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Research Article

A Study of Physical and Chemical Characteristics of Coriander Seeds Oil, Extraction Methods and Long-Term Stability

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ABSTRACT
Several methods of extraction of volatile oils from the *Coriadrum Satinium* L. seeds were experimented, and the best method was determined when the seeds were soaked with organic solvents at room temperature. The hexane is considered a good solvent to extract total and volatile coriander seeds oil. The following physical and chemical properties of extracted coriander seeds oil were studied: refractive index, density, acidity, acid value, moisture, volatile substances, saponification value and absorptivity in ultraviolet UV. It was found that coriander seeds oil is cannot be used as cooking or food oil. The long-term stability was studied for coriander seeds oil. It was found that the main total quantity of coriander seeds oil and its components was decreased interval of time.

Keywords: coriander seeds, long-term, stability, volatile oil, gas chromatography, extraction.

INTRODUCTION

Coriander is one of the oldest medicinal plants known to human, The ancient egyptians planted it and it was used in the therapeutic prescriptions by the Greeks and Romans. It was found in the tombs of the Pharaohs[1-3]. The original home of the coriander plant is the Mediterranean region, and its cultivation was spread in temperate and semi-tropical. The most important producing countries are Syria, Romania, Italy, Russia, Algeria, Morocco, Egypt and Iran[4]. Coriander seeds is good for healing abdominal cramps, removing bloating because it helps with digestion, and activates the immune system[5,6]. It also has many therapeutic properties for diseases nervous, bone, articular, headaches and flu[7]. In addition, coriander seeds oil lowers hypoglycemic by influence on the pancreas[4,8-11] and decreases of the triglycerides and cholesterol in the blood[12]. Coriander seeds volatile oil components are affected by the plant genetic factor, it is also affected by several physiological and environmental factors such as ripeness, climate, soil and storage conditions as well as the extraction method; and the difference in the composition of volatile oils is quantitative rather than qualitative[13]. Coriander is derived from Coriadrum (Latin) and its scientific name is *Coriadrum Satinium* L. Its seeds called coriander seeds[1,14]. There are several important methods to extract coriander seed oil[15-20].

Percolation with water vapor.

Extraction using Organic Solvents

There are two ways for extraction by organic solvents, the first method uses Soxhlet extraction apparatus at solvent boiling point, and the second method depends on soaking in the organic solvents at room temperature.

The physical and chemical properties of plant oils is determined by refractive index, loss on drying, insoluble materials, acidity, saponification value, unsaponifiable materials, iodine value and density[21,22]. Long-term stability studies on plants is considered recent approaches to follow active components in volatile oils, these types of studies are happened under conditions (25ºC ± 2ºC / 60% RH ± 5% RH) in stability chamber[23].

The aims of this research is to find the best method for separation coriander seeds oil and to determine its physical and chemical properties for found that coriander seeds oil is used as food oil. Study of coriander seeds stability under regular storage conditions (25ºC ± 2ºC / 60% RH ± 5% RH)

MATERIALS AND INSTRUMENTS

Materials

Coriander seeds was collected from Aleppo countrysides fields in Syria.

Hexane 99.8%, petroleum ether (b.p. 40-60ºC), diethylether 99.7%, methanol 99.9%, ethanol 99.9%, acetone 99.5%, dichloromethane 99.8%, tetrachloromethane 99%, Potassium hydroxyl 85%, Hydrochloric acid 32% and linalool 97% were obtained from MERCK (Germany).

γ-Terpinen 98% was obtained from Acros Organics (USA).

Geranyl acetate 98% was obtained from Sigma Aldrich (USA).

Instruments

UV/VIS Spectrophotometry, Jasco, model V-530, system software, made in Japan.

GAS Chromatography, Shimadzu, model GC-9A (FID), made in Japan.

Climatic chamber, Angelantoni, model CH1500, made in Italy.

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Table 1: Difference of extraction of coriander seeds volatile oil by different extraction methods

Separation method		Percentage of coriander seeds volatile oil %
Extraction of coriander seeds volatile oil using hexane solvent	Extraction of coriander seeds volatile oil with soaking in the hexane at room temperature	1.41
	Extraction of coriander seeds volatile oil using Soxhlet apparatus	1.39
extraction of coriander seeds volatile oil using percolation with water vapor		1.24

Table 2: change of Percentage of extracted coriander seeds oil by changing the type of solvent

Solvent	Percentage of total coriander seeds oil %
hexane	1.41
Petroleum ether (b.p. 40-60 C°)	1.28
Methanol	1.28
Ethanol	1.27
Acetone	1.29
Dichloromethane	1.42
Tetrachloromethan	1.39

Table 3: The physical and chemical properties of total coriander seeds oil

colour	Slight yelow
Refractive Index (n_d^{20})	1.468
density g/cm^3 (d^{20})	0.871
Acidity (weight / weight %)	0.78
acid value (KOH/g)	0.58
Saponification Value (KOH/g)	186.3
moisture and volatile compounds (weight / weight%)	8.98

Rotator Evaporator, Normschliff, model VV1, made in Germany.

Drying oven, MMM Croup, model Ecocell 55, made in Germany.

Balance, ±0.05mg, Sartorius, model ED224S, made in Germany.

Extraction methods of coriander seeds volatile oil

Two methods to extraction of coriander seeds volatile oil were performed. The aim of this research is to select of the best method to extract the coriander seeds volatile oil.

Extraction method of coriander seeds volatile oil using percolation with water vapor

50 g of coriander seeds were grinded and soaked in distilled water for an hour, then percolated with water vapor until depletion of oil from the specimen. After that, the extraction of volatile oil is completed from the aqueous extract; it was performed using diethyl ether. The etheric extract was evaporated using the rotating evaporator under low pressure at 40° C to get the coriander seeds volatile oil.

Extraction methods of coriander seeds volatile oil using organic solvents

Extraction method of coriander seeds volatile oil using Soxhlet apparatus

50 g of grinded coriander seeds were placed in a paper capsule in the middle part of Soxhlet apparatus. About 250 ml of hexane were added to the flask. The apparatus was assembled and the heating was made by electrical heater till the refluxed solution became colorless. After that, distillation of the resulting extract was performed under normal air pressure to get a hexane drop contains coriander seeds volatile oil. The hexanic extract was evaporated by the rotating evaporator under low pressure at 40° C to get the volatile oil.

Extraction method of coriander seeds volatile oil by soaking in hexane at room temperature

50 g of grinded coriander seeds were weighed and soaked in 250 ml hexane for one week at room temperature. After filtration, the hexane was removed using rotating evaporator at 40°C and the percentage of remaining oil was calculated (the total oil of coriander seeds). After that, the total oil was solved in hexane again, and then the resulting product was distilled using rotating evaporator under low pressure at 40°C. The hexane was removed rotating evaporator to get the volatile oil.

Studying the solvent influence on the quantity of total coriander seeds oil

50 g of fresh and dried coriander seeds were soaked in 250 ml hexane for one week at room temperature, and filtration of the extracted coriander seeds was repeated twice again in the same solvent (hexane).

The extracts were collected then the solvent was removed using rotating evaporator at 40°C. The percentage of total oil to the weight of seeds was calculated. The previous steps were repeated for the solvents: petroleum ether (b.p. 40-60°C), methanol, ethanol, acetone, tetra chloromethane and dichloromethane.

Determination of physical and chemical properties of coriander seeds oil

The physical and chemical properties were determined for coriander seeds oil by soaking in hexane at room temperature.

Determination of Refractive Index

The measurement was carried out at 20°C. The following correction equation was applied for the temperature differences[22].

$n^T = n^{T1} - (T - T_1)\ F,\ \ T_1 < T$

$n^T = n^{T1} - (T_1 - T)\ F,\ \ T_1 > T$

Where:

F: Correction factor = 0.00035

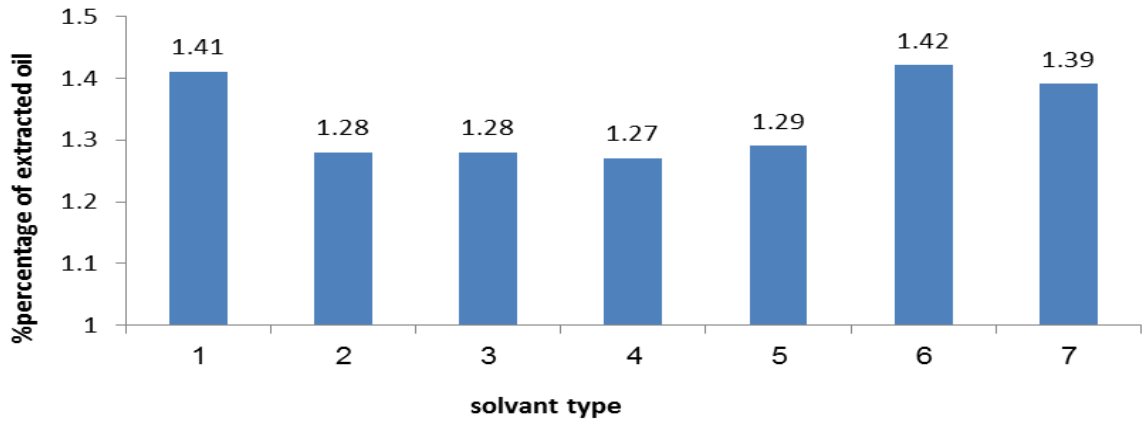
T_1: Temperature at measurement

T: = 20°C

Determination of density

The density was calculated (weight of oil to weight of distilled water) using 10 ml of volumetric flasks at 20°C. The following correction equation was applied for the temperature difference[21,24].

$d^T = d^{T1} - (T - T_1)\ K \quad ,\ T_1 < T$



(1): hexane (2): petroleum ether b.p. 40-60 C° (3): methanol (4): ethanol
(5): acetone (6): dichloromethane (7): tetrachloromethane

Figure 1: Percentage change of extracted coriander seeds oil by changing the type of solvent

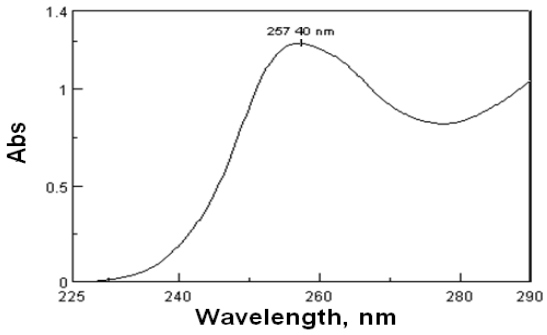


Figure 2: UV spectrum of the coriander seeds oil

$d^T = d^{T1} - (T_1 - T)K \quad , T_1 > T$

Where:

F: Correction factor = 0.00068

T_1: Temperature at measurement

T: = 20°C

Determination of moisture and volatile substances

20g of oil was dried in oven (its temperature increased 10°C/min until 105°C), and placed in the oven for 3 hours at same temperature (105 °C) and cooled in air-free conditions. The weight of sample was calculated. The previous steps were repeated until fixing the weight. The following equation was applied[22]

Moisture and volatile compounds % = (difference of two weights /oil weight) × 100

Determination of acidity and acid value

Acidity value is number of milligrams of potassium hydroxide that is added to neutralize the acidity of 1 gram of oil.

The calculation of acidity and free acidity were carrying out by solving 10 g of oil in 50 ml of neutralized mixture contain equal volumes of ethanol and diethyl ether. The resulting mixture was calibrated with potassium hydroxide in ethanol (0.1N) in the presence of phenolphthalein 1%. The percentage of acidity in oil was calculated from this equation[22].

Free acidity = a M N / 10 P

Where:

a: Volume of Reagent (ml)

M: Molecular weight of oleic acid = 282 g/mol

N: Normally Reagent

P: Mass of Oil (g)

The acidity value was calculated using this equation:

Acidity value = 56.1 a N / P

Where: 56.1 is the molecular weight of KOH.

Determination of saponification Value

Saponification value is number of milligrams of potassium hydroxide that is needed to saponificate 1 gram of oil.

25 ml of potassium hydroxide in ethanol (0.5N) was added to 2 g of oil. The mixture was refluxed for one hour. Titration of hot mixture with HCl (0.5N) was carried out in the presence of phenolphthalein 1%[22].

A control sample was analysed and the saponification value was calculated from the following equation:

Saponification Value = 56.1 N (a - b) / p

Where:

a: Volume of HCl for titration the excess of HCl.

b: Volume of HCl for titration the HCl in the control sample.

N: Normality of HCl (N)

P: Mass of the oil (g)

Absorptivity in Ultraviolet (UV)

The solution (1%) of coriander seeds oil in ethanol was prepared and UV scan was carried out at a range of (190-400 nm), and the result was compared with reference values[21].

Study of long-term effect on the quantity of main components in the coriander seeds oil

Samples of coriander seeds were placed in stability chamber under regular storage conditions (25°C ± 2°C / 60% RH ± 5% RH) for 2 years. Then, GC analyses of the coriander seeds volatile oil were carried out each 6 months[23]. The component of volatile oils was identified qualitatively and quantitatively using gas chromatography GC by comparison with standards, and the following conditions applied:

Chromatographic column is Carbowax 20M, 2 mm × 2 mm.

Micro-sized injected 5 µl.

Nitrogen carrier gas flow capacity of 0.5 ml / min.

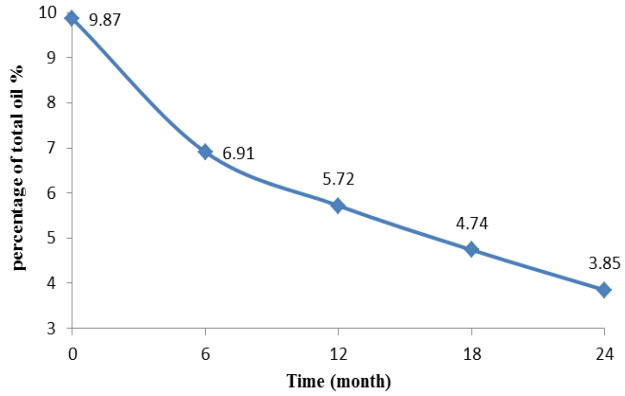


Figure 3: Decreased of total coriander seeds percentage with time under the long-term stability conditions (25ºC, 60%RH).

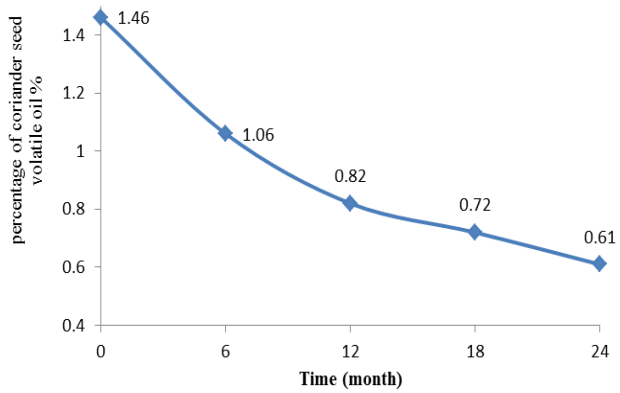


Figure 4: Decreased of coriander seeds volatile oil percentage with time under the long-term stability conditions (25ºC, 60%RH).

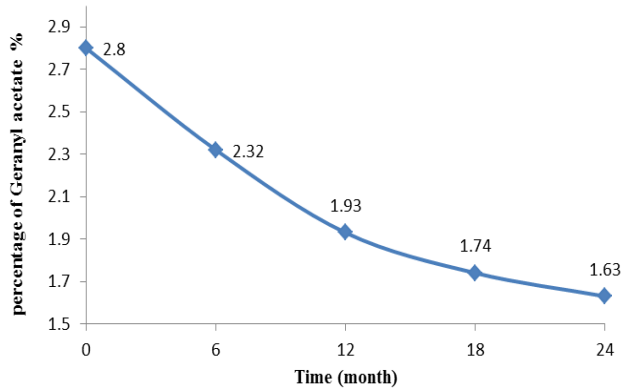


Figure 5: Decreased of Geranyl acetate percentage in volatile oil with time under the long-term stability conditions (25ºC, 60%RH).

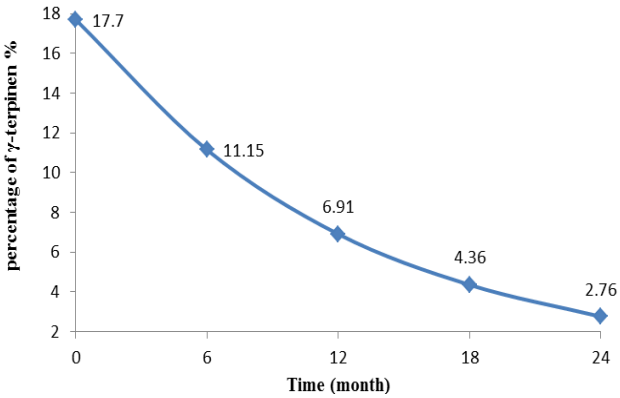


Figure 6: Decreased of γ-terpinen percentage in volatile oil with time under the long-term stability conditions (25ºC, 60%RH).

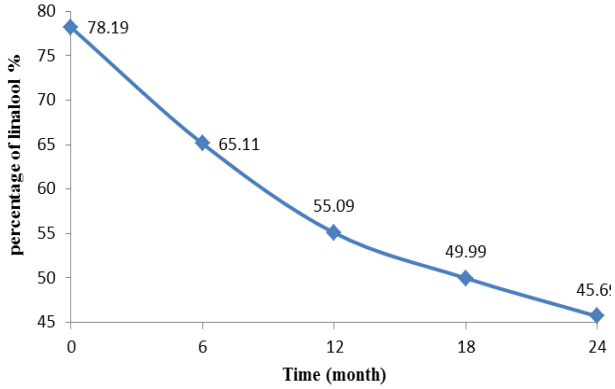


Figure 7: Decreased of linalool percentage in volatile oil with time under the long-term stability conditions (25ºC, 60%RH).

Degree Injector heat 200° C.

The degree of the primitive temperature 90° C is maintained for a period of 4min.

The final temperature 210° C is maintained for a period of 15min.

Heating speed of 2 degrees per minute within the range 90°C to 210°C.

The percentage was calculated for Geranyl acetate, γ-Terpinen and Linalool in the volatile oils using gas chromatographic technique (GC) that were compared with standard compounds. The areas of peaks were compared with the others that belong to standard compounds.

RESULTS AND DISCUSSION

Select of the best method for separation of coriander seeds volatile oil

Several methods were performed to extract coriander seeds volatile oil. The results in table (1) show the different yield of coriander seeds volatile oil by different extraction methods. By comparing the results in Table (1), we found the extraction of coriander seeds volatile oil using organic solvents have higher yield from percolation with water vapor. In addition, Also the extraction using soaking method with organic solvents at room temperature has higher yield from using the Soxhlet apparatus.

Studying of the Solvent Influence on the Quantity of the total coriander seeds Oil

The percentage of total extracted coriander seeds oil was calculated by changing the solvents (hexane, methanol, ethanol, acetone, petroleum ether (40-60ºC), tetrachloromethane and dichloromethane) and fixing the solvent volume and coriander seeds weight. The results are shown in Table (2). The figure (1) shown percentage change of extracted coriander seeds oil by changing the type of solvent. Table (1) and Figure (1) show that among the seven solvents, hexane, dichloromethane and tetrachloromethane are the strongest for the extraction of

Table 4: Decreased of main components quantity in the coriander seeds oil with time under the long-term stability conditions (25ºC, 60%RH).

Time (month)	total oil %	volatile oil %	Geranyl acetate in volatile oil %	γ-terpinen in volatile oil %	Linalool in volatile oil %
0	9.87	1.46	2.80	17.70	78.19
6	6.91	1.06	2.32	11.15	65.11
12	5.72	0.82	1.93	6.91	55.09
18	4.74	0.72	1.74	4.36	49.99
24	3.85	0.61	1.63	2.76	45.69

coriander seeds oil, followed by acetone and then methanol, ethanol and petroleum ether (b.p. 40-60 Cº).

It has been found the hexane, dichloromethane and tetrachloromethane are able to extract the main components of coriander seeds oil when using the different chromatographic methods. Therefore, hexane is considered the best solvent for extraction of coriander seeds oil, dichloromethane and tetrachloromethane should be avoided due its toxicity.

Determination of physical and chemical properties of total coriander seeds oil

The physical and chemical properties of total coriander seeds oil was determination and the results are shown in Table (3).

Maximum absorption was at 257 nm in the UV absorption spectrum of the coriander seeds oil (Figure 2).

Study of long term effect on the quantity of essential components in the coriander seeds oil

Under the long-term stability conditions (25ºC, 60%RH), the contents of quantity of main components in the coriander seeds oil was decreased (table 2). The following Figure show the changing of each the total coriander seeds oil, coriander seeds volatile oil and main components in the coriander seeds with the time. Table (4) and Figures (3), (4), (5), (6) and (7) shows that decreased of total coriander seeds oil, coriander seeds volatile oil, Geranyl acetate, γ-terpinen and linalool quantity in the coriander seeds with time under the long-term stability conditions (25ºC, 60%RH), where:

The total coriander seeds oil decreased 60.99%

The volatile coriander seeds oil decreased 58.21%

Geranyl acetate decreased 41.78%

γ-Terpinen decreased 84.40%

Linalool decreased 41.56%

CONCLUSION

In conclusion, extraction of volatile coriander seeds oil was performed by using different solvents. The best method of extraction was by soaking the coriander seeds in an organic solvent at room temperature. Hexane is a good solvent to extract total coriander seeds oil because of higher yield and less toxic properties. From the physical and chemical properties of the coriander seeds oil, it was found that coriander seeds oil is used as a therapeutic drug, but it cannot be used as cooking or food oil. Studying the long-term stability conditions (25ºC, 60%RH) for two years, it was found that the percentage of coriander seeds oil and its main components were decreased with time.

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