EXTRACTION OF ESSENTIAL OILS FROM JASMINE FLOWER USING SUPERCRITICAL CO_2 CO-SOLVENT EXTRACTION

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I declare that this thesis entitled "*Extraction of essential oils from Jasmine flower using Supercritical CO_2 co-solvent extraction*" is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

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DEDICATION

Special dedication to the memory of my beloved mother and father, Salasiah bt Daud and Che Din bin Yein, my aunty and uncle and all my family members that always inspire, love and stand besides me, my supervisors, my beloved friends, my fellow colleagues, and all faculty members

For all your love, care, support, and believe in me.Thank you so much.

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ABSTRACT

Pure essential oils are derived from various part of the plant. These essential oils have a very high commercial value due to its properties. They are widely used in the various fields of industries, such as perfumery industries and pharmaceuticals. Conventional technique to extracts this oil such as steam distillation is unsuitable since it induce thermal degradation of compounds in the oils. It is for this reason that the extraction of essential oils using supercritical fluid extraction method is said to be the most effective method. It also can avoid contamination of the oil. This extraction technique employed carbon dioxide (CO_2) as solvent due to CO_2 is stateless, odorless, non-toxic and chemical inertness and would not contaminate the environment and products. Thus, the material residues can be used without pretreatment. Beside that, the low critical properties make CO_2 the most preferable solvent in this technique. The used of co-solvent also affect the extraction of these essential oils because it could modify the CO_2 selectivity towards the fragrances compound. This can in turn produce higher quantity of products. For this purpose, two co-solvent had been employed, viz. ethanol and methanol.

ABSTRAK

Pati tumbuh-tumbuhan boleh di dapati di bahagian-bahagian tertentu tumbuhan tersebut seperti pada bunga, daun dan batang. Minyak pati ini mempunyai nilai komersil yang tinggi di dalam pelbagai bidang industri contohnya di dalam indutri wangian dan juga di dalam bidang industri farmasi. Teknik yang biasa digunakan untuk pengekstrakan seperti penyulingan wap air adalah tidak sesuai digunakan, hal ini kerana teknik ini boleh menyebabkan berlakunya thermal digradasi terhadap minyak tersebut. Di sebabkan itulah, teknik 'Supercritical Fluid Extraction' merupakan teknik yang paling efektif untuk tujuan pengekstrakan ini. Teknik ini juga boleh mengelakkan minyak yang di ekstrak itu daripada tercemar. Teknik ini mengunakan karbon dioksida (CO_2) sebagai bahan pelarut kerana ianya merupakan sejenis gas lengai, tidak berbau, tidak mengandungi toksik serta ianya tidak akan mencemarkan alam sekitar dan hasil pengekstrakan. Tambahan lagi, sisa bahan yang terhasil boleh digunakan tanpa rawatan khusus. Selain itu, CO_2 mempunyai ciri-ciri kritikal yang rendah jadi, ini menyebabkan CO_2 adalah pilihan bahan pelarut yang paling sesuai untuk teknik ini. Manakala penggunaan bahan pelarut sampingan juga memberi kesan terhadap hasil ekstrak kerana bahan pelarut sampingan ini mungkin mengubah sedikit pemilihan CO_2 terhadap kompaun wangian itu lantas memberikan hasil yang lebih banyak. Untuk tujuan ini, dua jenis bahan pelarut sampingan telah digunakan iaitu ethanol dan methanol.

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LIST OF ABBREVIATIONS

CO_2	=	Carbon dioxide
FID	=	Flame Ionization Detector
GC	=	Gas Chromatography
GC-MS	=	Gas Chromatography - Mass Spectrometer
RI	=	Refrective Index
SFE	=	Supercritical Fluids Extraction

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CHAPTER 1

INTRODUCTION

1.1 INTRODUCTION

Essential oils (EOs) are highly concentrated essences of aromatic plant. Technically, it cannot be classified as perfumes or fragrance oils. It can be extracted using a variety of methods such as steam distillation and solvent extraction. It is widely used in perfumery, aromatherapy, cosmetics, incense, medicine, household cleaning product as well as flavoring food and drink industries. EOs is also known as volatile oils and ethereal oils. Sometime it may also be referred to as oil of the raw plant materials from which it was extracted, for example oils of clave.

EOs can contain hundreds of organic constituents including hormones, vitamin and other natural elements. Therefore, it is important to understand the effect that the oils have and the way it works before using the EOs as part of an aromatherapy treatment.

There are many types of common essential oils; Rose, Bergamot, Lavender, Jasmine and etc. Each of these raw materials requires different methods of extraction. Plants like Rose and Jasmine which contains very little volatile oils needs solvent-extracted since it is too delicate to be distilled. Hence chemical solvents such as hexane and supercritical CO_2 are employed to extract the important aromatic ingredients.

Supercritical extraction of the fragrance compounds responsible for the fragrance contained in vegetable matrix is a promising field for the industrial application of supercritical food processing. Indeed, there is considerable interest in replacing the steam distillation and solvent extraction process traditionally used to obtain these products.

In most of the extraction, supercritical CO_2 is the supercritical solvent of choice in the extraction of fragrance compound due to its nontoxic properties. Furthermore, it allows supercritical operation at lower pressure as well as temperature. The advantage of using supercritical CO_2 is that the selectivity or solvent power of CO_2 is adjustable and can be set to values ranging from gas-like to liquid-like. Furthermore, through this method, the petrochemical residues in the solvent-extracted product can be avoided.

In this extraction method, co-solvent can also be used along with CO_2 as solvent. The used of co-solvent has been review by many researcher before and they found that it could increase the yield of essential oils.

Since the EOs is very sensitive to temperature and highly volatile, it requires a high degree of expertise to extract the oils from the raw material because the method used are time consuming and an expensive process.

Jasmine EOs has widespread applications. It can be used as antidepressant, antiseptic, antispasmodic, and expectorant for dry skin, labor pains, cough, headache, depression, exhaustion and sensitive skin. On top of that the aroma can be described as warm, floral and exotic.

1.2 PROBLEM STATEMENT

Jasmine flower contains very little volatile oil, and it needs to be solvent extracted since it is too delicate to be distilled. Conventionally, chemical solvents are employed to extract the important aromatic ingredients. In this study, supercritical CO_2 are used as solvent in supercritical extraction.

The reason of choosing CO_2 as main solvent is because of its low critical pressure and temperature where the P_C = 73.8 bar and T_C = 31.03 ^{O}C. It is also inexpensive, and exhibits nontoxic and nonflammable properties. Moreover, supercritical extraction using CO_2 are environmentally benign and efficient extraction technique for solids. In addition, supercritical extraction does not produce thermal degradation or solvent contamination of the product.

The aim of this research is to focus on the study of the effects using co-solvent. Co-solvent will modify the solvent power or the selectivity of CO_2 as well as to enhance the affinity of the solvent mixture towards polar compounds. Co-solvent is useful resource when the compounds to be extracted constitute complex mixture. Since co-solvents are liquid, it will be recovering together with the extracts and do not harmful the environment.

1.3 OBJECTIVE

The research to be described herein has been targeted at employing and subsequently promoting supercritical fluid extraction method (SFE) as an alternative technique to extract essential oils from jasmine flower.

1.4 SCOPE

To achieve the above objectives, the following research scopes had been identified:

i. To determine the optimum conditions (pressure and co - solvent) in extraction of essential oils using supercritical co-solvent fluid extraction method.

ii. To evaluate the quality and yield of the jasmine extracted.

iii. To analyze the product compositions obtained through this extraction process.

CHAPTER 2

LITERATURE REVIEW

2.1 ESSENTIAL OILS BACKGROUND

Essential oils are liquid that is generally distilled from various parts of plant that have strong aromatic components. These aromatic substances are made up of different chemical compounds that can be found naturally in the plant, for instance, alcohol, hydrocarbons, phenol, aldehydes, esters and ketones. Besides that, it may also contain hundreds of organic constituents, including hormones, vitamins and other natural elements. Essential oils 75 to 100 times more concentrated than the oils in dried herbs [1].

For the plant, essential oils act as agent to attract or repel insects (odors of the flowers); Leaf oils, wood oils, and root oils may serve to protect against plant parasites or depredations by animals as well as anti-bacterial agent which is utilizing the hormone in the oil. All in all there are only about 700 aromatic plants aromatic among all types of plants in the world. Thus they are all important for the production of essential oils [2].

Unlike fragrance oils or perfume, essential oils are derived from the true plants. Perfume oils are unnaturally created fragrances since it contains artificial substances. As such, it does not offer the therapeutic benefit that essential oils do. Consequently pure essential oils are very expensive. Fortunately, they are also highly effective because a few drops at a time are sufficient to achieve the desired effects.

These oils could promote healing of the body and the mind. Typically, these essential oils are used in three primary ways:

i. As odorants in cosmetics, perfumes, detergents, soap and etc
ii. As flavors in bakery goods, candies, meat, soft drinks, and also as food additives.
iii. As pharmaceuticals in dental products and group of medicines.

Today, we could also easily find synthetic essential oils in the market where the price would be cheaper than the pure ones. There are a few differences between synthetic essential oils and pure essential oils. Synthetic essential oils are produced by blending aromatic chemicals mostly derived from coal tar.

These oils may duplicate the smell of the pure essential oils, but the complex chemical components of each essential oil created in nature determine its true aromatic benefits. While synthetic essential oils are not suitable for aromatherapy, they add an approximation of the natural scent to crafts, potpourri, soap and perfume at a fraction of the cost. The reason of these synthetic products is mainly to reduce the cost of production.

The oil of the essential oils bears the name of the plant from which it was derived, for example Rose oil, and Bergamot oil. Such oils were called essential oils because they were thought to represent the very essence of odors and flavors.

The odors and flavor of these oils is usually dependent upon this oxygenated compound. Many oils are terpenoids, a few oils are benzene derivatives. Table 1 shows the important constituents of the more common essential oils [3].

Table 2.1: Essential Oils from Some Natural Plants.

Name	Part of the plant used	Botanical name	Important constituents	Uses
Lemongrass and citronella	Leaf	*Cymboposon spp*	Citral Citronella Terpenes	Perfumery Disinfectant
Eucalyptus	Leaf	*Eucalyptus globules* *Eucalyptus citriodora* *Eucalyptus dives*	Cineale Citronella Terpenes	Not mention
Cinnamon leaf	Leaf	*Cinnamon zeylanicum*	Eugenol	Used to make artificial vanilla
Clove	Bud	*Eugenia caryophyllus*	Eugenol	Dentistry flavouring
Turpentine	Not mention	*Pinus spp*	Terpenes	Paints
Lavender	Flower	*Lavendula intermedia*	Linalool	Perfumery
Sandalwood	Wood	*Santaium album*	Sanatols	Perfumery
Nutmeg	Nut	*Myristica fragrans*	Myristicin	Not mention
Almond	Nut	*Prunis communis*	Benzaldehyde	Not mention
Corainder	Seed	*Coriandrum sativum*	Linalool Terpenes	Not mention

2.1.1 Properties and uses of the essential oils

Each essential oil has it own properties and uses which can be classified and identified accordingly to the type of plant it was derived. Table 2.2 shows the properties and uses of the top essential oil [4]. From Table 2.2, it can be concluded that the significant use of the essential oil is mainly contribute to pharmaceuticals industry where most of it have the anti-depressant properties.

Table 2.2: Properties and Uses of the Top Essential Oils.

Essential oil	Biological Name	Properties	Uses
Clory Sage	*Salvia Sclarea*	Warming, soothing, antiseptic, anticonvulsive, astringent, antiphlogistic, digestive, deodorant, tonic, uterine, bactericidal, antidepressant.	Menstrual problems, anxiety, depression, high blood pressure, acne boils, oily skin and hair, cramp, migraine, the genitor-urinary system disorders such as amenorrhoea, wrinkles, ulcers.
Eucalyptus	*Eucalyptus Globulus*	Antiseptic, analgesic, antineuralgic, antirheumatic, antispasmodic, diuretic, expectorant, antiviral, hypoglycaemic, febrifuge, vulnerary, depurative, stimulant.	Muscular aches and pains, poor circulation, rheumatoid arthritis, asthma, bronchitis, flu, cold, epidermics, chicken pox, headaches, neuralgia, throat infections, skin disorders such as burns, cuts, herpes, wounds, insect bites.
Geranium	*Pelargonium Graveolens*	Soothing, refreshing, relaxing, antidepressant, astringent, antiseptic, antihaemarrhagic, deodorant, diuretic, fungicidal, anti-inflammatory	Anxiety, adrenocortical glands and menopausal problems, sore throat, tonsillitis, cellulites, engorgement of breast, broken capillaries, eczema, hemorrhoids, oily complexion, mature skin, ulcers, wounds.
Jasmine	*Jasminum Officinale*	Analgesic (mild), antidepressant, anti-inflammatory, antiseptic, antispasmodic, aphrodisiac, carminative, cicatrisant, expectorant, galactagogue, sedative, tonic (uterine)	Depression, nervous exhaustion and stress related conditions, jasmine is said to produce the feeling of optimism, confidence, euphoria, and it is especially good in cases of apathy, indifference, or listlessness. Jasmine is also used for catarrh, coughs, laryngitis, dysmenorrhoea, labor pains, uterine disorders, skin problem such as dry, greasy, irritated, sensitive skin, and for muscular spasms and sprains.
Lavender	*Lavendula Vera Officinalis*	Analgesic, anticonclusive, antidepressant, antimicrobial, antirheumatic, antiseptic,	Excellent first aid oil. It soothes cuts, bruises and insect bites. One of the most versatile therapeutic essences. For

		antispasmodic, antitoxic, deodorant, sedative, diuretic, choleretic, hypotensive, stimulant, tonic, vulnery, cytophylatic, insecticide	nervous system disorders such as depression, headache, hypertension, insomnia, migraine, sciatica, shock. Useful in treating skin conditions such as acne, allergies, athlete's foot, boils, dandruff, dermatitis, sunburn, eczema. Treatment of disorders such as rheumatism, throat infections, flu, bronchitis, and asthma.
Lemon	*Citrus Limonum*	Refreshing, antiseptic, stimulating, anti-anaemic, antirheumatic, antisclerotic, antitoxic, hypertensive, antiscorbutic, bactericidal, insecticidal, astringent, tonic,	Warts, depression, acne and indigestion, arthritis, cellulites, high blood pressure, nosebleeds, obesity, poor circulation, rheumatism, asthma, throat infections, bronchitis, cold, fever, flu. Treatment of anemia, brittle nails, corns, mouth ulcers, greasy skin, cuts, spots, and varicose veins.
Peppermint	*Menthe Piperita*	Digestive, cooling, refreshing, mentally stimulating, analgesic, anti-inflammatory, antimicrobial, antiseptic, antiviral, astringent, expectorant, stomachic, hepatic, cordial, antispasmodic.	Muscle fatigue, bad breath, toothache, bronchitis, indigestion, and travel sickness, neuralgia, muscular pains, asthma, sinusitis, spasmodic cough, cramp, dyspepsia, skin problem such as acne, dermatitis, ringworm, scabies, and nausea.
Ylang Ylang	*Cananga Odorata ver genuina*	Antidepressant, anti-infections, euphoric, relaxant, antiseptic, hypotensive, aphrodisiac, nervine, regulator, sedative (nervous), stimulant (circulatory), tonic	Depression, nervous tension, high blood pressure, hyperpnoea, (abnormally fast breathing), tachycardia, digestive upsets. For skin care such as hair growth, acne, hair rinse, oily skin, irritated and insect bites. For nervous system disorders such as frigidity, impotence, insomnia.

2.1.2 Hazardous essential oils

Not all essential oil are safe to use in aromatherapy. This is due to the high toxicity levels that the essential oils might have. Some of the oil can be hazardous as they can cause severe dermal irritation and even damage the mucous membranes and delicate stomach lining in undiluted form. Hence dermal application should be avoided as a general practice; it is advisable to use essential oils only for external remedies. Oils that fall under this category are bitter almond, calamus, camphor (brown & yellow), cassia, cinnamon (bark), fennel (bitter), pine (dwarf), rue, sage (common), thyme (red), wintergreen, garlic, onion, mustard and wormwood [5].

2.1.2.1 Toxicity

Essential oil such as Ajowan, Basil (exotic), Camphor (white), Cassia, Cedarwood (Virginian), Cinnamon (leaf), clove (bud), coriander, Eucalyptus, fennel (sweet), hyssop, juniper, nutmeg, pepper (black), sage (Spanish), tagetes, thyme (white), turmeric, should be used only on dilution (at least 1:3) and for a maximum of two weeks due to toxicity levels.

2.1.2.2 Photo toxicity

Some oils can cause skin pigmentation if the applied area is exposed to direct sunlight. Essential oils such as bergamot, cumin, ginger, lemon, lime, and orange should not be used either neat or on dilution on the skin, if the area will be exposed to direct sunlight.

2.1.2.3 Pregnancy

Essential oils should be used in half the usual stated amount during pregnancy due of the sensitivity of the growing child. Oils of adjoin, angelica, anise star, aniseed, basil, Cedarwood (all types), celery seed, cinnamon leaf, citronella, clary sage, clove, cumin, fennel (sweet), hyssop, juniper, nutmeg, Spanish sage, and thyme (white); should be totally avoided during pregnancy.

2.1.2.4 High blood pressure

Oils of hyssop, rosemary, sage (Spanish and common) and Thyme are to be avoided in case of high hypertension.

2.1.2.5 Dermal/skin irritation

Oils of basil (sweet), black pepper, borneol, cajeput, caraway, Cedarwood (Virginian), cinnamon (leaf), clove (bud), eucalyptus, garlic, ginger, lemon, peppermint, pine needle (scotch and longleaf), thyme (white) and turmeric; especially if used in high concentration may cause irritation to the skin [5].

2.1.3 Safety information of essential oils

Since some of the essential oil can promote hazard to the user, there are a few safety precaution should be emphasize when handling with these hazardous essential oils. The following step should be considered as a guideline to proper used of these precious aromatherapy arts.

i. Essential oils should never be used undiluted on the skin.
ii. Some oils can cause allergic reactions in some individuals.
    Do a skin patch on a small area of skin when using new oil for the first time. Place a small amount of the diluted essential oil inside of your elbow and apply a bandage. Wait about 24 hours to see if there is any form of reaction. This step should not be ignoring even if a particular essential oils is not known to cause irritation.
iii. Some essential oils should be avoided during pregnancy or by those with asthma, epilepsy, or with other health conditions.
iv. Never let children use essential oils without the presence of an adult knowledgeable about their use.
v. Essential oils should not be taken internally.
vi. Essential oils are flammable.

2.2 ESSENTIAL OILS QUALITY

To ensure that each of the oils is in highest quality as well as completely unadulterated, some scientific analyses are required on every essential oil. These are four major analyses that could be done.

2.2.1 Specific Gravity

The weight of essential oils is measured at $25^{O}C$. Every oil is made up of unique constituents. At given temperature, these constituents have a predictable weight. If the oil has been contaminated, the weight may be thrown off.

2.2.2 Optical Rotation

Measures direction, left or right and the degree to which light rays bend or rotate as they pass through an essential oil. If the oil has been contaminated, the speed and degree of refraction may be thrown off.

2.2.3 Refractive Index

Measures the speed at which light passing through the oil refracted. If the oil has been contaminated, the speed and degree of refraction may be thrown off.

2.2.4 Gas Chromatography

Separates the individual constituents of an essential oil and measures the amount of each constituent present. It confirms an oil botanical identity by comparing presence and amount of each constituent with its standard sample [1].

2.3 JASMINE FLOWERS

Table 2.3: Brief Description of Jasmine Flower

Common name	Jasmine
Botanical name	*Jasminum Grandiflorum,* *Jasminum Officinale*
Local name	Melur
Origin	France, Egypt, India
Family	Oleaceae
Color	Orange to brown, deep brown with a golden tinge
Extracted from	Flower
Perfumery note	Middle/Base
Chemical constituents	Benzyl acetate, linalool, benzyl alcohol, inclole, Benzyl benzoate, cis-jasmone, geraniol, methyl anthranilate, benzoic acid, benzaldehyde, y-terpineol, isophytol, cis-3 hexenyl, benzoate
Aromatic description	warm, floral, exotic
Properties	Antidepressant, Antiseptic, Antispasmodic, Expectorant.

Figure 2.1 Jasmine Flower

2.3.1 A background of Jasmine Flowers

Jasmine is a genus of shrubs and climbing vines with 200 species, native to tropical and warm temperature regions. The majority of species grow as climbers on other plants or on structures. The leaf shape is simple, trifoliate or pinnate with up to nine leaflets.

Jasmine flowers are white in most species, but with some species being yellow flowered. Unlike most genera in the Oleaceae which have four petals, jasmine often have five or six petals. They are often strongly and sweetly scented. Flowering is in spring or summer in most species. Nonetheless, few species like *J. nudiflorum* flowering in winter on the bare branches of this deciduous species. Normally jasmine flowers are picked at night where at the period, the aroma is most intense.

The common name 'jasmine' is often given to unrelated plants with pale, sweetly-scented flowers and dark green leaves, such as *Trachelospermum* species (confederate or star jasmine), *Gardenia jasminoides* (Cape jasmine), and *Gelsemium* species (Carolina jasmine).

2.3.2 Benefits of Jasmine Flower

Jasmine essential oil is one of the most expensive oils due to its potency and incredible fragrance power. *Jasmine grandiflorium* generally works with all oils. It helps to round out scents, and tends to work particularly well with other aphrodisiac oils such as Ylang Ylang and Sandalwood.

Jasmine essential oils are believed to encourage cell growth as well as increase skin elasticity. It is also used to help with muscle spasms and sprains. Additionally, it is recommended for use during childbirth, but not until the later stages of labor. At that point, it can be very useful as it strengthens the uterine contradictions and relieves pains. It is also effective in post-natal depression and promotes the flow of breast milk. Beside that it also makes a delightful uplifting perfumes or as room fragrances [9].

2.4 EXTRACTION OF ESSENTIAL OILS

2.4.1 Introduction

Essential oils have high alcohol components. Therefore, it has a higher volatility and a fast evaporation rate. In order to get the best quality and quantity of essential oils, extraction procedure seems to hold the key-controlling step. Factors worth considers are types of plant, chemical constituents of oils, location of oils within the plant (root, bark, wood, branch, leaf, flower, fruit and seed) and choosing the right extraction method.

Some plant like rose and jasmine contain very little essential oil. Their important aromatic properties are extracted using a chemical solvent. The end product, known as an absolute, contains essential oil along with other plant constituents. Though not a true essential oils, absolute are commonly used for fragrance cosmetic product like fine perfumes. Some of the important extraction methods are described herein.

2.4.2 Availability of extraction methods

2.4.2.1 Supercritical Fluid Extraction

When CO_2 is subjected to high pressure, the gas turns into liquid. This liquid is an inert and safe solvent which can be used to extract the aromatic molecules in a process similar to that used to extract absolutes. The chief advantage, of this technique is that no solvent residue remains. This is because at normal pressure and temperature, the carbon dioxide can simply slip back to gas phase and evaporates.

Compare to the steam distillation method the products of supercritical extraction seems to have richer, and more intense scent, since more aromatic chemicals are extracted through this process.

2.4.2.2 Solvent Extraction

Another method of extraction used on delicate plants is solvent extraction, which yields a higher amount of essential oils at a lower cost. A hydrocarbon solvent is added to the plant material to help dissolve the essential oil. When the solutions are filtered and concentrated by distillation, a substance containing resin, or a combination of wax and essential oil (concrete) remains. From the concentrate, pure alcohol is used to extract the oil and when the alcohol evaporates, the oils are left behind. This is not considered the best method for extraction as the solvents can leave small amount of residue behind which could cause allergies and affect the immune system.

2.4.2.3 Steam Distillation

Steam distillation is a special type of distillation process (separation process) for temperature sensitive materials like natural aromatic compounds. Through this process the botanical material is placed in a still and steam is forced over the material. The hot steam will help to release the aromatic molecules from the plant material. The molecules of these volatile oils are then escape from the plant material and evaporate into the steam. The temperature of the steam therefore needs to be carefully controlled. The temperature should be just high enough to force the plant material to release the essential oils, yet not too hot as it can degrade the plant material or the essential oils.

The steam containing the essential oil is passed through a cooling system to condense the steam, which then form a liquid from which the water and the essential oils is then separated. The steam is produced at greater pressure than the atmosphere and therefore it boils at above $100^{O}C$ which facilitates the removal of the essential oil at a faster rate. By doing so, it could prevent damage to the oil as well [12].

One example of application is Lavender which is heat sensitive. Using this method, it could prevent the oil from damage and ingredients like linalyl acetate will not decompose to linalool and acetic acid.

2.4.2.4 Enfleurage

Some flowers, such as jasmine or tuberose, have very low contents of essential oil or too delicate that heating them would destroy the blossoms before it could release the essential oils. In such cases, this method of extraction is sometimes used to extract the essential oils. Flower petals are placed on trays of odorless vegetable or animal fat, which will absorb the essential oils. Every day or every few hours, after the fat has absorbed the essential oils as much as possible, the depleted petals are removes and replaced with fresh ones.

This procedure continues until the fat or oil becomes saturated with the essential oil. Adding alcohol to this mixture will separate the essential oil from the fatty substance. After that, the alcohol will be evaporated and only the essential oils remains. One disadvantage of this process is it is very labor-intensive. Therefore, it is a very expensive method to obtain essential oil [13].

2.5 SUPERCRITICAL FLUID EXTRACTION

2.5.1 Properties of Supercritical Fluid

Supercritical fluid is defined as a substance above its critical temperature (T_c) and critical pressure (P_c). The critical point represents the highest temperature and pressure at which the substance can exist as a vapor and liquid in equilibrium. The phenomenon can be easily explained with reference to the phase diagram for pure carbon dioxide as shown in Figure 2.2 [10].

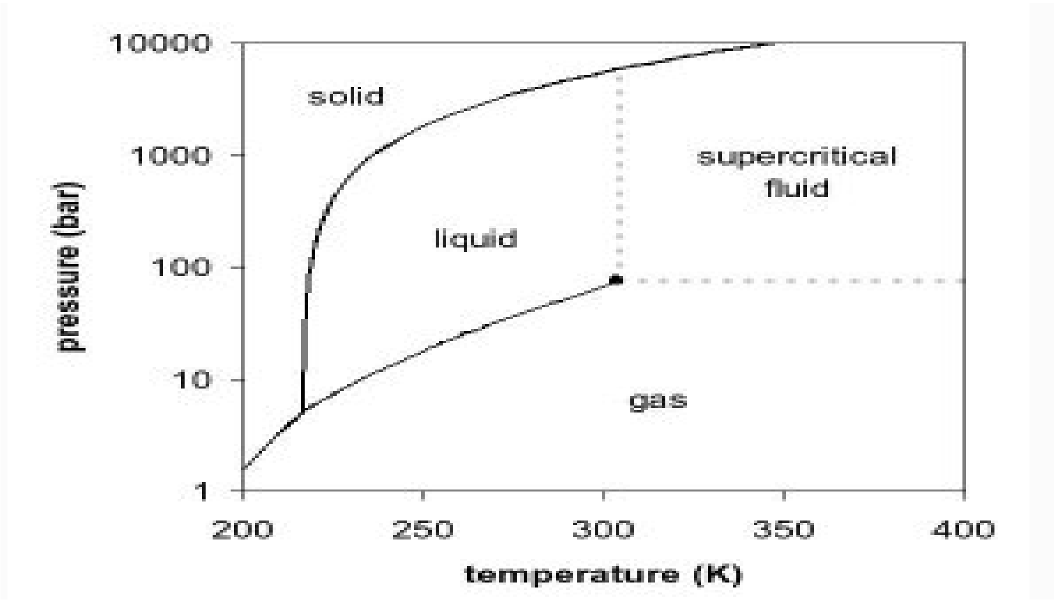

Figure 2.2 Phase Diagram of Carbon Dioxide

Supercritical fluid has unique ability to diffuse through solids like a gas as well as dissolving materials like a liquid. Additionally, it can readily change in density upon minor changes in temperature or pressure. These properties make it suitable as a substitute to organic solvents extraction in a process called Supercritical Fluid Extraction (SFE).

2.5.2 Solvent of supercritical fluid extraction

The choice of the SFE solvent is similar to the regular extraction. Principle considerations are the followings.

i. Good solving property
ii. Inert to the product
iii. Easy separation from the product
iv. Cheap
v. Low P_c because of economic reasons

Carbon dioxide is the most commonly used supercritical fluids, due to primarily to its low critical parameters ($T_c = 31.1^oC$, $P_c = 73.8$ bar). However, several other supercritical fluids have been used in processes. The critical properties of some commonly used solvent in supercritical fluids extraction are listed in Table 2.4.

Table 2.4: Critical Properties of Various Solvents.

Fluid	Critical temperature (K)	Critical pressure (bar)
Carbon dioxide	304.1	73.8
Ethane	305.4	48.8
Ethylene	282.4	50.4
Propane	369.8	42.5
Propylene	364.9	46.0
Trifluoromethane (fluoroform)	299.3	48.6
Chlorotrifluoromethane	302.0	38.7
Trichlorofluoromethane	471.2	44.1
Ammonia	405.5	113.5
Water	647.3	221.2
Cyclohexane	553.5	40.7
n-Pentane	469.7	33.7
Toluene	591.8	41.0

Organic solvents are usually explosive and this fact makes the investment more expensive because the SFE unit should be explosive proof. The organic solvents are mainly used in petrolchemistry. Among all the solvents, CFC are very good solvents in SFE due to their high density, but the industrial use of choro-fluoro hydrocarbons are restricted because their effects on the ozonosphere. CO_2 is the most widely used fluid in SFE.

Beside CO_2, water is the other increasingly applied solvent. One of the unique properties of water is that, above its critical point (374^oC, 218 atm), it becomes an excellent solvent for the organic compounds and very poor solvent for inorganic salts. This property gives the chance for using the same solvent to extract the inorganic and the organic component respectively.

2.5.3 Supercritical CO_2 extraction and its characteristics

From Figure 2.2 and Table 2.4, it is shown that the critical point for carbon dioxide occurs at pressure of 73.8 barg and temperature of 31.1°C. Thus, the supercritical extraction on the compounds responsible for the fragrances contained in vegetable matter is a promising field for the industrials application of supercritical fluid processing. Indeed, there is considerable interest in replacing the steam distillation and solvent extraction processes traditionally used to obtain these products.

CO_2 is the supercritical solvent of choice in the extraction of fragrance compounds, since it is non-toxic and allows supercritical operation at relatively low pressure and near room temperature. Generally speaking, supercritical CO_2 behaves like a lipophilic solvent but, compared to liquid solvent it has certain advantages like its selectivity is adjustable. Its characteristic also can be set to values ranging from gas-like to liquid-like [12]. Furthermore, less solvent residues left in the products. The other advantages of using SFE are shown here:

i. It is safe since CO_2 is cheap. On top of that, it is easily obtained and does not burn and explode easily thus removing the danger faced by organic solvent extraction.
ii. The solvents recovery is simple and convenient. Therefore, it is not an energy intensive process.
iii. CO_2 is stateless, odorless, non-toxic and chemical inertness and would not contaminate the environment and products. Thus, the material residues can be used without treatment.
iv. The permeability is strong. So, the extractive time may be greatly shorter than that of the extraction time of common solvent.
v. Manipulation of temperature is near to room temperature. Therefore, it is suitable for treating heat sensitive material. Using this method can obtain a high quality extraction product.

CHAPTER 3

METHODOLOGY

3.1 INTRODUCTION

This chapter focuses on the achievement of the conceptual study, laboratory work, analyzing and completion of the project. The detailed experimental procedure will be discussed throughout this chapter. There are three main stages in achieving jasmine essential oils through this experiment.

i. Sample preparation
ii. Jasmine essential oil extraction
iii. Analysis

3.2 OVERALL METHODOLOGY

The experiments were run according to the standard procedures outlined in Section 3.3.2.2. Experimental work was divided into two parts. The first part is to determine the optimum operating condition (pressure) where it had been conducted with three different pressures followed by examining the effects of using co-solvent.

Experimental results were analyzed using appropriate method and apparatus. It consists of analyses of the optimum condition, optimum co-solvent as well as the identifications of the extracts constituents. Figure 3.1 shows the outline of the methodology employed in this work.

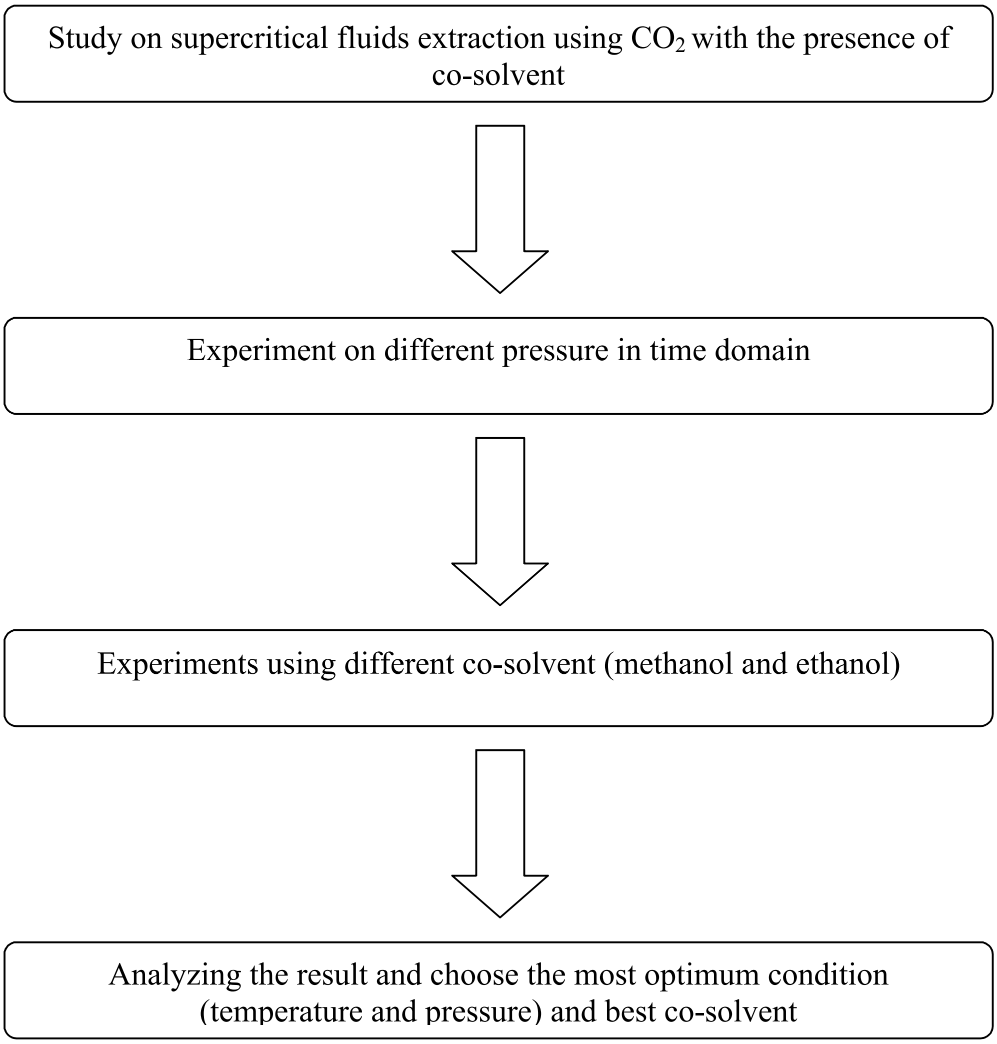

Figure 3.1 Outline of the overall methodology

3.3 EXTRACTION EXPERIMENTAL WORK

3.3.1 Sample Preparation

Jasmine flowers which were about to bloom were used. Since useful components are found in the petals, only the petals were used as sample. The selected jasmine's petals were dried in room temperature for about two to three days and blended to maximize the contact surface between the solute and the solvent.

3.3.2 Apparatus and Procedures

3.3.2.1 Supercritical Fluids Extraction Unit

Figure 3.2 shows the schematic diagram of the supercritical fluid extraction facility in Department of Chemical and Process Engineering, Universiti Kebangsaan Malaysia. The diagram shows that the main components of the SFE unit are pump, extraction chamber, a recovery chamber and a collection device.

Extraction from solids was performed as a semi batch process with continuous flow of CO_2. Liquid CO_2 was pumped from a reservoir, heated and pressurized to reach the supercritical conditions. Supercritical CO_2 and co-solvent is then enter the extraction chamber where contact with solid particles (jasmine flower) occurs. Afterward, the solute, CO_2 and co-solvent leave the extractor and extract is precipitated in separators, where CO_2 becomes gaseous. CO_2 was bled until the system pressure fell to atmospheric pressure.

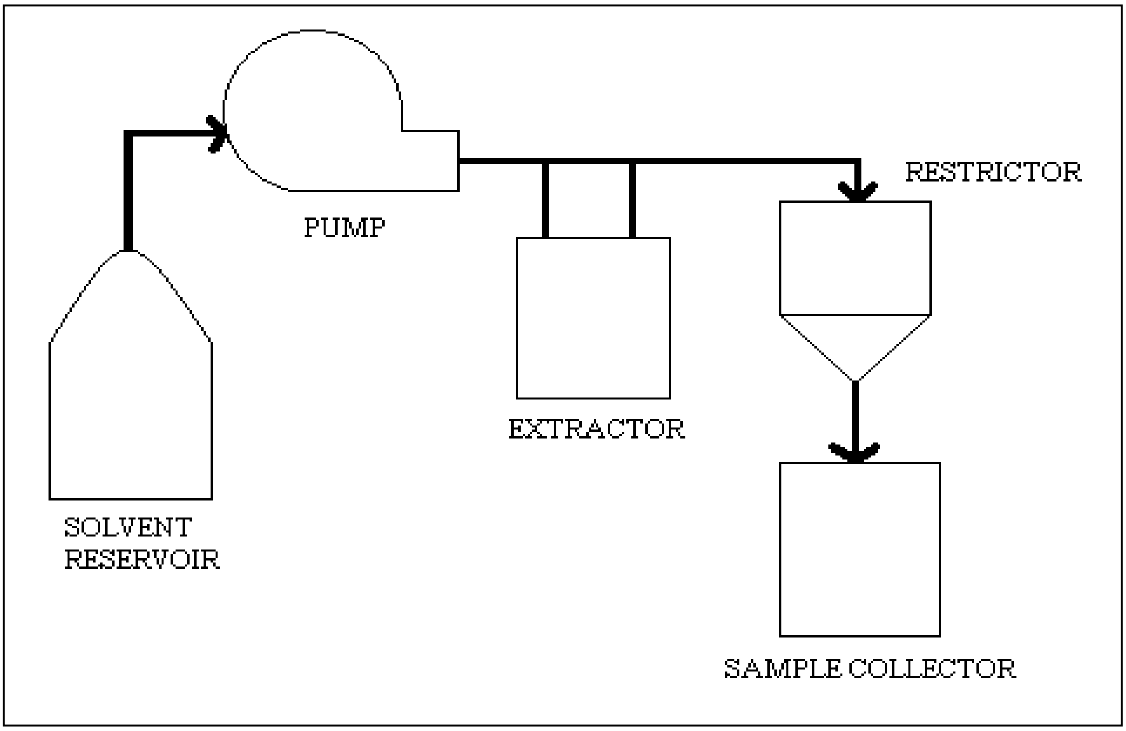

Figure 3.2 Schematic diagram of the supercritical fluid extraction unit

Figure 3.3 shows the facilities of the supercritical fluid extraction in Universiti Kebangsaan Malaysia. This unit comprises of CO_2 pump, CO_2 reservoir, co – solvent pump, back pressure regulator, pressure transmitter, sample collector as well as multipurpose oven where the extraction process took place in the extraction vessel inside the oven. This unit also consists of chiller where it is used to compress CO_2. Detailed function of each equipment will be discussed in section 3.3.2.2.

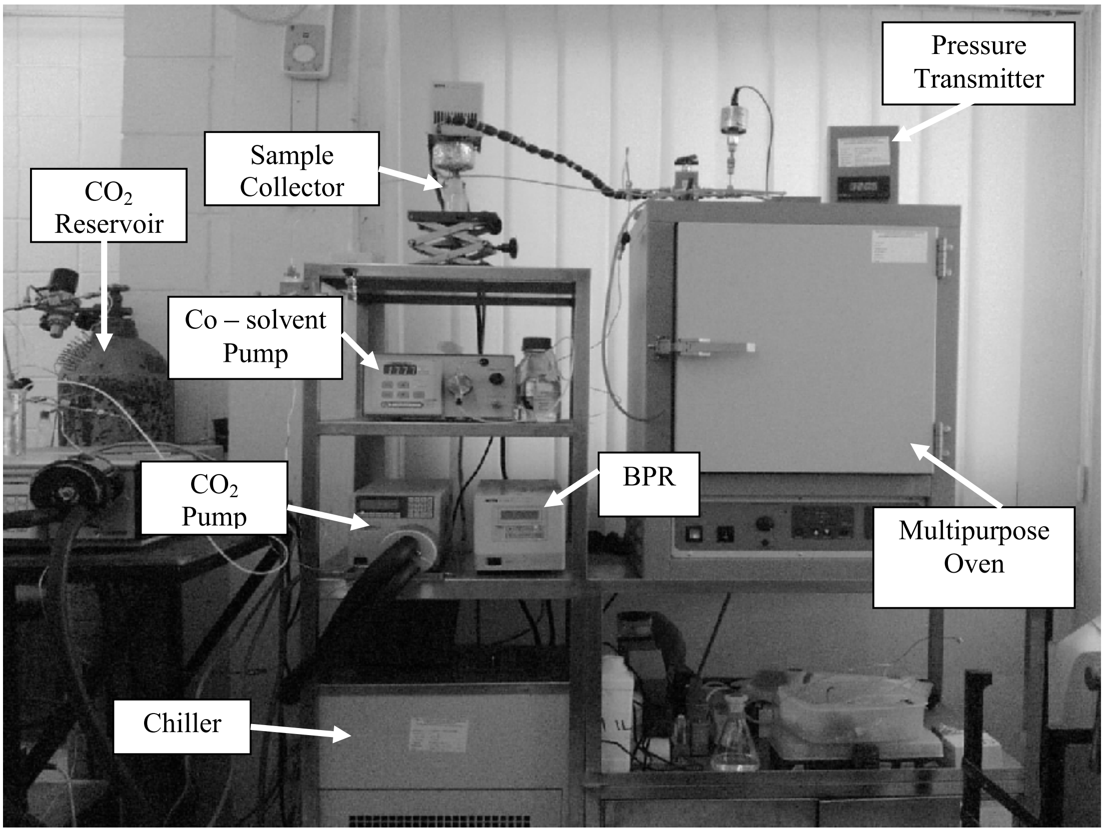

Figure 3.3 Supercritical fluid extraction laboratory scale

3.3.2.2 Experimental work

I. Start-Up of Operations

The chiller was set at 2°C to cool the CO_2 pump before switching on the CO_2 pump, co-solvent pump, back pressure regulator (BPR) and oven. All valves were ensuring to be in closed position. Extraction vessel which contains the sample was