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Research Paper

Spectroscopic Investigation of *Syzygium aromaticum* L. Oil by Water Distillation Extraction

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Abstract: Water distillation is commonly used for obtaining essential oils from aromatic herbs. In the present work Clevenger apparatus is applied to obtain essential oil from *Syzygium aromaticum* L. Clevenger extraction represents an environmentally method for oil herb extraction as it employs water as solvent during the distillation process. Essential oils are highly valued. They are traditionally used in medicines and health remedies. The purpose of the present work is to obtain essential oil from clove plant and subsequently identify the chemical composition by means of spectroscopic techniques. Essential oil samples were analysed by spectroscopic techniques FTIR spectroscopy and UV-Vis. spectrophotometer. Both techniques indicated presence of eugenol as main chemical component in the oil of clove in excellent agreement with reported literature data.

Keywords: *water distillation, clove flowers; essential oil; oil extract; FTIR; UV-Vis.*

Introduction

Hydro-distillation is commonly used for the extraction of essential oils from plants. It is reported that hydro-distillation can be categorized into steam distillation and water distillation (Dilworth *et al*., 2017). In addition, hydro-distillation can be a combination of water and steam distillation (Dilworth *et al*., 2017). Hydro-distillation is one of the most preferred methods as it represents an eco-friendly process (Taraj *et al.*, 2019a). In addition, its application has a low cost.

The distinction between steam distillation and water distillation is that in steam distillation, steam extracts essential oils at temperatures to approx. 100°C, whereas in water distillation the plant is plunged into water and subsequently boiled (Dilworth *et al*., 2017). Oil extracts are composed of aromatic constituents and they are traditionally used in medicines and health remedies.

In the current work, *Syzygium aromaticum* L. is used to attain essential oil by use of Clevenger apparatus and by employing water as extracting solvent. The aim is to isolate the oil and next to identify the chemical composition by means of spectroscopic techniques. In this work, eugenol is found to be the main chemical constituent in clove oil. Eugenol is a common odour in many dental practices. Additionally, for comparison reason Soxhlet extraction using hexane as solvent was carried out as well.

It is reported that eugenol quantity in clove oil is up to 90% (Yadava and Saini 1994, Li 2001, Guan *et al*., 2007, Rana *et al*., 2011, Mohammed *et al*., 2016). Additionally, spectroscopic techniques such as FTIR spectroscopy and UV-Vis. spectrophotometer were used for the characterization of essential oil samples. FTIR and UV-Vis. analyses are well reported in the literature (Schulz *et al*., 2005; Schulz & Baranska 2007; Andoni *et al*., 2015; Ciko *et al*., 2016a; Ciko *et al*., 2016b; Gakis 2016; Soto-Barajas *et al*., 2018; Taraj *et al*., 2013; Taraj *et al*., 2017; Taraj *et al*., 2018a; Taraj *et al*., 2018b; Taraj *et al*., 2019a; Taraj *et al*., 2019b). Infrared spectroscopy (FTIR) is well-known as a fast and accurate investigative technique (Soto-Barajas *et al*., 2018). It is reported that FTIR can be successfully used for the qualitative differences between samples (Andoni, 2009; Andoni *et al*., 2009; Andoni, 2014; Andoni *et al*., 2015; Andoni *et al*., 2018; Rodrıguez *et al*., 2018; Soto-Barajas *et al*., 2018; Taraj *et al.*, 2019a). Additionally, FTIR and UV-Vis. spectra of clove oil samples indicated

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presence of eugenol as main chemical component in good agreement with reported data (Yadava & Saini, 1994; Mohammed *et al*., 2016).

Materials and Methods

Sampling and extraction methodology

The origin of *Syzygium aromaticum* L. used in this work is from local Albanian herb. The hydro-distillation extraction was carried out, in a round bottom flask, using Clevenger equipment. The amount of *Syzygium aromaticum* L. used for hydro-distillation was 15 g. The water-herb mixture underwent distillation process for 4 hours (Taraj *et al.*, 2019a; European Pharmacopoeia,1983).

As noted in the introduction section, for comparison reason, Soxhlet extraction was carried out as well. The amount of *Syzygium aromaticum* L. used for Soxhlet distillation was 10 g. Additionally, the Soxhlet extraction was allowed to run for 4 hours by using hexane as solvent. The oils were dissolved in hexane and further used for FTIR and UV-Vis analyses.

FTIR spectra were acquired by Nicolet 6700 spectrometer, manufactured by Thermo Electron. FTIR spectra were recorded by using KBr plates. Additionally, the interval of measurements was in the range mid Infra-Red (4000-400 cm^{-1}). OMNIC software was used for additional analyses. UV-Vis spectra measurements were recorded by 6800 UV-VIS Jenway spectrophotometer.

Results and Discussion

Table 1 displays summarized results for Clevenger and Soxhlet extractions. These values are in good agreement with reported results by Ciko *et al*. (2016a), Taraj *et al*. (2019a) and Taraj *et al*., (2019b) for the relevant extraction methods.

Figure 1 represents UV-Vis. spectra of *Syzygium aromaticum* L. essential oil attained by Clevenger extraction. Additionally, in the insert the chemical structure of the main chemical compound i.e. eugenol is displayed (Yadava & Saini, 1994; Mohammed *et al*., 2016). The oil yield was 1.2% for 15 g herb used for the extraction.

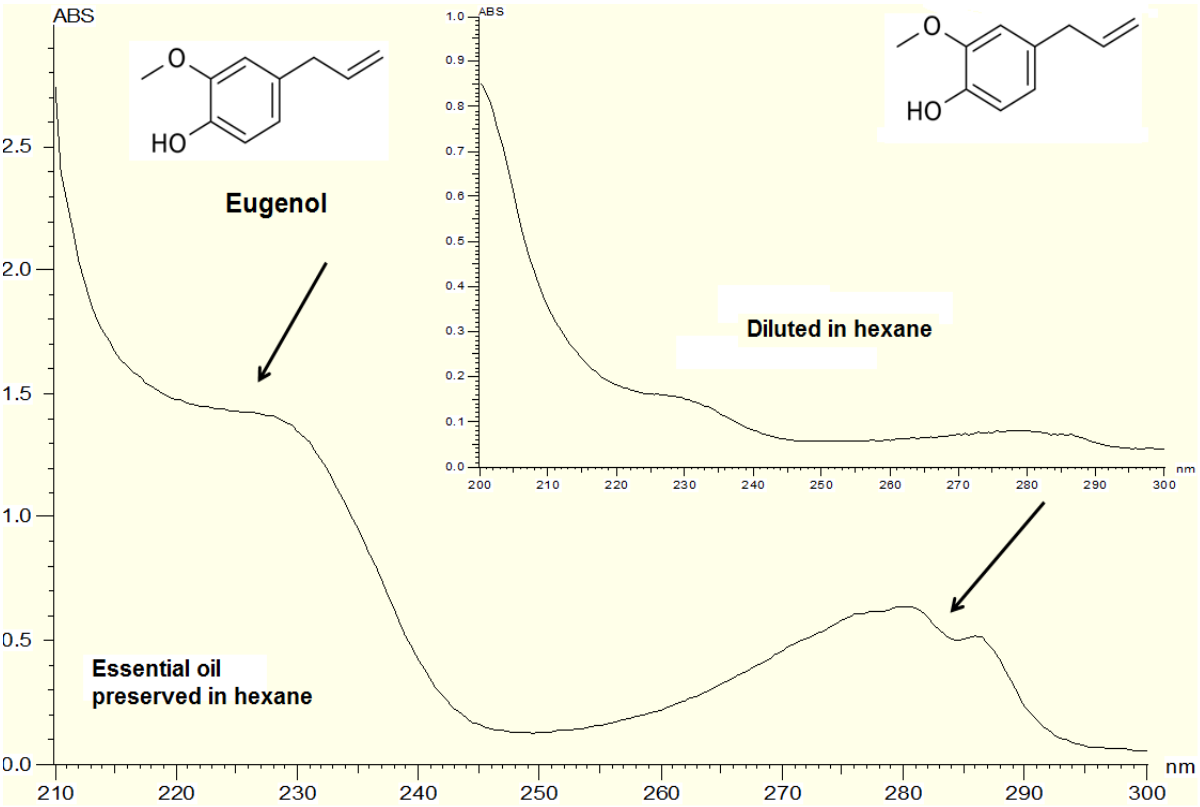

Figure 1. UV-Vis. spectra of *Syzygium aromaticum* L. essential oil obtained by Clevenger extraction.

Table 1: Tabulated results for the extractions of *Syzygium aromaticum* L.

Amount of *Syzygium aromaticum* L.	Extraction solvent	Extraction time	Extraction method	Yield of extract
15 g	Water	4 h	Clevenger	1.2%
10 g	Hexane	4 h	Soxhlet	12.5%

The UV-Vis. spectrum displays two absorption bands at 225-230 nm and at 270-290 nm. The absorption at 225-230 nm arises from the electronic transition of *n-π type of alcohol functional group -OH that links to the eugenol aromatic ring (Mohammed *et al*., 2016). Absorption band at 270-290 nm regards the electronic shift of *n-π type for C-O group that binds with eugenol aromatic ring (Mohammed *et al*., 2016).

Figure 2a displays FTIR spectrum for the extract of *Syzygium aromaticum* L. herb by Soxhlet extraction. Characteristic FTIR signals appear at 1637 cm^{-1}, 1606 cm^{-1} and 1513 cm^{-1}. It is reported (Yadava and Saini 1994, Smith 1999, Mohammed *et al*., 2016) that C-C vibrations of aromatic rings appear at 1630 cm^{-1}, 1604 cm^{-1} and 1508 cm^{-1}. Therefore, the wavenumbers at 1637 cm^{-1}, 1606 cm^{-1} and 1513 cm^{-1} belong to C-C aromatic rings vibrations of eugenol and eugenol acetate. In addition, C=C double bonds give rise to FTIR bands at 1640-1630 cm^{-1} (Smith, 1999).

The band at 1765 cm^{-1} is attributed to the ester group C-O or aromatic ketone group C=O which can be combined with more than one ring (Yadava & Saini 1994, Smith 1999, Mohammed *et al*., 2016). The peak at 1765 cm^{-1} is assigned to eugenol acetate presence. In addition, the intense FTIR bands in the interval 1300-1000 cm^{-1} belong to the C-O vibrations of ether and alcohol functional group of eugenol and eugenol acetate (Yadava & Saini 1994, Smith 1999, Mohammed *et al*., 2016). Other moderate bands appear at 794 cm^{-1} and 995 cm^{-1}. They belong to the groups CH_2 and C-H bonding vibrations, alkene monosubstituted, respectively (Yadava & Saini, 1994; Smith, 1999; Mohammed *et al*., 2016). Lastly, Figure 2b displays a Photo of *Syzygium aromaticum* L. oil extract. The oil appeared yellow in colour. Due to eugenol presence, the extract had a pleasant and spicy, clove like fragrance. The yield for the Soxhlet extraction was 12.5% for 10 g herb used.

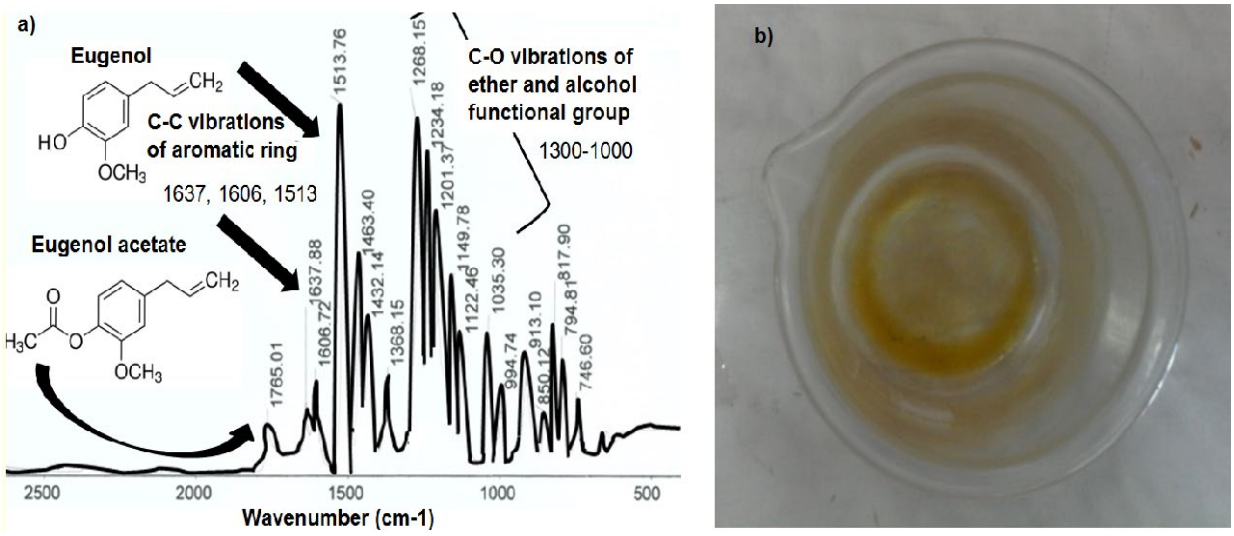

Figure 2: a) FTIR spectrum of *Syzygium aromaticum* L. extract obtained by Soxhlet apparatus. The main chemical compounds of *Syzygium aromaticum* L. oil extract i.e. eugenol and eugenol acetate are indicated in the inserts. b) Photo of *Syzygium aromaticum* L. oil extract.

Table 2 summarizes the most important FTIR bands (wavenumbers) identified in the essential oil and oil extract of *Syzygium aromaticum* L. compared with relevant data in the literature. It is evident that the obtained data are in good agreement with previous publications reporting IR spectra analyses of essential oil and oil extract from *Syzygium aromaticum* L. (Yadava & Saini 1994; Mohammed *et al*., 2016).

Table 2. FTIR and UV-Vis. assignments for the most characteristics bands of the oil extract compound for *Syzygium aromaticum* L.

Compound	FTIR (cm^{-1})	Assignments	UV-Vis. (nm)	Assignments
Eugenol	~1637, 1606, 1513 (this work), ~1650 (Yadava and Saini)	C-C of aromatic ring	~225-230 (this work), ~224 (Mohammed et al)	-OH
Eugenol acetate	~1637, 1606, 1513 (this work) 1650 (Yadava and Saini)	C-C of aromatic ring	~270-290 (this work), ~268 (Mohammed et al)	C-O
Eugenol	~1300-1000 (this work) 1070 (Mohammed *et al*)	C-O alcohol and ether		
Eugenol acetate	~1300-1000 this work)	C-O ether		
Eugenol acetate	~1765 (this work), ~1730 (Mohammed *et al*)	C=O ketone		

Conclusions

In this paper, distillation process was employed to attain essential oils from *Syzygium aromaticum* L. herb. Hydro-distillation was carried out by employing Clevenger apparatus. In addition to hydro-distillation extraction, Soxhlet extraction was utilized as well. FTIR spectroscopy and UV-Vis spectrophotometer were used as rapid techniques to analyze the extracts of *Syzygium aromaticum* L. Spectral analyses indicated presence of two chemical constituents i.e. eugenol and eugenol acetate in the oil extracts of *Syzygium aromaticum* L. This result was in good agreement with other relevant studies. The distinctive bands in the UV-Vis. spectrum were ~225-230 nm and ~270-290 nm, whereas the characteristic bands in the FTIR spectrum were in the intervals ~1637-1513 cm^{-1}, ~1300-1000 cm^{-1} and 1765 cm^{-1}.

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