Industrial Crops & Products 124 (2018) 633–642

Contents lists available at ScienceDirect

Industrial Crops & Products

journal homepage: www.elsevier.com/locate/indcrop

Microwave hydrodiffusion and gravity for rapid extraction of essential oil from Tunisian cumin (*Cuminum cyminum* L.) seeds: Optimization by response surface methodology

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ARTICLE INFO

Keywords:
Essential oil
Cuminum cyminum L. seeds
Hydrodistillation
Microwave hydrodiffusion and gravity
Response surface methodology

ABSTRACT

Recently, microwave hydrodiffusion and gravity (MHG) pushed the confines of the microwave assisted extraction (MAE) further toward an innovative, quick, efficient, and eco-friendly process for plant secondary metabolites extraction. The goal of this study was to optimize the microwave hydrodiffusion and gravity (MHG) process for the extraction of the essential oils (EO) from Tunisian cumin (*Cuminum cyminum* L.) seeds. The effects of different parameters: microwave irradiation time (8, 12, and 16 min), microwave irradiation power (150, 200, and 250 W), and moisture content (40, 50, and 60%), on the extraction yield were achieved using the response surface methodology (RSM) with central composite design (CCD). In order to elucidate the efficiency of the MHG, it was compared with conventional hydrodistillation (HD) in terms of kinetic, quantity and quality of obtained EO, environmental impact, and energy consumption. Results showed that, under the optimum conditions defined by the central composite face-centered (CCF) and calculated by the analysis of variance (ANOVA) (16 min of microwave irradiation time, 203.30 W of microwave irradiation power, and 44.67% of moisture content), the yield of cumin essential oil obtained with MHG was 1.579 ± 0.05%. Meanwhile, compared with the HD process, the MHG successfully improved the EO yield (1.579 ± 0.05% *versus* 1.550 ± 0.07% with HD) in shorter extraction time (16 min *versus* 150 min with HD), less electrical consumption, lower carbon dioxide emissions (CO_2), and smaller volume of waste water. Using the GC-FID and GC–MS analysis, the MHG provided more valuable EO with high amount of oxygenated compounds compared with the HD. Scanning electron micrograph (SEM) confirmed the efficiency of MHG for EO extraction. Thus, MHG appeared like rapid process, green technology, and desirable alternative protocol to enhance the quantity and the quality of the essential oil extracted from the medicinal and aromatic plants (MAPs).

1. Introduction

Over the past few years and with the development of the *Green Chemistry*, the application of the microwave assisted extraction (MAE) gained enormous popularity as a favored environment-friendly process for extraction of secondary metabolism in numerous laboratories and major industries (Filly et al., 2014; Yu et al., 2014). In fact, the MAE improved the quantity and the quality of the extract, reduced the time of extraction, decreased the cost and the energy consumption, and minimized the quantity of carbon dioxide (CO_2) emitted into the atmosphere compared with the conventional extraction methods as hydrodistillation (HD) (Farhat et al., 2011).

Recently, microwave hydrodiffusion and gravity (MHG) pushed the confines of the microwave assisted extraction further toward an innovative, quick, efficient, and eco-friendly process without any degradation in the quality of the extract (López-Hortas et al., 2016; Pérez et al., 2014). In reality, the key property of the MHG extraction was based on the combination of the microwave irradiation and the earth

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https://doi.org/10.1016/j.indcrop.2018.08.036
Received 30 May 2018; Received in revised form 12 August 2018; Accepted 13 August 2018

gravity at atmospheric pressure, without adding the solvent or the water (Binello et al., 2014; López-Hortas et al., 2016).

Moreover, the MHG extraction showed the best results addressed in a wide variety of applications focused on the extraction of the active compounds such as: antioxidant molecules, essential oils, colorants, pectin, and polyphenols from the medicinal and aromatic plants (Al Bittar et al., 2013; Boukroufa et al., 2015; Bousbia et al., 2009a; Périno et al., 2016; Zill-e-Huma et al., 2009). The success of the MHG in the laboratory scale pushed the researchers to expand its application area. Périno et al. (2016) used a pilot scale of MHG system for the extraction of the polyphenols from lettuce.

Cumin is one of the medicinal and aromatic species belong to the Apiaceae family (Umbelliferae). It originated in Turkistan, Egypt, and countries neighboring the Mediterranean area (Rebey et al., 2012; Thippeswamy and Naidu, 2005). Cumin is the most popular aromatic spice in the world after pepper (*Piper nigrum* L.) (Hajlaoui et al., 2010). In fact, cumin and its essential oil were commonly used as an aromatic spice in the food industry due to its strong aromatic quality (Wang et al., 2006). Furthermore, its essential oil was also used in the cosmetics and the perfumery industries (lotions, creams, and perfumes) (Dubey et al., 2017). In particular, the pharmaceutical industry used cumin volatile oil as an excellent antioxidant, antimicrobial, antitumor, cytotoxic, anti-inflammatory, antifungal, and antidiabetic agent (Bettaieb et al., 2011; Rebey et al., 2012; Einafshar et al., 2012; Hajlaoui et al., 2010; Li and Jiang, 2004; Petretto et al., 2018; Sharma et al., 2016).

Since the yield and the chemical composition of the essential oil may differ significantly with: agricultural factors, seasonal variations, ecological situations, and extraction methods (Moghaddam et al., 2015; Telci et al., 2006), the cumin essential oil was extracted in this study by the microwave extraction to improve the quantity and the quality of the volatile oil.

The complete MHG method was herein presented, where the effects of some parameters such as: the microwave irradiation time, the microwave irradiation power, and the moisture content on the MHG extraction were studied using the response surface methodology (RSM) as the powerful statistical method. The yield of Tunisian cumin essential oil was followed as a key parameter to achieve the optimum for MHG extraction. Then, the optimal MHG response was compared with the hydrodistillation (HD) in terms of the extraction kinetic, the quantity and quality of the essential oils, the environmental impact, and the energy consumption. Finally, the morphological of cumin seeds before and after extractions was also done by the scanning electron microscope (SEM) to better understand the effectiveness and the quickness of the MHG on the extraction of the essential oil from the medicinal and aromatic plants (MAPs).

2. Materials and methods

2.1. Plant material

Cumin seeds used for all extraction experiments were bought from a local market in Gabes, Tunisia (33°53′12″N/10°05′36″E), in May 2014. The plant samples were identified by the head of the herbarium of the regional station of Gabes, National Institute of Research in Rural Engineering, Waters, and Forests (INRGREF, Gabes, Tunisia). The particle size chosen was less than 2 mm. The initial moisture content of the cumin seeds was verified in the laboratory as 8.53 ± 0.04%.

2.2. Hydrodistillation

One hundred grams of cumin seeds were subjected to hydrodistillation using a Clevenger apparatus (Dubey et al., 2017). The hydrodistillation was performed to cumin seeds in distilled water with the ratio of 1:10. The digital Wattmeter LCD was placed in the electrical heater entrance monitored the consumptions of the power during the extraction of the essential oil, which was read directly from the digital displayer in kWh. In fact, to extract all quantities of the essential oil contained in the cumin seeds, 150 min was enough. The HD extraction was repeated in triplicate, and the mean values were reported.

2.3. Microwave hydrodiffusion and gravity

The microwave hydrodiffusion and gravity (MHG) was carried out in the domestic microwave oven (ME 711 K, 20 L, 2450 MHz) modified in the mechanical engineering hall in the National Engineering School of Gabes, ENIG, Gabes, Tunisia. The dimensions of the interior cavity of the microwave oven were 33 cm × 30.9 cm × 21.1 cm. Its maximum power was 800 W. The digital Wattmeter LCD placed at the microwave generator entrance monitored continuously the consumptions of the power during the extraction of the cumin essential oil, which was read directly from the numeric display in kWh. The extraction vessel was made of Pyrex glass, it was composed of the round bottom flask having a capacity of 500 ml, the receiving flask, the condenser, the glass thimble which was used as the connector coupling the neck of the round bottom flask and the top of the condenser through the bottom hole of the microwave cavity. In fact, the perforated glass disc contained in this connector, let the steam pass through it at the same time supported the plant materials. Finally, the container, which connected to the condenser and opened from the bottom.

In the MHG procedure performed at atmospheric pressure, one hundred grams of the cumin seeds were heated using a fixed microwave power and time without adding any solvent or water after being soaked in the distilled water to reach the moisture content in the experimental design. The earth's gravity and the direct interaction of the microwave irradiations with the *In Situ* water facilitate the diffusion of the essential oil to the outside of the cells of the plant tissues. The condenser placed outside the microwave oven condensed the distillate. All essential oils were collected in amber vials, dried over anhydrous sodium sulfate and stored at 4 °C prior to analysis.

2.4. Experimental design of response surface methodology and statistical analysis

The response surface methodology (RSM) was used to define the optimal conditions, providing the highest extraction yield of the essential oil from the cumin seeds assisted by the microwave hydrodiffusion and gravity.

The microwave irradiation time (X_1: 8–16 min), the microwave irradiation power (X_2: 150–250 W), and the moisture content (X_3: 40–60%) were selected as the three main independent variables. The response was the yield of the cumin essential oil (%).

The central composite face-centered (CCF) (Boukroufa et al., 2015), was employed in this study. Based on the preliminary experiments, the range of the three selected variables that affected the MHG extraction was chosen and investigated at three levels (− 1, 0, + 1) with the star points which were at the center of each face of the factorial space ($\pm \alpha = \pm 1$). The range and the level of the variables were summarized in Table 1.

In this CCF, twenty different experiments, including the six replicates at center point were employed to fit the full quadratic equation model that had the following form:

Table 1
Factors and their levels.

Factor	Levels		
	−1	0	+1
X_1: Microwave irradiation time (min)	8	12	16
X_2: Microwave irradiation power (W)	150	200	250
X_3: Moisture content (%)	40	50	60