

Yield Study for Extracted Tea Leaves Essential Oil Using Microwave-Assisted Process

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There have been developments on the use of microwave to accelerate the digestion of solid materials, the heating of materials, and, more recently, the extraction of solute from solids via solvent as extracting medium. Microwave-assisted process (MAP) was used to accelerate the extraction of target compounds. It can be used for the extraction of compounds from various plant and animal tissues or of undesirable components from raw materials. Tea tree leaves were used in this study to investigate the applicability of microwave irradiation for essential oil extraction. The microwave parameters studied were tea tree leaves/ethanol ratio as well as required dose of microwave and time of irradiation. Different ratios of tea tree leaves/ethanol had been examined in order to obtain the optimal feed/solvent ratio that would give the highest yield of extracted essential oil. The required number of microwave doses that provided an accomplished extraction process had been ascertained. The optimal time of microwave exposure was found to be at 3 min. The measurements of extracted tea tree essential oil constituents (i.e., cineole, α -pinene, and γ -terpinene) that represent the major constituents were performed using gas chromatography (GC) analysis to estimate the yield of extracted tea tree essential oil.

Keywords: Essential oil, ethanol, extraction, microwave-assisted process (MAP), and tea tree leaves.

INTRODUCTION

Essential oils, like traditional herbal remedies, are very valuable yet are simple and easy-to-use for self-medication and first aid because of their effectiveness in the prevention and treatment of many common conditions.

Essential oils can be broadly defined as volatile oils that differ fundamentally from fixed fatty oils, such as those from linseed, coconuts, and olives, in being more mobile and volatile. Many essential oils, aside from being used in first aid or for treatment of common complaints, are also ideal for bath oils,

perfumes, or room fresheners. Even when they are used purely for esthetic purposes, they still fulfill a positive preventive and therapeutic role (Lawless 1997).

The conventional essential oils extraction methods includes tumbling and shaking, oil infusion, cold expression, steam distillation, and Soxhlet extraction, which can take from hours to days and require significant amounts of solvent. The review of microwave extraction studies discussed in the following paragraphs shows that extraction using microwave technology is a good alternative to conventional extraction techniques.

Extraction by microwave as a new technique has its own specific parameters that need to be characterized for every plant that contains essential oil. Microwave applicability for the extraction of various types of compounds from plant, food, and soil had been investigated by Ganzler et al. (1986). The production of volatile material from plant exposed to microwave energy in an air stream had been discussed by Craveiro et al. (1989).

Chen and Spiro (1994) studied the microwave heating characteristics of extracting oils from rosemary and peppermint leaves suspended in hexane, ethanol, and mixture of hexane and ethanol. Pare (1995) patented a general extraction method for biological matter using microwave energy and a microwave transparent solvent.

Whish et al. (1996) studied the influence of the dryness of tea leaves on the oil content yield using distillation process. It was found that drying tea leaf on the stem increased the oil content yield from 5.8 to 7.4 % volume/dry weight. This increase was not a result of changing moisture content but appeared to be due to an active form of postharvest oil uptake or production. These results showed that the need for immediate distillation of cut foliage would be unnecessary, since the delay would actually improve the yield rather than reduce it.

Weseler et al. (2002) studied the antibacterial activity of Australian tea tree oil against several strains of *Malassezia pachydermatis*. Different solvents, such as ethanol, trichloromethane, cyclohexane, and *n*-hexane, were used to study the effect of solvency on the extraction of artemisinin from *Artemisia annua* using microwave-assisted process (MAP) by Jin-yu Hao et al. (2002).

Marie et al. (2004) used a new microwave technique—solvent-free microwave extraction (SFME)—to extract essential oil from basil (*Ocimum basilicum* L.), garden mint (*Mentha crispa* L.), and thyme (*Thymus vulgaris* L.). The findings showed an aromatic profile of microwave exposure similar to those obtained by hydrodistillation at 4.5 h.

The present work is concerned with the optimization of three microwave parameters: tea tree/ethanol ratio, required dose of microwave, and time of exposure. Different ratios of tea tree

leaves/ethanol were examined, the required number of microwave doses that would provide an accomplishing extraction process was ascertained, and the optimal time of microwave exposure was determined.

EXPERIMENTAL PROCEDURES

Materials and chemicals

Standard samples of the major tea tree constituents (i.e., cineole, α -pinene, and γ -terpinene) of high purity purchased from Sigma-Aldrich were used in this study for Gas Chromatography (GC) calibration. The calibration was done by preparing known concentrations of each component in ethanol solution. Tree tree leaves (Figure 2) were supplied by Nasuha Enterprise Plantation (Malaysia), while ethanol of 96 v/v % was supplied by Fluka—Switzerland.



Figure 1. Tea Tree



Figure 2. Tea Tree Leaves

Equipment

In this study, GC (Perkin Elmer-Auto System XL Gas Chromatograph) was utilized for extracts analysis. Computerized microwave solvent extraction system (Milestone-Ethos Sel microwave lab station) was used for microwave extraction, as shown in Figures 3 and 4.



Figure 3. Microwave Oven

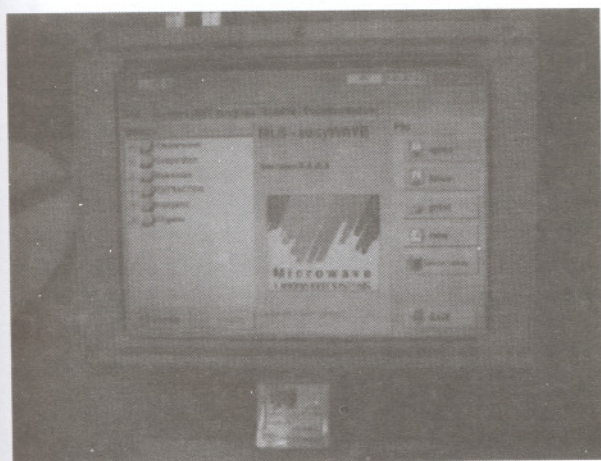


Figure 4. Lab Terminal Controller

Procedure

In order to measure the optimal ratio, required doses, and time, the following procedure was followed: samples of tea tree leaves were placed in sealed containers of 270 mL each. The same volume of solvent was used throughout the tests, 100 mL ethanol. A magnetic stirrer was used to agitate the solution. In between each dose of exposure, the solutions were allowed to cool back

to room temperature (28°C). Sampling of 2 mL was carried out at the end of each dose of exposure; thus, the solute concentrations were corrected taking into account the change in solution volume. The extraction was done in sealed vessels; no evaporation was observed. All solute concentrations were reported in mg/L. A power of 1,000 W and an exposure time of 60 sec were used in all the experiments. Eq. (1) was used to correct the concentration due to sampling:

$$C = C_n (V - 2 [n-1]) / V \quad (1)$$

where:

- C = corrected concentration (mg/L)
- C_n = concentration obtained directly from chromatogram (mg/L)
- N = number of sampling
- V = initial volume of solvent (100 mL)

RESULTS AND DISCUSSIONS

Influence of tea tree/ethanol ratio on microwave extraction

Tables 1 to 5, on the one hand, show the constituents concentrations for the different ratios of tea tree to ethanol at the same conditions of time and power. Figures 5 to 7, on the other hand, show the concentrations of each constituent at different ratios and for ten successive doses.

All these tables and figures underscore the increasing trend in the amount of extracted constituents, namely cineole, α -pinene, and γ -terpinene, that microwave extraction yielded with each increase in exposure dose.

The results showed that maximum extraction was obtained at a ratio of 6 g tea tree leaves to 100 mL ethanol, wherein the essential oil solution reached saturation. Beyond this ratio, however, the extraction would no longer be economical. The obtained yield from fresh tea tree leaves was 11.11 mg/g. The percentages for each constituent were 54.3% γ -terpinene, 35% α -pinene, and 10.7% cineole.

Table 1. Constituent Concentration (mg/100 mL) at 2 g Tea Tree/100 mL Ethanol, 60 sec Exposure, and 1000 W

Constituents	No. of Doses									
	1	2	3	4	5	6	7	8	9	10
cineole	1.20	1.28	1.38	1.45	1.58	1.77	1.96	2.1	2.38	2.4
α -pinene	2.40	3.00	3.60	4.20	5.00	5.70	6.30	6.8	7.66	7.8
γ -terpinene	5.90	6.60	7.30	8.10	8.80	9.40	10.5	11.0	12.0	12.1

Table 2. Constituent Concentration (mg/100 mL) at 4 g Tea Tree/100 mL Ethanol, 60 sec Exposure, and 1000 W

Constituents	No. of Doses									
	1	2	3	4	5	6	7	8	9	10
cineole	2.3	2.48	2.6	2.64	3.0	3.55	3.84	4.1	4.6	4.7
α -pinene	4.7	5.90	7.2	8.10	10.3	11.20	12.20	13.2	15.3	15.5
γ -terpinene	11.6	12.80	14.3	15.90	16.4	18.20	20.60	22.0	23.8	24.0

Table 3. Constituent Concentration (mg/100 mL) at 6 g Tea Tree/100 mL Ethanol, 60 sec Exposure, and 1000 W

Constituents	No. of Doses									
	1	2	3	4	5	6	7	8	9	10
cineole	3.40	3.7	3.8	4.0	4.4	5.0	5.78	6.2	7.0	7.2
α -pinene	6.80	8.8	10.4	12.3	14.8	16.9	18.5	19.8	23.1	23.3
γ -terpinene	16.90	18.	21.5	23.9	26.1	28.7	30.8	33.3	36.0	36.2

Table 4. Constituent Concentration (mg/100 mL) at 8 g Tea Tree/100 mL Ethanol, 60 seconds exposure, and 1000 W

Constituents	No. of Doses									
	1	2	3	4	5	6	7	8	9	10
cineole	3.9	4.1	4.4	4.8	5.2	6.1	6.44	7.1	7.78	7.8
α -pinene	8.7	10.9	13.2	15.7	18.9	21.2	23.2	26.8	28.5	28.8
γ -terpinene	21.6	24.8	27.3	30.1	33.4	35.2	37.8	41.0	44.6	44.7

Table 5. Constituent Concentration (mg/100 mL) at 10 g Tea Tree/100 mL Ethanol, 60 sec Exposure, and 1000 W

Constituents	No. of Doses									
	1	2	3	4	5	6	7	8	9	10
cineole	3.2	3.3	4.0	4.2	5.0	6.0	6.14	7.0	7.3	7.4
α -pinene	8.3	10.0	12.7	15.1	18.3	21.2	23.0	26.3	28.0	28.1
γ -terpinene	20.8	23.0	28.6	29.1	32.3	34.0	36.5	40.0	43.1	43.2

Influence of exposure doses on microwave extraction

The microwave extraction technique works by exposing the material to a sudden heat caused by the microwave field. To integrate this process successive exposure and cooling is required. After each microwave exposure, the solution was allowed to cool to room temperature

(28°C). The natural cooling process helped release more solutes from the solid matter and allowed the solute-contained gland to return to its original size.

Tables 1 to 5 show that ten doses of exposure are required due to the adjacent values of the obtained concentrations at 9 and 10 doses. Hence, the required number of successive

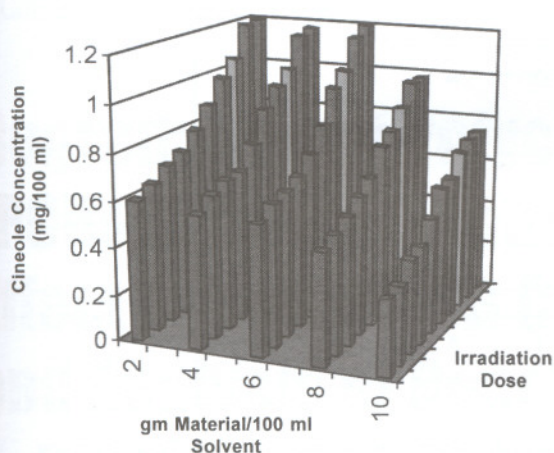


Figure 5. Cineole Cumulative Concentration—Microwave Dose Plot of 1000W and 60 sec at Different Tea Tree/Ethanol Ratios

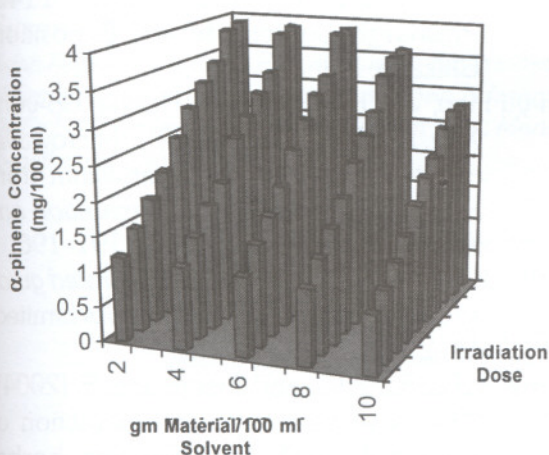


Figure 6. α -pinene Cumulative Concentration—Microwave Dose Plot of 1000 W and 60 sec at Different Tea Tree/Ethanol Ratios

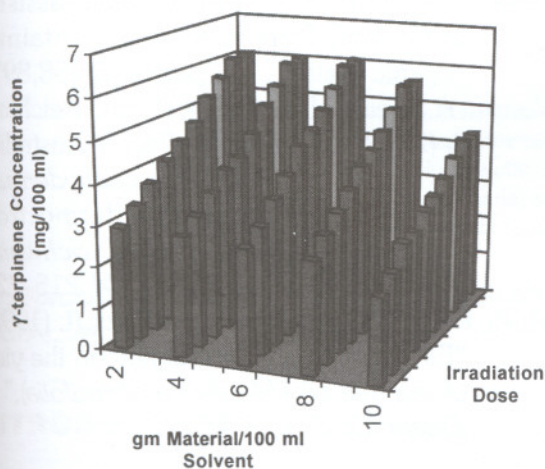


Figure 7. γ -terpinene Cumulative Concentration—Microwave Dose Plot of 1000 W and 60 sec at Different Tea Tree/Ethanol Ratios

Hence, the required number of successive exposures and cooling for an accomplishing extraction process to achieve the equilibrium of extraction is ten.

Influence of microwave exposure time on tea tree essential oil extraction

A microwave exposure time range of 1–5 min was used in the investigation of the effect of irradiation time on essential oil extraction. The solution temperature was recorded after microwave exposure at different exposure times for a sample ratio of 6 g tea tree leaves to 100 mL ethanol, 1,000 W, and 10 microwave doses.

Figure 8 illustrates significant increments in the concentration of γ -terpinene and α -pinene for an exposure time of 1–3 min, while their curves approached plateau at an exposure time exceeding 3 min. For cineole, the constituent with lowest concentration in tea tree essential oil, the curve reached plateau from the first minute of irradiation.

Tea tree essential oil yield was 26.8 mg/g tea tree fresh leaves with the constituent percentages at 53.5% γ -terpinene, 35.7% α -pinene, and 10.7% cineole.

These results indicated that to ensure the maximum yield of essential oil extracted from tea tree leaves, 3 min of exposure time should be adopted during the microwave extraction.

The essential oil yield attained by this study is higher than that reported by Jessie et al. (1994), which was 24.0 mg/g of essential oil acquired by steam distillation from fresh tea tree leaves.

To a plateau curve for a time beyond 3 min may be ascribed the greater loss of moisture content at a longer exposure time; subsequently, a drop in ΔT results from the microwave exposure which eventually slows down the extraction.

The molecular movement that promotes the extraction process under microwave exposure is much stronger in the presence of water and other solvents with high dipole movement.

CONCLUSIONS

Microwave-assisted extraction is a potential method for essential oils production because it employs sudden and uniform heat propagation

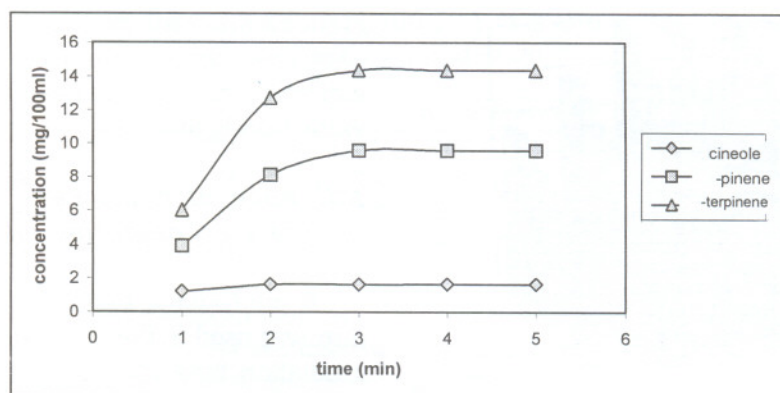


Figure 8. Tea Tree Essential Oil Constituent Concentrations per Gram of Tea Tree Leaves at Different Microwave Time Exposures

as a result of microwave application to enhance essential oil yield.

Material-solvent ratio has an important influence on microwave extraction of tea tree leaves in ethanol.

In particular, the results of this study showed that the:

- ratio of 6 g tea tree leaves to 100 mL ethanol obtained the highest concentration;
- required number of exposure doses to achieve the equilibrium of extraction was 10;
- 3 min of microwave exposure time was good enough for microwave extraction process to give optimum yield; and,
- yield of tea tree essential oil was 26.8 mg/g fresh leaves.

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