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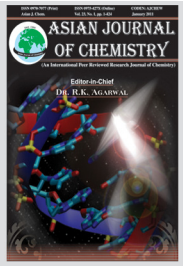

Microwave-Assisted Hydrodistillation of Essential Oil from *Thymus vulgaris* L.

P. ABEROOMAND AZAR[1,*], A. PORGHAM-DARYASARI[1], M. SABER-TEHRANI[1] and M. SOLEIMANI[2]

[1]Department of Chemistry, Science and Research Branch, Islamic Azad University, P.O. Box 14155/4933, Tehran, Iran
[2]Department of Chemistry, Faculty of Science, Islamic Azad University, Lahijan Branch, P.O. Box 1616, Lahijan, Iran

*Corresponding author: Tel: +98 21 44817146; E-mail: pabroomand@yahoo.com

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Microwave-assisted hydrodistillation was used to isolate an essential oil from the aerial parts of *Thymus vulgaris* L. and the results compared with those obtained by conventional hydrodistillation.The essential oils extracted by microwave-assisted hydrodistillation for 20 min were similar to those obtained by hydrodistillation for 3.5 h. The microwave-assisted hydrodistillation method yields an essential oil with higher amounts of more valuable oxygenated compounds and allows substantial savings of costs, in terms of time, energy and plant material.

Key Words: Essential oil, *Thymus vulgaris* L., Microwave-assisted hydrodistillation.

INTRODUCTION

The genus thymus consists of about 215 species that Mediterranean region can be described as the center of the genus[1]. Fourteen species of the genus thymus found in Iran that four of which are endemic[2].

Thymus vulgaris L. (common *Thyme*) is member of the labiatae family that has antiseptic, antimicrobial and antioxidant effect[3-6]. Many studies on composition of essential oils from different thymus species have been carried out. The results indicated that major volatile constituents from aerial parts of the plant are thymol, *p*-cymene, γ-terpinene, β-caryophyllene[7-11]. Various different methods can be used for the extraction of essential oils, *e.g.* hydrodistillation, steam distillation and simultaneous distillation-extraction[12,13]. Use of microwaves as an alternate extraction technique was first introduced by Ganzler *et al.*[14]. Microwave-assisted extraction is one way of using microwave in an extraction process where a higher extraction rate along with a lower cost of operation[15].

The aim of this work is to investigate the composition of essential oil from aerial parts of *Thymus vulgaris* L. obtaind by microwave-assisted hydrodistillation and to compared with the conventional hydrodistillation.

EXPERIMENTAL

The aerial parts of *Thymus vulgaris* L. were collected from northeastern of Iran, located in height of Binaloud Mountain, in June 2009. The plants were identified and authenticated by Dr. Mozaffarian at the herbarium of the Research Institute of Forest and Rangelands (TARI), Tehran, Iran.

Extraction of the essential oil: For hydrodistillation, 100 g the air-dried aerial parts of *Thymus vulgaris* L. in 2 L flask were subjected to hydrodistillation for 3.5 h using a Clevenger type according to the standard procedure described in the European pharmacopeia. In microwave-assisted hydrodistillation, 50 g the air-dried aerial parts of *Thymus vulgaris* L. were placed in a 1 L flask containing deionized water (1000 mL) and the flask was setup within the microwave oven cavity and a condenser was used on the top (outside the oven) to collect the extracted essential oils. The microwave oven was operated at 600 W power levels for 20 min. After trapping the oil with *n*-hexane (Merck-Germany) decanting and drying of the oil over anhydrous Na_2SO_4 (Merck-Germany), the corresponding pale-yellow oil isolated and was stored at 4 ºC until analyzed.

Microwave apparatus was a millestone "Microsynth" microwave oven that had a multimode microwave reactor 2.45 GHz with a maximum delivered power of 1500 W variable in 1 W increments. Temperature was monitored by an external fiber optic sensor. A cooling system outside the microwave oven condenses the distillate continuously. Excess water was refluxed to the extraction vessel in order to restore the water to the plant materials. The dimensions of the PTFE coated cavity are 55 cm × 55 cm × 55 cm. During experiments; time, temperature, pressure and power were controlled with the

"easy-control" software package. Temperature was monitored by a shielded thermocouple (ATC-FO) which inserted directly into the sample container and was controlled by a feedback to the microwave power regulator without using correction factors.

GC analysis: Analysis of the oil was carried out on a Hewlett-Packard-6890 gas chromatograph equipped with a split/splitless (20:1) injector (250 °C) and a flame ionization detector (250 °C). N_2 was used carrier gas (1 mL/min).The capillary column used was DB-5 (30 m × 0.25 mm, 0.32 μm film thickness).The oven temperature was held at 60 ºC for 3 min, then heated to 220 °C with a 5 °C rate and kept constant at 220 °C for 5 min. Quantitative data were obtained from GC (FID) area per cent.

GC/MS analysis was performed using a Hewlett-Packard 6890/5973 GC/MS equipment whit a 30 m × 0.25 mm, film thickness 0.32 μm HP-5MS column. Helium (99.999 %) was used as carrier gas (1.0 mL/min). The temperature program was as GC. MS spectra were taken at 70 eV. The GC/MS was equipped with chemstation software and Wiley 275 library. Identification of constituents of the oil was made by comparison of their mass spectra and retention indices (RI) with those given in the literature and authentic samples[16].

Statistical analysis: All extractions with hydrodistillation and microwave-assisted hydrodistillation were performed in duplicate and a general linear model (GLM) procedure from SAS (Statistical Analysis Software, version 9.1, SAS Institute Inc., Cary, NC, USA) was used to compare among the means.

RESULTS AND DISCUSSION

Comparison of extraction yield and time: One of the advantages of the microwave-assisted hydrodistillation is rapidity. The extraction temperature is equal to the boiling point of water at atmospheric pressure for both the microwave-assisted hydrodistillation and hydrodistillation method. To reach the extraction temperature and obtain the first essential oil droplet, it is necessary to heat for 5 min with microwave-assisted hydrodistillation compared with 0.5 h for hydrodistillation. This is due to the more efficient heat flow involved with microwaves. Unlike the classical conductive heating methods; microwaves can heat the entire sample almost simultaneously and at a higher rate[17]. Full recovery of essential oils was achieved within the first 15 min of operation with microwave-assisted hydrodistillation. In the case of hydrodistillation, a time period of at least 3.5 h was necessary for such purpose. As is shown in Table-1, an extraction time of 20 min with microwave-assisted hydrodistillation provides yields comparable to those obtained after 3.5 h by means of hydrodistillation.

Composition of essential oil: The identified constituents in the extracted essential oils from both methods are shown in Table-1. For hydrodistillation and microwave-assisted hydrodistillation 21 and 23 components, respectively were characterized, according for 98.2 and 98.8 % of the oil. In the hydrodistillation technique, the oil consists of 59.8 % oxygenated compounds and 34.8 % monoterpene hydrocarbons with thymol (43.8 %), *p*-cymene (21.1 %), γ-terpinene (6.6 %), carvacrol (5.7 %) as the major component in the oil.

TABLE-1
CHEMICAL COMPOSITION OF ESSENTIAL OILS OBTAINED BY HYDRODISTILLATION (HD) AND MICROWAVE-ASSISTED HYDRODISTILLATION (MAHD) of *T. vulgaris* AERIAL PARTS

No.	Compound[a]	RI[b]	Relative peak area (%)[c]	
			HD (%)	MAHD (%)
1	α-Thujene	930	1.7±0.0	0.8±0.2
2	α-Pinene	940	1.2±0.2	0.5±0.3
3	Camphene	957	1.1±0.1	0.4±0.0
4	1-Octen-3-ol	974	0.4±0.4	0.9±0.4
5	Myrcene	989	1.2±0.1	0.9±0.5
6	α-Terpinene	1022	1.4±0.2	0.8±0.2
7	*p*-Cymene	1030	21.1±0.0	17.3±0.3
8	Limonene	1035	0.5±0.2	0.4±0.1
9	1,8-Cineol	1040	0.4±0.0	0.8±0.1
10	γ-Terpinene	1064	6.62±0.1	3.2±0.2
11	*cis*-Sabinene hydrate	1075	0.9±0.3	2.1±0.0
12	Linalool	1101	2.4±0.0	4.0±0.2
13	*trans*-Sabinene hydrate	1108	–	0.6±0.0
14	Camphor	1164	0.4±0.1	0.7±0.4
15	Borneol	1184	2.1±0.0	3.4±0.2
16	4-Terpineol	1193	0.6±0.3	0.8±0.1
17	Estragole	1210	–	8.1±0.3
18	Thymol methyl ether	1243	0.8±0.3	0.8±0.0
19	Carvacrol methyl ether	1254	0.9±0.1	1.1±0.1
20	Thymol	1300	43.8±0.2	43.3±0.5
21	Carvacrol	1310	5.7±0.1	5.5±0.4
22	β-Caryophyllene	1455	3.6±0.1	1.3±0.2
23	Caryophyllene oxide	1628	1.4±0.2	1.1±0.3
Total oxygenated fraction (%)			59.8	73.2
Total peak area (%)			98.2	98.8
Total extraction time (min)			210	20
Yield (%)			1.5±0.2	1.6±0.1

[a]Compounds listed in order of elution.
[b]RI, Kovat's indices as determined on a DB-5 column using the homologous series of *n*-hydrocarbons (C9-C19).
[c]Mean ± SD (n = 2), In each component identified, the means are not significantly different ($p > 0.05$) for each component identified.

Comparatively the main constituent of the oil in microwave-assisted hydrodistillation technique were thymol (43.3 %), *p*-cymene (17.3 %), estragole (8.1 %), carvacrol (5.5 %). Furthermore the oil consists of 73.2 % oxygenated compounds and 24.3 % monoterpene hydrocarbons.

Conclusion

Microwave-assisted hydrodistillation offer substantial advantages over conventional hydrodistillation. Essential oils obtained by hydrodistillation and microwave-assisted hydrodistillation are very close in their compositions but the time required for microwave-assisted hydrodistillation was much shorter than hydrodistillation and allowed substantial saving in energy. Higher amounts of oxygenated compounds and lower amounts of monoterpene hydrocarbons are present in the essential oil extracted by microwave-assisted hydrodistillation in comparison with hydrodistillation. Monoterpene hydrocarbons are less valuable than oxygenated compounds in terms of their contribution to the fragrance of the essential oil[18]. Conversely, the oxygenated compounds are highly odoriferous and hence, the most valuable. All this advantages make microwave-assisted hydrodistillation a good alternative for the extraction of essential oil from plants.

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