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Analytical Methods

Comparison of two isolation methods for essential oil from rosemary leaves: Hydrodistillation and microwave hydrodiffusion and gravity

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ABSTRACT

Traditional hydrodistillation (HD) and innovative Microwave Hydrodiffusion and Gravity (MHG) methods have been compared and evaluated for their effectiveness in the isolation of essential oil from fresh *Rosmarinus officinalis* leaves. The microwave method offers important advantages over traditional alternatives, namely: shorter isolation times (15 min against 3 h for hydrodistillation), environmental impact (energy cost is fairly higher to perform HD than that required for rapid MHG isolation), cleaner features (as no residue generation and no water or solvent used), increases antimicrobial activities, increases antioxidant activity and provides a more valuable essential oil (with high amount of oxygenated compounds). It offers also the possibility for a better reproduction of natural aroma of the essential oil from rosemary leaves than the HD essential oil. Moreover, microwave procedure yielded essential oils that could be analysed or used directly without any clean-up, solvent exchange or centrifugation steps. Scanning electron microscopy shows important structural changes for MHG extraction in contrast to those obtained by HD. Electron micrographs show clearly that the cells are broken and damaged during microwave treatment. Finally, the mechanism of Microwave Hydrodiffusion and Gravity is proposed and discussed.

1. Introduction

Rosemary (*Rosmarinus officinalis* L.) is a perennial herb with fragrant evergreen needle-like leaves. It is native to the Mediterranean region and it has been cultivated for a long time. It belongs to the Lamaiaceae family, which comprises up to 200 genera and about 3500 species. The leaves are evergreen, with dense short woolly hairs. Rosemary has been a significant herb since antiquity, although rosemary is more familiar to contemporary Westerners as a kitchen herb used to add a spicy or slightly medicinal flavour to some foods, it was traditionally used as an antiseptic, astringent, and food preservative before the invention of refrigeration. Rosemary's antioxidant properties are still used to extend the shelf life of prepared foods (Cuvelier, Richard, & Berest, 1996). Rosemary is also known medicinally for its powerful antioxidant activity (Ibanez et al., 2003), its antibacterial and antimutagenic properties, and as a chemopreventive agent (Oluwatuyi, Kaatz, & Gibbons, 2004). Besides the therapeutical application, the essential oil is widely applied in the cosmetic industry producing various Cologne waters, bathing essences, hair lotions and shampoos and as a component of disinfectants and insecticides (Boelens, 1985).

The essential oil secreted by glandular trichomes is mainly located in leaves. Essential oil can be isolated using a number of isolation methods, e.g. hydrodistillation, steam distillation and organic solvent extraction. Nevertheless, monoterpenes are well known to be vulnerable to chemical changes under steam distillation conditions, and even conventional solvent extraction is likely to involve losses of more volatile compounds during removal of the solvent (Presti et al., 2005). Many of these methods are moreover time-consuming and energy intensive. There are many publications dealing with the extraction of *R. officinalis* using alternative techniques. Recently, the supercritical fluid extraction of rosemary with CO_2 has been the object of a lot of research (Carvalho, Moura, Rosa, & Meireles, 2005) and has become a valid alternative to the more conventional extraction procedures, mainly because the dissolving power of the extracting medium can be adjusted by regulating the pressure and temperature conditions. However, the technological conditions required for the use of supercritical fluid extraction are onerous and the high cost of producing specific products has limited its use. Moreover, in certain cases, the extractive power of supercritical CO_2, towards specific analytes, is insufficient under conventional conditions (Lucien & Foster, 2000) and

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inversely excessive with the extraction of undesirable compounds as vegetable waxes or resins (Guinamant, 1992). Microwave-assisted solvent extraction (MASE) (Chen & Spiro, 1995) appeared to be particularly attractive for isolation of essential oil from rosemary. The popularity of microwave technique is due to the rapid rates of heat transfer which allows quicker times of extraction. Ultrasounds were also used to increase the solvent extraction efficiency of antioxidants from rosemary (Albu, Joyce, Paniwnyk, Lorimer, & Mason, 2004). The application of ultrasound irradiation facilitated the low-temperature rupturing of plant cell membranes, thereby liberating molecules from cellular structures. However, the principal drawback of these methods is that the solvent used cannot be completely separated from the extract at the end of the process. Tigrine-Kordjani, Meklati, and Chemat (2006) have been recently developed a microwave-assisted distillation (MAD) with free solvent for laboratory scale applications in the extraction of essential oils from different kinds of aromatic plant. Recently, Bendahou et al. (2008) have been reported the extraction of *Origanum glandulosum* essential oil with MAD. Nowadays, more and more advanced techniques utilising microwave radiations in the extraction process have been documented in the literature (Golmakani & Rezaei, 2008; Wang et al., 2008). Rezzoug, Boutekedjiret, and Allaf (2005) have been isolated essential oil from rosemary leaves by an innovative process called "Detente Instantannée controlée" or controlled instantaneous decompression. This process involves subjecting plant material for a short time to a steam pressure followed by an instantaneous decompression to a vacuum.

Recently, a new greener extraction technique, Microwave Hydrodiffusion and Gravity was designed and developed. The essential oil isolation based on this technique, which was successfully tested for isolation of essential oil from mint plants (Abert Vian, Fernandez, Visinoni, & Chemat, 2008) is an interesting alternative not only to standard techniques of essential oil isolation, such as isolation with solvent or steam distillation, but also to more effective processes described above. This process is based of an original combination of microwave heating and earth gravity at atmospheric pressure.

In this paper, the essential oil from rosemary (*R. officinalis*) obtained by Microwave Hydrodiffusion and Gravity has been compared with those obtained by conventional hydrodistillation. We make appropriate comparisons in terms of kinetics of isolation, quality and quantity of essential oil, antimicrobial activities, antioxidant activity and energy consumption. We have also proposed a mechanism for MHG technique. This study was supplemented by scanning electron micrographs to shed light on the isolation mechanism.

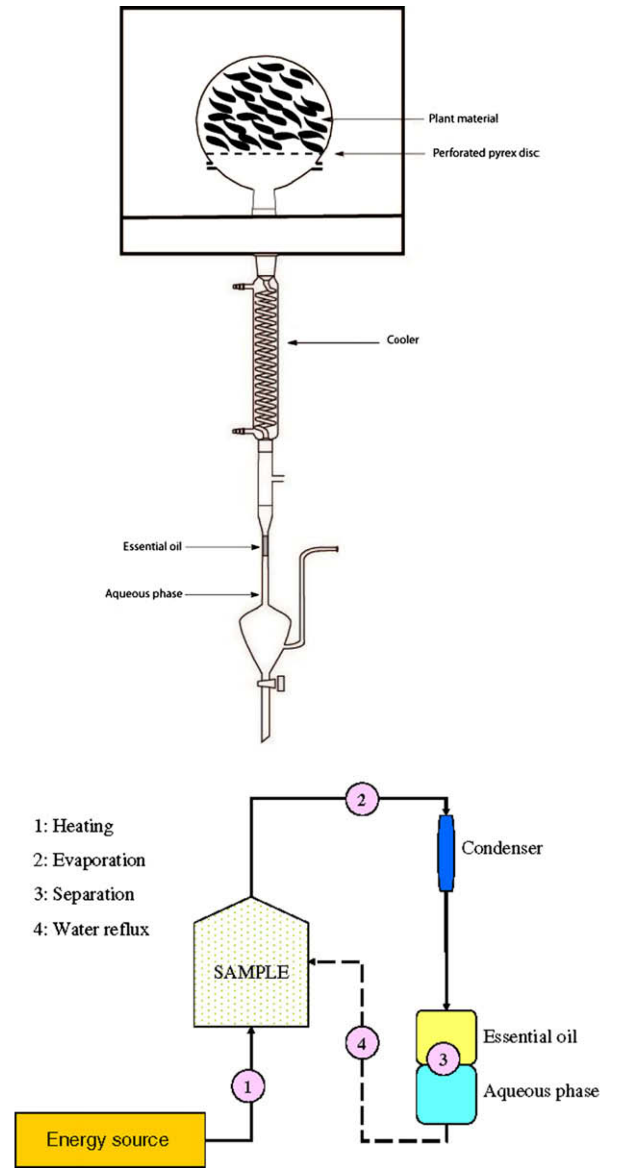

Fig. 1. Isolation methods of essential oil from *Rosmarinus officinalis* (MHG and HD).

2. Experimental

2.1. Plants material

Leaves of the cultivated plants of rosemary (*R. officinalis* L.) were collected in 2007 from the Institut National Agronomique (INA-El Harrach – Alger). The initial moisture of leaves was 60.2%.

2.2. MHG apparatus and procedure

Microwave Hydrodiffusion and Gravity has been performed using the "DryDist" microwave oven illustrated in Fig. 1. This is a multimode microwave reactor 2.45 GHz with a maximum delivered power of 1000 W variable in 10 W increments. Temperature was monitored by an external infrared sensor. In a typical MHG procedure performed at atmospheric pressure, 500 g of fresh plant material were heated using a fixed power density of 1 W g^{-1} for 15 min without addition of solvent or water.

The direct interaction of microwaves with biological water (i.e., steam produced from the water present in the fresh plant material) favours the release of essential oils trapped inside the cells of plant tissues. A mixture of hot "crude juice" and steam (*in situ* water) move thus naturally by earth gravity downwards on a spiral condenser outside the microwave cavity where it condensed. The oily condensate is collected continuously in a receiving flask (similar to separator funnel) where essential oil forms a film on the surface of the water and the film is skimmed off the top. At the end the essential oil is collected, dried with anhydrous sodium sulphate and stored at 4 °C until used.

2.3. Hydrodistillation apparatus and procedure

Five hundred grams of each aromatic herb were submitted to hydrodistillation with a clevenger-type apparatus (Conseil de l'Europe and Pharmacopée Européenne 1, 1996) according to the European Pharmacopoeia and extracted with 3 l of water for 90 min (until no more essential oil was obtained). The essential oil was collected, dried under anhydrous sulphate and stored at 4 °C until used.