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Short communication

Solvent-free microwave extraction of essential oils from *Thymus vulgaris* L. and *Melissa officinalis* L.

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ABSTRACT

The aim of this study was the comparison of essential oil of two herbs from lamiaceae family namely Thyme and Melissa via solvent-free microwave extraction (SFME) and hydrodistillation (HD). The process of SFME was optimized by central composite design (CCD) and response surface methodology (RSM). The essential oils were analyzed by gas chromatography–mass spectrometry (GC–MS). The compounds identified in higher relative abundance, for Thyme were, Thymol (HD: 28.92%, SFME: 32.17%), Carvacrol (HD: 26.18%, SFME: 29.25%), *p*-Cymene (HD: 13.25%, SFME: 9.51%) and γ-Terpinene (HD: 5.09%, SFME: 3.62%). In regard to Melissa oils, Caryophyllene oxide (HD: 23.71%, SFME: 24.95%), E-Citral (HD: 19.53%, SFME: 24.19%), Z-Citral (HD: 16.39%, SFME: 18.45%) and α-Muurolene (HD: 12.17%, SFME: 12.50%) were the most abundant compounds. The results showed that SFME was excellent in terms of saving energy and extraction time (42 min, compared to 4 h in HD). The research suggests that SFME is an outstanding alternative for extraction essential oils in industrial applications.

1. Introduction

Thyme (*Thymus vulgaris* L.) and Melissa (*Melissa officinalis* L.) are aromatic-medicinal species that belong to lamiaceae family. A large range of medical uses for essential oils from these plants have been reported. These include antibacterial, antitumor, antifungal and antioxidant properties (Pereira et al., 2014; Duda et al., 2015; Shakeri et al., 2016; Boubaker et al., 2016). Essential oil composition and the ratio between different oil constituents play a role in their bioactivity. Besides, Essential oil composition depends on various environmental and genetic factors, as well as on postharvest processing factors (Jordán et al., 2006; Lemos et al., 2017).

solvent-free microwave extraction (SFME) is an innovative, clean and environmental friendly technique with particular interest for the extraction of essential oil from herbs or plants. Isolation and concentration of volatile compounds are performed by a single stage (Tigrine-Kordjani et al., 2006; Li et al., 2013; Filly et al., 2014). The SFME apparatus, a combination of microwave heating and dry distillation, is carried out at atmospheric pressure. The process is based on heat generated by ionic conduction and dipole rotation. The extraction occurs when the water inside the plant absorbs energy coming from microwave and increase pressure inside the material causing the cell structure to break (Lucchesi et al., 2004; Bayramoglu et al., 2008).

In this paper, we present a comparative study of the ability of two different techniques to extract the essential oils from Thyme and Melissa leaves to find the most advantageous in term of essential oil quantity and quality.

2. Materials and methods

2.1. Samples and chemicals

n-hexane and analytical grade anhydrous sodium sulfate were purchased from Merck (Darmstadt, Germany). The Thyme and Melissa leaves used was collected from Shiraz, Iran in August 2016.

2.2. GC–MS analysis

The Analyses were performed using a GC–MS system (Hewlett-Packard 6890 gas chromatograph coupled to a HP-5973 mass spectrometer), equipped with split/splitless injector in the splitless mode and at 250 °C during the chromatographic run. The volatile compounds were separated in a capillary column (30 m × 0.25 mm, 0.25 µm film thickness, HP-5MS) using N_2 (99.99%) as a carrier gas at a 1 mL/min

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flow rate. The oven temperature was varied as follows: 60 °C (5 min), then warmed to 250 °C at 5 °C/min, then held 10 min at 250 °C (Khalili et al., 2017). The mass detector conditions were: transfer line temperature of 220 °C, ion source temperature of 220 °C and ionization mode with electron impact at 70 eV. The Kovats retention index (RI) values were calculated using a homologous series of n-alkanes (C_8–C_{25}) and values were compared with values reported in the literature and available Adams data (Adams, 2007). All mass spectra were also compared with the data system library (Wiley 275). Identification of volatile compounds was achieved by matching mass spectra and retention indices (RI) with those obtained from Wiley library as well as from literature data.

2.3. Solvent free microwave extraction (SFME)

The microwave-accelerated reaction system (NEOS, Milestone, Italy) was utilized for the experiment. This is a 2.45 GHZ multimode microwave reactor with a maximum power of 900 W delivered in 10 increments. This instrument is equipped with an infrared temperature sensor, an electromagnetic stirrer, a time controller, and a circulating water-cooling system. The plant materials were wetted before extraction by soaking in certain proportion of water for 1 h, and then removal the excess water. The wetted material was placed inside the reaction flask. Essential oil was condensed in the receiving flask with circulating water- cooling system. The extraction time, extraction temperature, irradiation power can be controlled by an electronic control panel. SFME extractions were conducted at irradiation power 700 W and extraction time 42 min. These optimized conditions were obtained by central composite design (CCD) and response surface methodology (RSM). Fig. 1. Revealed the interaction between extraction time and irradiation power on the extraction yield of essential oil. The anhydrous sodium sulfate was used for drying the isolated essential oil.

2.4. Hydrodistillation (HD)

Thyme and Melissa leaves were shade dried at room temperature for 72 h. The dried Thyme and Melissa leaves were loaded to hydrodistillation using a Clevenger-type apparatus, according to European Pharmacopeia, and extracted with 1 L of water for 240 min (until no more essential oil was reused). The anhydrous sodium sulfate was used for drying the isolated essential oils.

3. Results and discussion

3.1. Essential oil analysis

The identified constituents in the extracted *Thymus vulgaris* L. and *Melissa officinalis* L. essential oils by HD and SFME techniques are present in Table 1. At first, for *Thymus vulgaris*, by hydrodistillation and solvent-free microwave extraction 22 and 20 compounds, respectively were characterized, according to 97.22% and 98.92% of the essential oils. In the hydrodistillation technique, the essential oil consists of oxygenated compounds 62.46% with, Thymol (28.92%), Carvacrol (26.18%), *p*-Cymene (13.25%) and γ-Terpinene (5.09%) as the main components in the oil.

Comparatively, the main constituents of the oil using solvent-free microwave extraction technique was Thymol (32.17%), Carvacrol (29.25%), *p*-Cymene (9.51%) and γ-Terpinene (3.62%). Furthermore, the oil consists of 75.06% oxygenated compounds.

Fig. 2 shows the main compounds in Thyme essential oil extracted by hydrodistillation and solvent-free microwave extraction, it was noticed that higher amounts of Thymol and Carvacrol were present in the essential oil by SFME.

Then, for Melissa, by hydrodistillation and solvent-free microwave extraction 10 and 10 compounds, respectively were characterized, according to 90.41% and 99.95% of the essential oils. In the hydrodistillation technique, the essential oil consists of oxygenated compounds 63.09% with, Caryophyllene oxide (23.71%), E-Citral (19.53%), Z-Citral (16.39%), and α-Muurolene (12.17%) as the main components in the oil.

Comparatively, the main constituents of the oil in solvent-free microwave extraction technique were Caryophyllene oxide (24.95%), E-Citral (24.19%), Z-Citral (18.45%) and α-Muurolene (12.50%). Furthermore, the oil consists of 70.87% oxygenated compounds.

Fig. 3 shows the main compounds in Melissa essential oil extracted by hydrodistillation and solvent-free microwave extraction, it was noticed that superior amounts of Caryophyllene oxide, Z-Citral and E-Citral were present in the essential oil by SFME.

The results show that higher levels of oxygenated compounds were found in the herbs essential oils extracted using SFME, regarding the microwave extraction time. This may be due to the reduced heating time required, which partially prevented decomposition oxygenated compounds by thermal and hydrolytic reactions (Lucchesi et al., 2004; Bendahou et al., 2008). Additionally, antioxidant and antimicrobial

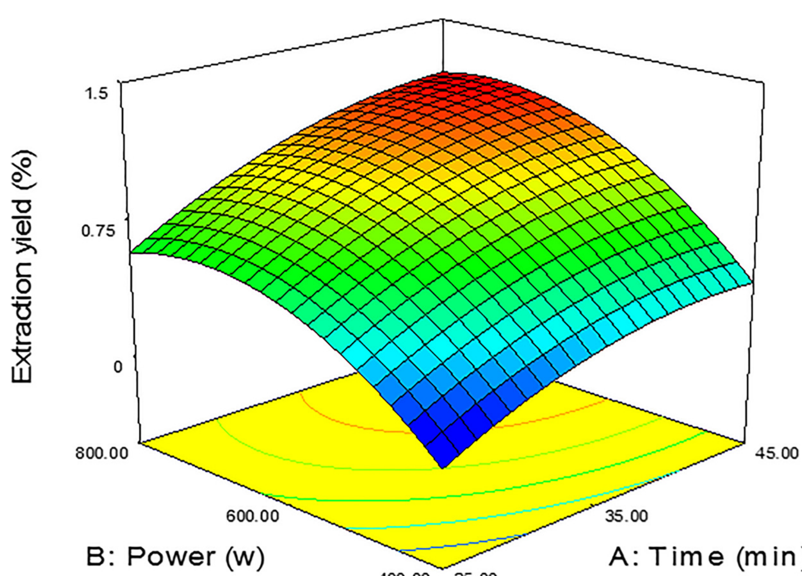

Fig. 1. Response surface representation for essential oil from *Thymus vulgaris* L.