos, J. F. Tyson, and D. J. Curran, "Determination of acetaminophen by flow injection with on-line chemical derivatization: Investigations using visible and FTIR spectrophotometry," Anal. Chim. Acta, vol. 364, no. 1–3, pp. 107–116, 1998