



Analytical Methods

Extraction of *Cuminum cyminum* essential oil by combination technology of organic solvent with low boiling point and steam distillationX.-M. Li^{a,*}, S.-L. Tian^a, Z.-C. Pang^a, J.-Y. Shi^b, Z.-S. Feng^{c,*}, Y.-M. Zhang^{a,*}^aThe Institute of Aricultural Product Processing and Storage of GanSu Academy of Agricultural Sciences, Lanzhou 730070, PR China^bThe Institute of Modified Variety of Livestock Grass of GanSu Academy of Agricultural Science, Lanzhou 730070, PR China^cSchool of Food science, Xinjiang Agricultural University, urumqi 830052, PR China

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ABSTRACT

Extraction of essential oil from *Cuminum cyminum* seeds using a combination of organic solvent with low boiling point and steam distillation was explored. The effect of different parameters, such as particle size (40, 60, 80 mesh), temperature (°C) 10, 15, 20 and extraction time (3, 5, 8 h), on the extraction yield was investigated using three-level orthogonal array design. The experimental results showed that the temperature had the largest effect on the yield of the extract (oleoresin), followed by extraction time and particle size. The optimum parameters, such as temperature, particle size, and extraction time were in turn 20 °C, 80 mesh, and 8 h. Essential oil of *C. cyminum* seeds obtained by supercritical fluid extraction (SFE), hydrodistillation (HD), combination technology of organic solvent with low boiling point and steam distillation (OS-SD) were further analysed by gas chromatographic/mass spectrometric detection to compare the extraction methods. Forty-five compounds in the *C. cyminum* essential oil were identified, showing that the composition of the extraction by different methods was mostly similar, whereas relative concentration of the identified compounds was apparently different. General characteristics of the *C. cyminum* essential oil obtained by different methods were further compared, and OS-SD was considered as the optimum process among the three processes to obtain *C. cyminum* essential oil for high quality, simple technology and low cost.

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1. Introduction

Herbs and spices have been used for generations by humans as food condiments and to treat ailments duo to being less toxic and generally free from side effects (Valiathan, 1998). Scientific evidence is accumulating that the spices do have medicinal properties that alleviate symptoms or prevent disease. *Cuminum cyminum* is one of the popular spices regularly used as a flavouring agent. In Chinese traditional medicine, the seeds of the plant have been used for treatment of toothache, dyspepsia, diarrhoea, epilepsy and jaundice (Eikani, Goodarznia, & Mirza, 1999; Nostro et al., 2005). It also has diuretic, carminative, emmanogogic and antispasmodic properties (Janahmadi, Niazi, Danyali, & Kamalinejad, 2006; Singh, Kapoor, Pandey, Singh, & Singh, 2002). China is known as the home of *C. cyminum* and is an important exporter of this commodity. Besides its use in traditional medicine in the treatment of some ailments, *C. cyminum* is widely used in food. The spice contains essential oil that imparts a characteristic aroma to it. The proximate composition and the physicochemical properties of the essential

oil have been reported (Li & Jiang, 2004). The spice is well known appetizers and is considered essential in culinary art all over the world. In traditional usage, *C. cyminum* seeds are generally ground to a powder, but this may bring about disadvantages such as bad taste and rapid loss of flavour.

Because the essential oil of *C. cyminum* seeds is volatile, and some volatile components may vary significantly according to such factors as the storage time and the environmental conditions, this prompted us to further investigate a new, practical extraction technology of the *C. cyminum* essential oil. The essential oil contained in *C. cyminum* seeds is usually obtained by hydrodistillation, steam distillation, supercritical fluid extraction (SFE) or conventional solvent extraction. These processes are expensive, or can induce thermal degradation, hydrolysis and water solubilisation of some fragrance constituents. Extracts obtained by conventional solvents contain residues that pollute the foods and fragrances to which they are added (Faborode & Favier, 1996). These disadvantages can be avoided by using a combination technology of organic solvent with low boiling point and steam distillation (OS-SD) process.

Organic solvents with low boiling range between 30 and 50 °C may overcome many disadvantages that conventional solvents with boiling range between 60 and 80 °C bring about in the process of oleoresin extraction, such as difficult separation of product from

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extraction solvent and high remnant solvent (Babatunde, Ajibola, & Ige, 1988; Debet & Gidley, 2006). Success of oleoresin manufacturing industry always uses low cost solvent, that can be recovered. Recovery of organic solvents with low boiling, e.g., *n*-pentane is 98.6% during oleoresin preparation according our past experience (Li, Feng, & Kong, 2004). Application of organic solvents with low boiling in extraction of oil and essential oil have been reported (Carrín & Crapiste, 2008; Kouri, Tsimogiannis, Bardouki, & Oreopoulou, 2007; Tanaka, Haneda, Tanaka, Kimura, & Seki, 1996). However, organic solvents with low boiling range easily expand when heated. This increases dangerous factors in the process of oleoresin extraction, so they are scarcely utilised as extraction solvent. In the present study, we extracted essential oil by using a combination technology of organic solvent with low boiling point and steam distillation in modified extraction equipments. Each test was repeated thrice, the percentages of essential oil extraction and standard error were calculated. This extraction way is a new technique based on the use of a air pump connected to the modified extraction equipment, which may cause low pressure condition. Under low pressure condition, the boiling point of organic solvent is further reduced, and this not only accelerates the removal of remnant solvent, but in comparison with conventional procedures significantly minimises the risk of explosion. Then, essential oil is separated from *C. cyminum* oleoresin with the help of steam distillation. In conclusion, the technology, due to some advantage, e.g., simple equipment, large extraction quantity and low cost, was promising in industrial scale. Compositions of *C. cyminum* essential oil were analysed by gas chromatography (GC) and gas chromatography–mass spectrometry (GC–MS). The aim of this work is to investigate the applicability of the combination technology of organic solvent with low boiling point and steam distillation (OS-SD) for *C. cyminum* essential oil extraction.

2. Material and methods

2.1. Plant material

C. cyminum seeds were collected from the southern part of Urumqi (China) in September 2003. The dried seeds were stored in a dark place (4 °C and 30% relative humidity) until use. The sample was ground in a blender to produce a fine powder (70 mesh).

2.2. Reagents

Ligarine, diethyl ether, dichloromethane and *n*-pentane were purchased from the NanJing JianChen Biotechnology Lt. Cod (NanJing, China). Other reagents used were of analytical grade.

2.3. Effect of extraction parameters on the yield of the extract

2.3.1. Effect of solvents on the yield of the extract

Twenty gram, 70 mesh *C. cyminum* powder was placed in four 500 ml round bottom flasks with a magnetic stirrer. Then four times volume of solvents (ligarine, diethyl ether, dichloromethane, *n*-pentane) were respectively added into the flasks. The extracts were constantly stirred with a magnetic stirrer in the flasks in a heated water bath shaker (DKS-12; Jiaying Zhongxin Medical Instruments Co., Ltd; Jiaying city, China) for extraction (33 °C, 2.5 h) and vacuum filtered to remove solvents, which was reutilized up to two times to obtain maximal yield of *C. cyminum* oleoresin in the same way. Combined filtrate was vacuum evaporated to recover the solvent at 25 °C. Evaporation of the organic solvent under reduced pressure gave the oleoresin extract, which was weighed to give the yield of the extract (*C. cyminum* oleoresin).

The yield of the extract (%)

$$= [\text{Cuminum cyminum oleoresin(g)}/\text{Cuminum cyminum powder(g)}] \times 100\%$$

2.3.2. Effect of temperature on the yield of the extract

Extraction temperature was set at 7, 10, 13, 16, 19, 22, 25, 28 °C, respectively. *n*-Pentane was chosen as solvent and other conditions were the same to those described in Section 2.3.1.

2.3.3. Effect of extraction time on the yield of the extract

Extraction time was set at 3, 4, 5, 6, 7, 8, 9, 10 h, respectively. *n*-Pentane was chosen as solvent and other conditions are the same to those described in Section 2.3.1.

2.3.4. Effect of particles size of powder on the yield of the extract

Particles size was set at 30, 40, 50, 60, 70, 80, 90 mesh, respectively. *n*-Pentane was chosen as solvent and other conditions are the same to those described in Section 2.3.1.

2.3.5. Effect of ratio of liquid to solid (v/m) on the yield of the extract

Ratio of liquid to solid (v/m) was set at 1.5:1, 2:1, 2.5:1, 3:1, 3.5:1, 4:1, 4.5:1, 5:1, respectively. *n*-Pentane was chosen as solvent and other conditions are the same to those described in Section 2.3.1.

2.3.6. Effect of extraction number on the yield of the extract

Extraction number was set at 1, 2, 3, 4, 5, respectively. *n*-Pentane was chosen as solvent and other conditions are the same to those described in Section 2.3.1.

2.4. The optimisation of the experimental conditions

On the basis of single-factor test, an attempt was made to optimise three parameters, which are confirmed to significantly affect the yield of the extract, as ratio of liquid/solid, particle size, extraction time to obtain good yield of *C. cyminum* oleoresin by orthogonal test. Each of these parameters was varied at three levels; temperature at 10, 15 and 20 °C, particle size at 40, 60 and 80 mesh, and time of extraction at 3, 5 and 8 h.

2.5. Separation of essential oil from *C. cyminum* oleoresin by steam distillation

Steam was introduced into *C. cyminum* oleoresin. Essential oil was rapidly carried out with the help of steam and condensed to obtain desired product (40 min). At the same time, non-volatile oil was collected.

The yield of essential oil (%)

$$= [\text{essential oil weight(g)}/\text{Cuminum cyminum powder weight(g)}] \times 100\%$$

2.6. Preparation of *C. cyminum* essential oil by direct hydrodistillation

Twenty g, 70 mesh *C. cyminum* powder was placed in a 500 ml round bottom flask and 250 ml water was added. The flask was connected to a hydrodistillation apparatus and the water was boiled for 3 h. Thereafter, the resulting water extract was removed and the extraction process was repeated with fresh extractant a further three times. The water extracts were combined, filtered, reduced in vacuo (45 °C), freeze-dried and stored at 4 °C. The yield of essential oil was calculated in the same way described above.

The oils, taken in 2 ml of capillary GC grade *n*-pentane and dried over anhydrous sodium sulphate, were stored at –20 °C, subsequently analysed by GC and GC–MS.

2.7. Supercritical fluid extraction (SFE) of *C. cyminum* essential oil using carbon dioxide

Extraction of *C. cyminum* essential oil using supercritical carbon dioxide was conducted by Heikes' method (2001).

A Suprex MPS/225 system (Pittsburgh, PA) in the SFE mode was used for all the extractions. The extraction vessel was a 100 ml stainless steel vessel. The supercritical fluid extraction involved five successive extraction phases with increasing extraction strength (from very mild to very harsh) by subsequently raising the temperature and density of the fluid (Phase 1: $\rho = 0.25$ g/ml; $T = 40$ °C; Phase 2: $\rho = 0.25$ g/ml; $T = 60$ °C; Phase 3: $\rho = 0.63$ g/ml; $T = 60$ °C; Phase 4: $\rho = 0.63$ g/ml; $T = 120$ °C and Phase 5, $\rho = 0.63$ g/ml, $T = 120$ °C plus 5% (v/v) methanol as modifier). The *C. cyminum* powder (30 g, 60–70 mesh) mixed well with 2 mm diameter glass beads was charged into the 10-ml extraction vessel, and extracted statically for 20 min followed by another 6 h dynamically under 30 MPa. The supercritical CO₂ flow rate through the Duraflow restrictor was approximately 0.3–0.4 ml/min (compressed), and the extract in the supercritical fluid was depressed directly into a separate vessel. Total essential oil content was determined.

2.8. Measurement of remnant solvent in essential oil

Remnant solvent in essential oil was measured by headspace gas chromatography (Camarasu, 2000). The GC and headspace parameters are as follows: the carrier gas was helium and the average column linear velocity was 35 cm/s. Nitrogen was used as the make-up gas at a flow rate of 50 ml/min. The injector was maintained at 160 °C with a flow rate of 5:1 and the detector at 250 °C. The column temperature was programmed at 40 °C for 20 min, then raised at a rate of 10 °C/min to 240 °C. The headspace injector parameters were 80–140 °C equilibration temperature, 60 min thermostating time, 3 min pressurisation time, 0.04 min injection time and sample volume of 1.0 ml. The needle and transfer line temperatures were set at 10 and 20 °C higher than equilibration temperature, respectively. The headspace vial was 22-ml capacity and a polyperfluoroethylene coated butyl rubber septum and an aluminium crimp cap were used to seal the vial.

2.9. Analysis of chemical component of essential oil

2.9.1. Gas chromatography

Gas chromatographic (GC) analyses of oil samples were performed employing a Perkin-Elmer gas chromatograph Model 8500 (Perkin-Elmer, Waltham, Massachusetts, USA) fitted with flame ionisation detector (FID), GP-100 printer-plotter and an electronic integrator, using a bonded phase fused silica capillary column BP-1 (25 m length \times 0.5 mm i.d.; film thickness 0.25 μ m) (PerkinElmer Life And Analytical Sciences, Inc. 940 Winter Street Waltham, Massachusetts, USA) coated with polydimethylsiloxane. Nitrogen at a flow rate of 40 ml/min (linear velocity 34 cm/s) and 10 psi inlet pressure was the carrier gas employed. Temperature was programmed from 60 to 220 °C at 5 °C/min ramp rate with a final hold time of 10 min. Injector and detector were maintained at 250 and 300 °C, respectively. The oil samples (0.1–0.2 μ l) were injected neat with 1:80 split ratio.

2.9.2. Gas chromatography-mass spectroscopy

Gas chromatography-mass spectroscopy (GC-MS) analyses of oil samples were carried out on a Hewlett-Packard 5890 gas chromatograph (Hewlett-Packard, EquipNet, Inc. 50 Hudson Road Canton, MA 02021 USA) coupled to a HP 5970 mass-selective detector (MSD) (Hewlett-Packard, EquipNet, Inc. 50 Hudson Road Canton, MA 02021 USA) using a fused silica ultra performance cross-linked

methyl silicone column (50 m length \times 0.2 mm i.d.; film thickness 0.25 μ m). Temperature programming was done from 100 to 280 °C at 4 °C/min. Helium was used as the carrier gas at 1 ml/min flow rate. Mass spectra were recorded over 40–400 amu range at 1 scan/s with ionisation energy of 70 eV and ion source temperature of 250 °C.

3. Result and discussion

3.1. Assay of effect of different solvents on the yield of the extract

For selection of the best organic solvent for *C. cyminum* oleoresin production, different organic solvents (diethyl ether, ligarine, dichloromethane, *n*-pentane), were examined under given condition. As shown in Table 1, maximum extraction yield differed with different solvents and was in turn 16% when using diethyl ether as the solvent, 15.6% when using dichloromethane, 17.89% when using ligarine, 17.78% when using *n*-pentane. The effect of different extraction solvents on extraction rate was in agreement with the solubility/extraction trends reported for other oil crops in the literature (Piggott, Ghisalberti, & Trengove, 1997). Result indicate that maximum yield of the extract was achieved when *n*-pentane was used. Moreover, recovery rate of solvent was also the highest. Therefore, *n*-pentane was used as the solvent for *C. cyminum* oleoresin production in the experiment.

Table 1

Effect of solvents, extraction temperature, extraction time, particles size, ratio of liquid to solid, and extraction number on the yield of the extract.

		The yield of the extract (%)
Solvent	Diethyl ether	16
	Dichloromethane	15.6
	Ligarine	17.89
	<i>n</i> -Pentane	17.78
Extraction temperature (°C)	7	6.2
	10	9.64
	13	11.06
	16	12.06
	19	13.9
	22	17.48
	25	17.67
Extraction time (h)	28	17.6
	3	9.54
	4	10.9
	5	12.19
	6	13.4
	7	15.7
	8	16.97
Particle size (mesh)	9	16.97
	10	16.97
	30	9.5
	40	12.71
	50	15.3
	60	17.88
	70	17.1
Ratio of liquid to solid (v/m)	80	16.72
	90	15.03
	1.5	4.9
	2	6.05
	2.5	8.1
	3	12.63
	3.5	17.9
Extraction number	4	17.98
	4.5	17.96
	5	17.99
	1	7.1
	2	15.3
	3	18.1
	4	18.12
5	18.13	

3.2. Assay of effect of extraction temperature on the yield of the extract

The influence of temperature on extraction was investigated since it affects both the equilibrium (solubility) and mass transfer rate (diffusion coefficient) (Hayes, 1997). To investigate effect of extraction temperature on the yield of *C. cyminum* oleoresin, a study was conducted by employing different extraction temperatures in the range of 7 to 28 °C (Table 1). As can be seen, the yield of oleoresin increased with increasing temperature and reached a peak value at 22 °C. The percent oil yield inversely decreased after this optimum temperature value. It means that the mass transfer of *C. cyminum* component from the surface of the solid phase into the water phase regulated most of the extraction process. Increase of extraction temperature resulted in increase of solvent velocity and quicker mass transfer (Elbashir, Al-Zahrani, Abdul Mutalib, & Abasaheed, 2002; Matsuihiro, Lillo, Sáenz, Urzúa, & Zárate, 2006). The main disadvantage of applying higher temperature is increasing the solvent boil off, reducing effective contact area between solid and liquid phases, and consequently, lower yield of the final extracts. In practice, the best extraction temperature must be selected considering two important factors including safety factor and the yield of the extract. Therefore, 22 °C was chosen as optimum extraction temperature.

3.3. Assay of effect of extraction time on the yield of the extract

Table 1 shows the effect of extraction time on the yield of the extract. Under the given conditions, as shown in Table 1, there was marked correlation between the extraction time and the extraction yield of oleoresin. The yield of the extract was found to increase from 9.54 to 16.97% when the extraction time increased from 3 to 8 h. The yield of the extract no longer increased with further increase in extraction time, suggesting that when *C. cyminum* powder was extracted for 8 h, remanent oil contained in powder particles had been dropped to a minimum value. Similar findings have been reported by Enujiugha and Akanbi (2005) for African bean oil seeds. Boskabady, Kiani, and Azizi (2005) working with finely ground *C. cyminum* found that there was significant increase in oil yield when the extraction time was increased from 2 to 7 h, but yield was either unchanged or decreased slightly when the time was continuously increased. These confirm that extraction time of 8 h is sufficient for optimum extraction. On the basis of preliminary single-factor experiments, we still analyse effect of extraction time on the yield of the extract when quantity of *C. cyminum* powder scale up to 500 g or kg batch for the design of large-scale essential oil extraction systems. Data obtained indicate that effect of optimal extraction time on oil yield is not affected by varying sample quantity (data not given). According to the obtained results, extraction time was chosen between 7 and 9 h for later orthogonal optimisation tests.

3.4. Assay of effect of particle size on the yield of the extract

Table 1 shows the effect of particle size on the yield of the extract. It was demonstrated that the decrease of particle size from 30 to 70 mesh resulted in a rapid increase in the extraction yield of oleoresin, while the increase of particle size from 60 to 90 mesh did not result in an increase of the oil yield. As it was expected, the extraction yield enhanced significantly (*C. cyminum* < 0.05) with decrease of particle sizes, due to the increase of the solubility of the oil components. This is attributed to the decrease of particle sizes, which results in the increase of oil dissolving ability (Cardarelli, Crapiste, & Mattea, 2002; Valdez et al., 2006). The specific surface area of a sample particle is the total surface area contained in a unit mass of plant material. *C. cyminum* particle with high specific surface areas have high oil holding capacities, more adsorption of

solvent, and greater swell potentials. Specific surface is closely tied to particle size distribution. Specific surface is therefore an important parameter (Cardarelli et al., 2002). The specific surface area of sample increases as the particle size decreases, which consequently decreases solvent penetration path length and this influences solubility. However, when the particle size becomes excessively small, this becomes a bottle-neck of raising extraction yield again. This can be explained by the fact that increased specific surface area with reducing particle size raises the adsorbability of particles. As a result, diffusion of solvent inside and outside particles became difficult. This will reduce oleoresin extraction yield and increase difficulty of filtering extraction solution of high density. Therefore, in the present work, we adopt 60 mesh *C. cyminum* particle.

3.5. Assay of effect of ratio of solvent to solid on the yield of the extract

Generally, during the conventional extraction process, the percentage of active component in the liquid phase will increase until equilibrium is reached (Bédouet, Denys, Courtois, & Courtois, 2006; Molero Gómez & Marfinez de la Ossa, 2002). It is important to consider and perhaps examine the ratio of solvent to solid to exploit maximum extractability while scaling up or down the sample preparation method (Shogren, Fanta, & Felker, 2006). In order to determine the optimal ratio of solvent to solid, the effects of ratio of solvent to solid on the extraction yield was examined as shown in Table 1. It can be observed that the extraction rate gradually increased with increasing ratio of solvent to solid and reach the maximal value as the ratio of solvent to solid was 4/1 (v/m) under the given condition. As far as a given quantity of *C. cyminum* powder is concerned, increasing quantity of solvent can decrease concentration of oleoresin in solvent and raise concentration gradient of interface between oleoresin contained in *C. cyminum* particles and solvent. This raises mass transfer ratio and consequently extraction yield. Because oleoresin contained in *C. cyminum* particles has been almost thoroughly extracted, increasing quantity of solvent no longer change extraction yield. Therefore, ratio of solvent to solid (4:1) was adopted in the work. Solvent ratio is important to achieve the best results but also depends upon the batch size and particle size. In single-factor test, extraction yield was not be obviously affected by varying quantity of *C. cyminum* (data were not be given) *C. cyminum* as the ratio of solvent to solid was 4/1 (v/m) under the given condition. On the basis of before-mentioned experiment result (Table 1), the relationship between solvent ratio and particle size will be further studied in later orthogonal optimisation tests.

3.6. Assay of effect of extraction number on the yield of the extract

The effect of extraction number on the yield of the extract is shown in Table 1. The extraction yield was found to increase from 7.1% to 18.1% as the extraction number increased from 1 to 3. Extraction yield no longer significantly changed as the extraction number increased from 3 to 5. For further experiments, the optimum value for extraction number was selected as 3 in our work.

3.7. Optimisation of extraction technology of *C. cyminum* oleoresin

The three factors (extraction time, particle size and ratio of liquid to solid) were conducted through orthogonal optimisation tests under the individual three levels (Table 2). Based on the analytical data obtained in Table 2, the optimal extraction condition was determined as A₃B₃C₃ when all levels of the three factors were considered (Table 2). Therefore, the optimal parameters combinations are as following: temperature 20 °C, extraction time between 7 and 8 h, particle size 80 mesh. Further variance analysis shows

Table 2
Optimisation of extraction parameters of *Cuminum cyminum* oleoresin.

No.	Factors			Extraction rate (%)
	A	B	C	
1	1	1	1	7.07
2	1	2	2	14.13
3	1	3	3	18.8
4	2	1	3	9.78
5	2	2	1	15.56
6	2	3	2	10.03
7	3	1	2	7.97
8	3	2	3	9.63
9	3	3	1	15.45
K ₁	24.82	26.73	38.1	
K ₂	39.32	25.23	32.13	
K ₃	44.28	42.33	38.21	
R	19.46	15.6	6	

A. Ratio of liquid to solid.

B. Extraction time (h).

C. Particle size (mesh).

that the decreasing order of effect of the three factors on extraction yield of *C. cyminum* oleoresin were temperature > extraction time > particle size.

The oleoresin extraction yield of five batches of sample was examined under the optimal extraction condition described as above. As shown in Table 3, mean extraction yield of *C. cyminum* oleoresin was 17.77%.

3.8. Assay of isolation of essential oil from *C. cyminum* oleoresin by steam distillation

Under the optimal extraction condition described as above, average extraction yield of *C. cyminum* oleoresin was 17.71%. Yield of essential oil isolated from *C. cyminum* oleoresin by steam distillation was 4.81%. Consequently, yield of non-volatile oil was 12.2%.

Table 3
Validation test for optimal extraction condition.

Optimum condition			Mean yield ^a (%)
A. temperature (°C)	B. Extraction time (h)	C. Particle size (mesh)	
20	8	80	17.77±1.83

^a Mean ± standard deviation (N = 5).

Table 4
Comparison of different extraction methods.

Index	SFE	OS-SD	HDE
Yield of essential oil (%)	5.24	4.87	3.16
Extraction time (h)	2	<9	12
Appearance	Clear and transparent liquid	Clear liquid	Clear liquid
Colour	Light yellow	Yellow	Light yellow
Odour	Special <i>Cuminum cyminum</i> fragrance	Special <i>Cuminum cyminum</i> fragrance	Special <i>Cuminum cyminum</i> fragrance
Remnant solvent	Zero	<3 ppm	Zero
Relative density (d ₄ ²⁵)	0.911	0.921	0.908
Optical rotation (α)	+5		+7
Refractive index (n _D ²⁰)	1.506		1.510

Note: SFE: supercritical fluid extraction; OS-SD: combination of organic solvent and steam distillation; HDE: hydrodistillation extraction.

3.9. Advantage of combination technology of organic solvent with low boiling point and steam distillation in extracting *C. cyminum* essential oil

As shown in Table 4, compared with supercritical fluid extraction (SFE) using carbon dioxide, despite of the lower total extraction yield (4.87 for OS-SD against 5.24% for SFE) and longer extraction time for combination technology of organic solvent with low boiling point and steam distillation (OS-SD) (9 h for OS-SD against 2 h for SFE), the method can offer larger extraction capacity, and need more simple equipment and technology and from cost point of view, the final extract using OS-SD method was

Table 5
The chemical composition analysis of essential oil from *Cuminum cyminum* seed.

Peak No.	Compound	SFE	OS-SD	HDE
1	2-Methyl-5-(1-methylethyl)-bicyclo [3.1.0] hex-2-ene	0.15	0.14	0.12
2	Cyclohexane,1-methylethylideneyl	-	0.02	0.02
3	α-Pinene	0.40	0.34	0.42
4	Camphene	0.25	0.24	0.21
5	β-Phellandrene	0.17	0.33	-
6	Sabinene	2.47	2.36	2.22
7	β-Myrcene	0.27	-	-
8	3-Carene	0.09	0.04	0.07
9	1-Methyl-2-(1-methylethyl) benzene	3.05	2.98	2.69
10	D-Limonene	0.15	0.14	0.12
11	1,8-Cineole	0.25	0.25	0.43
12	1-Methyl-4-(1-methylethyl)-1,4-cyclohexadiene	6.12	5.93	5.87
13	β-Terpinol	0.36	0.45	0.33
14	L-Fenchone	0.54	0.62	0.58
15	Linalool	-	-	0.11
16	6,6-Dimethyl-2-methylene-bicyclo[2.2.1]heptan-3-one	0.06	0.12	0.09
17	Pinocarveol	0.55	0.61	0.59
18	Anethole	0.07	-	-
19	(5R)-5-Methyl-2-(1-methylethylidene)cyclohexanone	3.65	3.64	3.54
20	Ledene	-	0.29	0.23
21	r-Elementene	0.57	-	0.56
22	Cuminal	31.73	30.63	29.84
23	4-(1-Methylethyl)-1-cyclohexene-1-carboxaldehyde	3.53	3.51	3.41
24	γ-terpinene	17.13	17.12	17.07
25	2-Ethylidene-6-methyl-3,5-heptadienal	9.13	8.94	8.20
26	α-Propyl-benzenemethanol	8.05	7.87	7.84
27	4-(1-Methylethyl)-1,4-cyclohexadiene-1-methanol	0.37	0.44	0.34
28	6-Isopropylidene-1-methyl-bicyclo [3.1.0] hexane	0.06	0.02	0.05
29	Phenol, 2-methoxy-4- (1-propenyl)	-	0.04	0.09
30	trans-Geraniol	0.10	-	-
31	Myrtenal	0.11	0.13	0.06
32	E-2-Nonenal	0.43	0.42	0.38
33	Rhodinol	0.79	0.72	0.69
34	7,11-Dimethyl-3-methylene-1,6,10-dodecatriene	0.26	0.28	0.24
35	2-Isopropyl-5-methyl-9-methylene-bicyclo[4.4.0]dec-1-ene	0.08	-	0.06
36	p-Allylphen	0.42	0.42	0.40
37	E -Hexadecen-6-yne	0.04	0.04	0.01
38	Octahydro-3,8,8-trimethyl-6-methylene-1H-3a,7-methanoazulene	0.16	0.15	0.14
39	Thujopsene	0.42	0.36	0.40
40	1-(1,5-Dimethyl-4-hexenyl)-4-methylbenzene	-	-	0.02
41	5-(1,5-Dimethyl-4-hexenyl)-2-methyl-1,3-cyclohexadiene	0.14	0.12	0.13
42	Copaene	0.08	0.09	0.08
43	β-Sesquiphellandrene	0.09	0.03	0.08
44	Beta-acoradiene	0.09	0.04	-
45	Carotol	0.12	0.15	0.09

Note: SFE: supercritical fluid extraction; OS-SD: combination of organic solvent and steam distillation; HDE: hydrodistillation extraction; -: not detection.

relatively better and more valuable. Compared with the hydrodistillation extraction, essential oil extraction yield for combination technology of organic solvent with low boiling point and steam distillation was significantly higher than the former (4.87% for OS-SD against 3.16% for HDE). Moreover, the method still possessed other advantages, such as shorter extraction time (9 h for OS-SD against 12 h for HDE), lower energy-consumption, and full utilisation of non-volatile oil, in which contains plenty of unsaturated fatty acid, such as oleic acid (26.5–31%), linolenic acid (32–41%) and docosahexaenoic acid (7–21%) (Wang et al., 2006). Unsaturated fatty acids (UFAs) may affect gene transcription, messenger RNA processing and modulate posttranslational modifications of proteins (Kersten, 2002) and are known to suppress lipogenic gene transcription by downregulating the expression of the sterol regulatory element-binding proteins (SREBPs) (Wahle, Rotondo, & Heys, 2003) and they may function as antagonists of liver X receptors (LXR) and as activators/ligands for the peroxisome proliferator-activated receptors (PPARs) (Ham-Pichavant, Sèbe, Pardon, & Coma, 2005; Hannah, Ou, Luong, Goldstein, & Brown, 2001). Undoubtedly, the non-volatile oil in *C. cyminum* seeds has extremely high nutrition and health protection value (Lu, Feng, & Li, 2008). As shown in Table 5, it is interesting to note that the five main chemical compositions (Cuminal, 31.73% for SFE, 30.63% for OS-SD, 29.84% for HDE; Safranal, 17.13% for SFE, 17.12% for OS-SD, 17.07% for HDE; 2-Ethylidene-6-methyl-3,5-heptadienal, 9.13% for SFE, 8.94% for OS-SD, 8.20% for HDE, α -Proyl-benzene-methanol, 8.05% for SFE, 7.87% for OS-SD, 7.84% for HDE, 1-methyl-4-(1-methylethyl)-1,4-cyclohexadiene, 6.12% for SFE, 5.93% for OS-SD, 5.87% for HDE) of essential oil from *C. cyminum* seed employing three different extraction methods are mostly alike and do not display statistical significance. Moreover, forty-five chemical compositions can all be nearly detected in essential oil obtained by the three methods. This indicates that quality of essential oil obtained by the three methods does not show much difference. In addition, remnant solvent in essential oil was less than 3 ppm, which was within the range of safety.

4. Conclusions

The potential of the essential oil by combination technology of organic solvent with low boiling-point and steam distillation (OS-SD) has been compared with the conventional hydro-distillation method and supercritical fluid extraction (SFE) using carbon dioxide, for the extraction of essential oil from *C. cyminum* seeds. The results show that the three variables chosen, namely extraction time, temperature, particle size all have a positive influence on the yield of oleoresin using the extraction technology of organic solvent with low boiling point. GC analysis indicated that the composition of the essential oil by different extraction methods (OS-SD or SFE or HDE) did not demonstrate significant difference. However, the OS-SD method offers important advantages over HDE and SFE, namely: shorter extraction time (9 h against 12 h for hydrodistillation), cost (energy cost or equipment investment is fairly higher for performing hydrodistillation or SFE than that required for conducting OS-SD) and higher yield rate (4.87% against 3.02% for hydrodistillation).

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