# Supercritical Carbon Dioxide Extraction of Selected Herbal Leaves: An Overview

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# Supercritical Carbon Dioxide Extraction of Selected Herbal Leaves: An Overview

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**Abstract.** Supercritical fluid extraction of carbon dioxide (SC-$CO_2$) is one of new alternative extraction method that has been widely used to isolate bioactive components from variety of plant materials. The method was proved to be clean and safe, compatible for the extraction of edible products such as spices, food additives, medicines and nutritional supplement products compared to traditional extraction techniques such as solvent extraction, hydro distillation and steam distillation. The SC-$CO_2$ extraction was known as highly influenced by its process parameter such as temperature and pressure for obtaining maximum yield. Therefore, a clear review on the optimum range of temperature and pressure for herbal leaves extraction using SC-$CO_2$ is necessary for future reference. The aim of this work is to analyze the effect of temperature and pressure of SC-$CO_2$ process without modifier on extraction yield of some selected herbal leaves i.e clubmoss, drumstick leaves, kratom leaves, mallee and myrtle leaves. The values of investigated parameters were; pressure from 8.9 to 50 MPa and temperature from 35 to 80°C. The results showed that the highest extraction yields were obtained when the pressure and temperature were above 30 MPa and 40°C. The interaction between pressure and temperature for SC-$CO_2$ extraction of plant leaves are crucial since the values cannot be very high or very low in order to preserve the quality of the extracts.

## 1. Introduction

The application of a green extraction method of supercritical carbon dioxide (SC-$CO_2$) in isolating bioactive compounds from plant materials has been vigorously studied. Carbon dioxide ($CO_2$) in its supercritical condition (i.e. above its critical pressure and temperature of 7.38 MPa and 31.1°C), is an ideal solvent for extracting functional foods, supplement and medicines since it is nontoxic, noncorrosive and chemically inert which is safer to consumer and environment. Traditionally, many herbs from plant leaves have been used as alternative treatments by extracting oil from leaves using classical extraction methods such as solvent extraction, steam distillation and hydro distillation. However, these methods possess several weaknesses such as long exposed to the hazardous chemicals in solvent extraction (i.e. methanol, ethanol, dichloromethane, hexane, ethyl acetate, acetone etc.), high operating temperature used in steam and hydro distillation process that led to the losses of many important volatile compounds, long extraction period which consumes lots of energy and cost, and impurities to the final product that can cause many health problems [1][2]. Even though these methods could extract the medicinal compounds from the plant, but the probability of the organic solvent presence in the product and the losses of volatile compounds are high [3].

Therefore, a clean extraction method of SC-$CO_2$ that is free from any chemical solvent and only deals with $CO_2$ of 99.99% purity (without modifier) has been applied for the leaves extraction process. With a mild operating temperature used in SC-$CO_2$ system, it will avoid the losses of important compounds during the extraction process and has a potency to recover lots of volatile components from plant leaves. In general, the content of oil in the leaves consists of a complex mixture of compounds including polar and non-polar compounds, whereas $CO_2$ is well-known to be a nonpolar molecule. However, with the special properties owned by SC-$CO_2$ fluid i.e. the solvating power, has made this method proficient in extracting some of polar compounds or moderately polar compounds [4]. As published by Pronyk and Mazza [5], the ability of SC-$CO_2$ in extraction process is highly influenced by its temperature and pressure in which a slight change of temperature and/or pressure was greatly affect the output produced. Moreover, $CO_2$ at its supercritical state has the ability to penetrate into the solid matrix of the sample and dissolve the desired compound on account of its dual gaseous and liquid-like properties [6]. The increase in pressure of SC-$CO_2$ system could increase the fluid density which will enhance the solvating power. More oil or solutes will dissolve into the supercritical fluid promoting higher extraction yield. As for the temperature, the effect on the yield may be increase or decrease since it depense on the competing effect between fluid density and solutes vapour pressure [7]. When the tempertaure is higher, the density of supercritical fluid will decrease reducing the solvent strength to dissolve compounds. However, the increment of tempereture also increase the volatility of compounds which enhancing the oil solubility into the fluid. Therefore, in this study, the effect of extraction parameters i.e. pressure and temperature towards extraction yield were reviewed on the several selected herbal leaves. The knowledge on the parameters in SC-$CO_2$ process is significant in order to obtain highest oil yield as well as in designing the optimum process including equipment sizing within its ideal operating conditions. This article will become a guidance to choose the best range of parameters that is suitable for the extraction of herbal leaves under SC-$CO_2$ condition.

## 2. Supercritical carbon dioxide extraction on selected herbal leaves

Clubmoss (*Lycopodium clavatum L*) is a low-growing green plant consist of high medicinal compounds and has been used as alternative therapy and homeopathic medicines by traditional practitioners. Recent studies have showed that the extract from clubmoss could treat cervical cancer disease [8]. Therefore, da Silva [9] applied supercritical carbon dioxide (SC-$CO_2$) method to extract oil from clubmoss plant under the following condition; pressure range from 20 to 30 MPa, temperature range from 40 to 60°C and at constant flow rate of 800 g/h (table 1). The result showed that the oil yield increases with an increase in pressure from 20 to 30 MPa and a decrease in temperature from 60 to 40°C. The influence of increasing the pressure is expected since it will increase the density of supercritical fluid as well as increasing the fluid solubility which promoted to the higher extraction yield. However, the effect of temperature on the extraction yield can be varied (increase or decrease) depending on the complex balance between the vapour pressure of solute (i.e. the volatility of the extracted compounds) and the density of SC-$CO_2$ fluid [10]. In this case, the effect of density dominates the effect of solute vapour pressure since the highest yield of 1.36% was obtained at the highest pressure of 30 MPa (figure 1 (a)) and at the lowest temperature of 40°C (figure 1 (b)).

**Table 1.** Amount of extracted oil and process parameters in the supercritical carbon dioxide extraction of the selected leaves

| Leaves oil | Extraction yield (%) | Process parameter | Ref. |
|---|---|---|---|
| Clubmoss (*Lycopodium clavatum L.*) | 1.36 | P = 20 – 30 MPa; T = 40 - 60°C; F = 800 g/h; | [9] |
| Drumstick leaves (*Moringa oleifera*) | 6.34 | P = 30 – 50 MPa; T = 40 – 80°C; F = 2 L/min; | [11] |
| Kratom leaves (*Mitragyna speciose*) | 1.36 | P = 8.9 – 34.5 MPa; T = 40 – 80°C; F = 24 mL/min; | [12] |
| Mallee (*Eucalyptus loxophleba*) | 4.78 | P = 10 – 50 MPa; T = 40 – 80°C; F = 2 L/min; | [13] |
| Myrtle leaves (*Myrtus communis L.*) | 4.89 | P = 10 – 30 MPa; T = 35 – 50°C; F = 0.42 kg/h; | [14] |

P: pressure, T: temperature, F: flowrate

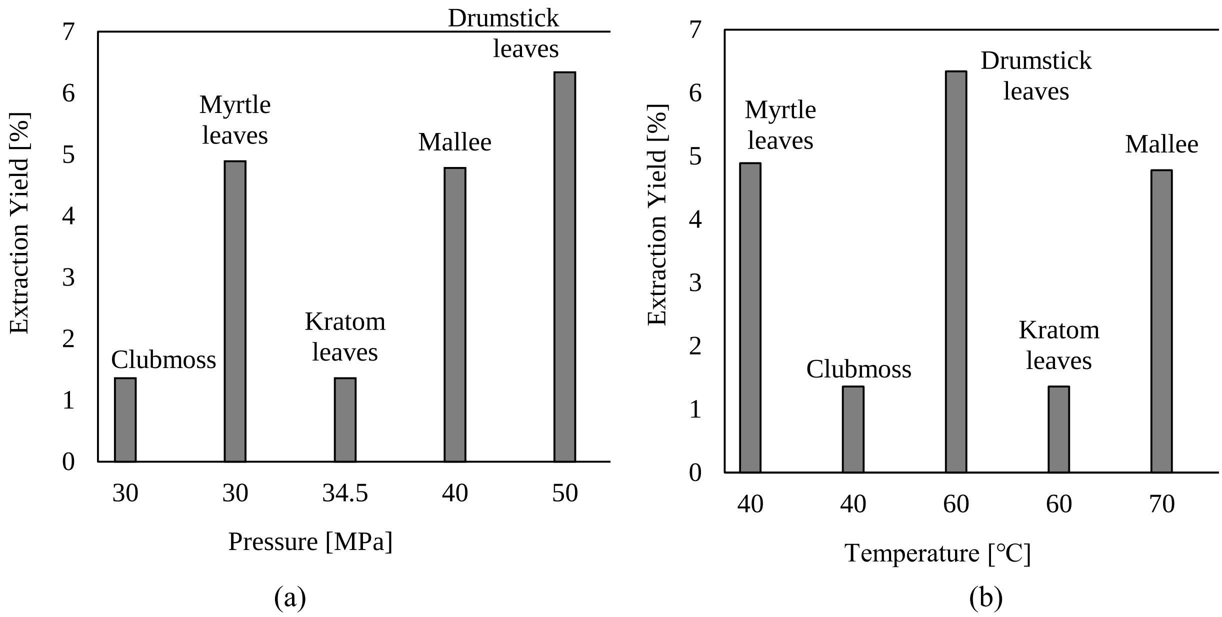


**Figure 1.** Pressure (a) and temperature (b) of the highest extraction yield in supercritical carbon dioxide extraction of the selected plant leaves.

Drumstick leaves (*Moringa oleifera*) contain both nutritional and medicinal values including vitamins, minerals and amino acids [15]. The plant also was consumed in food preparation such as curry. Zhao and Zhang conducted the extraction of oil from the drumstick leaves by using both traditional solvent extraction and SC-$CO_2$ methods [11]. The effect of different pressure (30 – 50 MPa) and, temperature (40 – 80°C) on the extraction yield were examined. As expected, when the pressure increases in isothermal condition, the extracted oil increases significantly due to the enhancement of the fluid density to solubilize more solute from the solid sample. For temperature effect, the increment from 40 to 60°C resulting in an increase in the extraction oil yield. When the temperature is high, the solute vapour pressure increased make it easier to be extracted out from solid matrix. Nevertheless, the further increment of temperature from 60 to 80°C has led to a slight decrease in the yield. This can be explained as, at that moment, the density of fluid started to reduce and thus lowered the yield. As a conclusion of this research, the maximum extraction yield can be obtained by manipulating the process parameters of temperature and pressure since the highest oil yield was achieved at 50 MPa and 60°C (figure 1). Kratom leaves (*Mitragyna speciose*) locally known as 'ketum' is a controversial plant which contains various medicinal values. The plant has been applied as traditional practice in folk medicine for the treatment of muscle pain, fever, wound, diarrhea and cough [16]. The extraction of oil from kratom leaves by using SC-$CO_2$ method has been conducted by Tohar et al. [12] at 8.9 to 34.5 MPa of pressure and 40 to 80°C of temperature with a constant flow rate of 24 mL/min (table 1).

In general, the extraction yield improved with the increment of pressure from 8.9 to 34.5 MPa. Conversely for the temperature effect, the yield started to decrease when the temperature was above 60°C. This phenomenon described that the further increment of temperature more than 60°C from Kratom leaves extraction will directed to a reduction in solvent density and thereby resulting to the lower yield. This is similar to the study conducted by Zhao and Zhang [11] discussed previously on the temperature effect of the extraction oil yield. In this study, the highest yield of 1.36% was obtained at the highest pressure of 34.5 MPa and at 60°C of temperature (figure 1 (a) and (b)). Zhao and Zhang [13] investigated the relationship between temperature and pressure on SC-$CO_2$ extraction of *Eucalyptus* leaves since the extract from the leaves contains numerous medicinal benefits such as anti-inflammatory, anti-allergenic, anti-asthmatic, antiseptic, aquaculture antiviral, anti-bacterial and anti-malarial. The laboratory experiments were carried out at a range of pressure from 10 to 50 MPa and temperature from 40 to 80°C (table 1). Based on this research, the extraction yield under a clean SC-$CO_2$ method also trailed the same trend for the increment in pressure. A rise in pressure from 10 to 40 MPa has risen the solvating power of the fluid and thus giving rise to more extraction yield. It is also stated that the increase in yield improved as the temperature increase from 40 to 70°C. The yield started to decrease when the pressure is further increases to 50 MPa and when the temperature is above 70°C. Such condition of yield reduction is due to the decreasing of fluid diffusion coefficient at a very high pressure. Thus, a very high pressure and temperature is not always recommended for SC-$CO_2$ extraction. The optimum interaction between pressure and temperature on *Eucalyptus* leaves was achieved at 40 MPa and 70°C with the highest oil yield of 4.78% and can be seen in table 1.

Zermane et al. [14] presented the result on the extraction of essential oil from Myrtle (*Myrtus communis*) leaves by using SC-$CO_2$ method. The extraction was carried out by varying the pressure from 10 to 30 MPa and temperature from 35 to 50°C. The optimum condition was achieved at 30 MPa and 40°C. Similar to the previous reviewed an increase in pressure leads to an increase in the oil recovery. Pressure gives a big impact to the yield since a slight change of pressure will change the solvent strength of the fluid which improving more extraction yield. As for the temperature effect, the oil yield shows a different trend. At initial stage, increasing temperature from 35 to 40°C increased the yield since raising temperature increases the solute vapour pressure, resulting in the enhancement of SC-$CO_2$ solubility. Then, a further increase in temperature up to 50°C decreased the yield. This is due to the reduction of the fluid density of $CO_2$ which reduce the solvation power. The overall effect of temperature towards extraction yield in SC-$CO_2$ extraction depends on whether solute vapour pressure or solvent density dominates at the particular condition.

## 3. Analysis of temperature and pressure on extraction yield

According to the survey of all selected plant leaves above (i.e. clubmoss, drumstick leaves, kratom leaves, mallee and myrtle leaves) that were undergone the SC-$CO_2$ process, the effect of the extraction pressure towards oil yield showed similar trend line. It can be explained that, as the pressure of the system increase at constant temperature, the density of supercritical fluid also increase. Density plays an important role in the extraction process since it triggers the solubility of fluid make it able to penetrate inside the solid sample to extract out the important oil at the solid surface. As can be seen in figure 1(a), most of the plant leaves achieved the maximum oil yield at pressure above 30 MPa.

Many authors agreed that the influence of temperature on the extracted yield is defined in different manners. In reviews on the extraction of the selected plant leaves above, the results of temperature effect are different. The extraction yield can be increased or decreased. Near the critical pressure, the density of fluid is very sensitive to temperature. At constant pressure, an increase in temperature will lead to a decrease in fluid density and reduces the solvent power. However, the increase in temperature will also speed up the mass transfer and enhance the yield. This is due to the increase in vapour pressure of the extractable compounds [1]. Therefore, it is hard to predict the effect of temperature in SC-$CO_2$ extraction.

There are other factors that can affect the extraction yield such as fluid flow rate, extraction time, sample particle size, moisture content and addition solvent as modifier. However, this paper only focus on the effect of pressure and temperature since these two factors have the biggest impact on the extraction yield based on supercritical fluid properties such as gas-like diffusivity and liquid-like density. Apart from that, most of the leaves oils consist of a complex mixture of compounds including polar and non-polar compounds. The interactive combination of pressure and temperature in SC-$CO_2$ system can increase the mass transfer of fluid to penetrate inside the solid sample and thus extracting more oil. The perfect combination of temperature and pressure also promotes to the increasing solute vapour pressure which is crucial for the isolation of polar substance from plant sample.

## 4. Conclusion

In this study, the influence of temperature and pressure on the extraction of clubmoss, drumstick leaves, kratom leaves, mallee and myrtle leaves under SC-$CO_2$ extraction were presented. Based on the comparison result on the effect of temperature and pressure used, the extraction yield from 1.36 to 6.34% was achieved within the range of pressure from 30 to 50 MPa and temperature from 40 to 70°C. The review of the range of parameter is necessary for the optimum design of supercritical extraction process and could be used as future reference in supercritical fluid field.

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