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Ultrasonic extraction of waste solid residues from the *Salvia* sp. essential oil hydrodistillation

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ABSTRACT

Enormous quantities of aromatic plants such as *Salvia* sp. are used in the production of essential oils all over the world, thereby creating a huge waste solid residue. Since the spent plant material (SPM) may contain biological compounds, it could be exploited as secondary raw material for obtaining different bioactivities using solvent extraction, with possible technological, economical and ecological justification. A novel extraction technique, namely the ultrasonic extraction, and two extracting solvents of distinct polarity (petroleum ether and 70% (v/v) aqueous ethanol solution) were used to obtain extractive substances (ES) from the SPM of two *Salvia* sp. The investigation was focused on the ES yield, the extraction kinetics and the composition of the final extracts obtained. The maximum concentration of ES in the liquid extracts was reached after about 20–30 min of sonication. The kinetics of ultrasonic extraction was described by the unsteady-state diffusion through the plant material and the film theory. The chemical composition of the extracts depended on the extraction conditions and the type of the plant material. Many biologically active compounds were detected in the extracts. So far the "waste" SPM from the *Salvia* sp. essential oil hydrodistillation may be considered as a possible source of the exploitable natural products. The benefit of the ultrasound action was related to shortening of the extraction time (60 min) compared to the classical extraction (6 h).

1. Introduction

Hydrodistillation is normally used to isolate essential oils from aromatic and medicinal plants. Besides the essential oil, which is the main product, the aromatic water ("hydrosol") and the spent plant material (SPM) are also obtained by hydrodistillation. Hydrosols are rich in water-soluble hydrophilic components of the original plant, and are considered as ready-to-use products such as in aromatherapy and cosmetic industry. Remaining SPM is usually treated as a waste material with no commercial value with seldom use as cheap solid fuel energy, cattle feed or a fertilizer.

There are a few reports on the use of SPM remaining after the essential oil hydrodistillation as a source of compounds which might possess biological activities. The water-soluble extracts recovered from the steam distilled, essential oil extracted aerial parts of several aromatic plants were shown to possess antioxidant properties [1]. The SPM remaining after the essential oil water distillation from the aerial parts of two *Salvia* species was shown to contain considerable quantities of flavonoids [2]. It has already been demonstrated that the ethanolic extract obtained from the *S. officinalis* SPM exhibited the antimicrobial activity against the bacteria, yeasts and fungi [3]. Another example is the extraction of glycosides with the boiling water from the SPM remaining after the hydrodistillation of the essential oil from *Mentha aquatica* L. [4]. There is a lack of information in the literature about the content and the constituents of the extracts from SPM remaining after the hydrodistillation of aromatic plants.

As in the case of any plant material, different solvent extraction techniques can be used for obtaining extractive substances (ES) from SPM. In the last decade, ultrasonic extraction has been more extensively used for the extraction of ES from different parts of many plants. For instance, it has already been shown that ultrasound promotes the extraction of bioactive substances [5–7] and polysaccharides [8] from *S. officinalis*. As a novel technique, the ultrasonic extraction has recently been reported to be very promising and effective for obtaining bioactive substances from sage, ensuring higher yields of the ES in much shorter times than the classical extraction [9]. The extracts obtained by ultrasonic extraction

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Nomenclature

b	washing coefficient according to the film model, 1
b'	washing coefficient according to the unsteady diffusion model, 1
c	concentration ES in a liquid extract during the extraction (g/L)
c_f	final concentration of ES in a liquid extract (g/L)
c_s	concentration of ES in a saturated liquid extract (g/L)
k	slow extraction coefficient according to the film model (min^{-1})
k'	slow extraction coefficient according to the unsteady diffusion model (min^{-1})
q	content of ES in the plant material during the extraction (g/100 g)
q_f	content of ES in the plant material at the end of the extraction (g/100 g)
q_0	content of ES present in the plant material (g/100 g)
t	time (min)

from the SPM remaining after the essential oil water distillation from the *Salvia* species contained more flavonoides than those obtained by classical extraction [2]. The ultrasonic enhancement of the extraction is attributed to the cell destruction, capillary effects, better solvent penetration and mass transfer intensification [10].

In this work the ultrasonic extraction was employed to obtain the ES from the waste solid residues from the hydrodistillation of essential oil from the ground, dry aerial parts of garden (*S. officinalis*) and glutinous (*S. glutinosa*) sage using petroleum ether and 70% (v/v) aqueous ethanol solution as extracting solvents of different polarity. The main goal was to estimate if these waste residues could be exploited as secondary raw materials for obtaining bioactive compounds. The attention was focused on the yield and the composition of the final extracts obtained, as well as on the extraction kinetics.

2. Experimental

2.1. Materials

The aerial parts of the garden (*S. officinalis*) and glutinous (*S. glutinosa*) sage, originating from east Serbia, were used. The additional information on harvesting, drying, packaging and storing of the herb materials can be found elsewhere [9]. Before being used, the plant material was comminuted by a hammer mill and sieved through a 6 mm screen.

Petroleum ether (boiling temperature: 50–70 °C) and ethanol were purchased from Zorka Pharma (Šabac, Serbia) and Merck, respectively.

2.2. Hydrodistillation

The ground herb material (400 g plus 5 L of distilled water) was submitted to hydrodistillation in a Clevenger-type apparatus equipped with a 10 L distillation flask for 2 h. The essential oil was separated from the aromatic water, dried over anhydrous Na_2SO_4, diluted by ethanol (1 mL of the oil and 99 mL of ethanol) and immediately analyzed. The hydrosol was separated by vacuum filtration and the remaining SPM was dried in a well-aired place in a thin layer for 5 days. The moisture content, determined by drying at 105 °C to constant mass, was about 12% for the SPM of both plant species.

2.3. Initial content of ES in SPM

The SPM (10 g) and petroleum ether or the 70% (v/v) aqueous ethanol solution as the extracting solvent (200 mL) were placed in an Erlenmayer flask, and heated under reflux for 4 h. The liquid extract was separated from the solid residue by vacuum filtration. The plant residue was returned to the Erlenmayer flask and a fresh solvent (200 mL) was added. The suspension was heated under reflux for 30 min. The liquid extract was separated and the plant residue was washed with the fresh solvent (50 mL). Three filtrates were collected, and the solvent was evaporated at 50 °C. The extract was then dried under vacuum at 50 °C to constant weight. The values of the initial content of ES present in the SPM of two *Salvia* sp. are given in Table 1.

2.4. Concentration of ES in saturated liquid extracts

The SPM (10 g) and the extracting solvent (100 mL) were added to an Erlenmayer flask placed in an ultrasonic cleaning bath (Sonic, Niš, Serbia; total nominal power: 3× 50 W; and internal dimensions: 30 cm × 15 cm × 20 cm) operating at 40 kHz and sonicated at 40 ± 0.1 °C for 20 min. Then, the liquid extract was separated by vacuum filtration and used for extracting the ES from a further portion (10 g) of the fresh SPM. The extraction was repeated with the liquid extract and a further quantity of fresh SPM (10 g). The mass of the dry extract was not increased after the fourth extraction cycle, meaning that the saturated solution of ES in the extracting solvent had been prepared. After the third extraction cycle, the solvent was evaporated under vacuum at 50 °C. The extract was then dried under vacuum at 50 °C to constant weight. The concentration of ES in a saturated liquid extract was calculated from the mass of the dry extract and the known volume of the liquid extract. The values of the concentration of ES in the saturated liquid extracts for the two extracting solvents are given in Table 1.

2.5. Kinetics of ultrasonic extraction

The SPM (10 g) and the extracting solvent (100 mL) were placed in a series of Erlenmayer flasks (250 mL) and the flasks were soni-

Table 1
The initial content of ES of the plant materials, the concentration of ES of the saturated liquid extracts, the final concentration of ES in liquid extracts and the extraction efficiency for different SPM and extracting solvents (mean values of two experiments)

Origin of SPM	Extracting solvent	q_0[a] (g/100 g of SPM)	c_s (g/L)	c_f[b] (g/L)	Extraction efficiency[c] (%)
S. officinalis	Petroleum ether	4.7	8.5	1.9	40.4
	Aqueous ethanol solution, 70% (v/v)	14.6	12.9	6.0	41.1
S. glutinosa	Petroleum ether	2.2	6.5	1.3	59.1
	Aqueous ethanol solution, 70% (v/v)	12.6	10.5	4.6	36.5

[a] Symbols are shown in Nomenclature.
[b] After the 80-min extraction.
[c] Defined as $1 - q_f/q_0 = c_f/q_0$.