ORIGINAL ARTICLE

Supercritical carbon dioxide extraction of oil from *Chromolaena odorata* leaves

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Abstract:

Relatively all previous researches on *Chromolaena odorata* were carried out by using traditional method such as hydrodistillation and soxhlet extraction methods which have their own disadvantages such as long extraction time and non-solvent free oil extract. Thus, a clean extraction method of Supercritical Fluid Extraction (SFE) using Supercritical Carbon Dioxide (SC-CO_2) was proposed for the extraction of *Chromolaena odorata* leaves to produce oil with bioactive components. The SC-CO_2 extraction of *Chromolaena odorata* was studied by using measurement of solubility of the oil. The extraction was conducted at constant CO_2 flowrate of 24mL/min extraction time of 60 minutes within a range of temperature (40°C, 45°C and 50°C) and range of pressures (3000, 3500, 4000, 4500 and 5000 psi). The highest solubility of 7.619 mg oil/ g CO_2 in SC-CO_2 was obtained at pressure 4000psi and temperature 50°C. Based on the this study, the solubility increased with increases pressures because the density of CO_2 that leads to the improvement of solvating power of CO_2. The solubility also increased with increasing temperature as the vapour pressure of solute in the sample increased the energy for the sample to travel into the SC-CO_2.

Keywords: *Chloromolaena odorata*, SFE, Solubility

1. INTRODUCTION

Chromolaena odorata is a type of flowering plant that falls under the family of Asteraceae which is also have common names "Pokok Kapal Terbang", Siam Weed, "Independence Weed", Bitter Bush, Christmas Weed or Jack in the Bush [1, 2]. 80% of world population used *Chromolaena odorata* as a traditional medicine for treating coughs, colds, skin diseases, and wound healing as well as an antiseptic agent [3–5]. Besides the traditional used, it is also used pharmaceutically as an antispasmodic, antiprotozoal, anti-trypanosomal, anti-bacterial or antimicrobial, anti-fungal, antihypertensive, anti-inflammatory, anti-gonorrhoeal, astringent, antipyretic, analgesic, diuretic and hepatortropic agent [1, 4].

Many studies recognized the high amount of natural antioxidant content in *Chromolaena odorata* as a valuable component to replace synthetic antioxidant which could be used in anticancer activity for cancer treatment. Besides that, antioxidant components such as α-pinene, β-pinene stored in *Chromolaena odorata* leaves are beneficial to pharmaceutical industries. As an example, Chakrabotry *et. Al* [2] revealed that *Chromolaena odorata* has a lot of pharmacological activities such as anti-oxidant, anti-microbacterial, antibiotic, blood coagulation and wound healing activity [2]. Moreover attention are focusing on natural antioxidants as the synthetics counterpart safety issues are doubtful.

Traditional extraction method such as hydrodistillation and soxhlet extraction are normally used in the extract an oil from *Chromolaena odorata* [4, 6,7-9]. However, these long-established methods of oil extraction have their own disadvantages. As stated by Illés et al [10], hydrodistillation extraction method gives chemical alterations to the plant essential oil extracted. Besides that, the compounds that are sensitive to heat can merely be destroyed which means that the oil quality is low. In soxhlet extraction, the disadvantages is that agitation is not possible to occur, the possibility of thermal decomposition must not be ignored due to long extraction time, the valuable volatiles compounds could lost during solvent evaporation and the extracted procedure is not solvent free [10, 11]. Other than that, these two extraction methods used hexane as a solvent which might become residual and have some level of environmental contamination during the oil extract [12].

Therefore, in this study a clean extraction method which is Supercritical Fluid Extraction with carbon dioxide, CO_2 (SFE-CO_2) as a solvent and ethanol as co-solvent are introduced for the extraction of *Chromolaena odorata* oil. Recent developments in extraction technology such as Supercritical Fluid Extraction (SFE) allowed the extraction of bioactive components of herb to be produced in the form of essential oil. Supercritical carbon dioxide (SC-CO_2) as a solvent offers a lot of benefits compared to traditional extraction method such as being nontoxic, non-flammable, environmental friendly method, easy to separate the solvent, mild operating conditions, short extraction time, and offers high quality of final product [12–15]. Handa [16] states that SFE is an alternative extraction method that reduced the use of organic solvents and increased the product of extracted oil. Co-solvent such as ethanol is added during SFE process to increase the solvent power of supercritical CO_2 towards polar molecules [17]. When the solvent power increase, the extraction oil yield increases [18]. The co-solvent is collected in the collection vessel together with the extracted oil. Therefore, rotary evaporator is used to separate the co-solvent from the desired product. According to Pourmortazvi and Hajimirsadeghi [19], more than 90% of SFE is performed by using CO_2 due to its low critical temperature (31.1°C) and low critical pressure (7.38MPa). It is where CO_2 in liquid and gas phase disappear and form one single supercritical phase.

As for solubility, it is an important property prior to optimizing the extraction yield [20]. Solubility is dependent on the content of components from different chemical categories existing in the plant. De melo et al [21] stated that in order to calculate the solubility, it is either to consider the oil as a single body or the solute as the unique compound to be dissolved by SC-CO_2. According to Sapkale et al., when the pressure is above critical pressure, solubility will increase with temperature [12]. This is because, the solubility is highly manipulated by pressure and temperature of the supercritical solvent [22]. The increase in pressure will increase the density of CO_2 which leads to the improvement of its solvating power. The increase in pressure diffuse the SC-CO_2 rapidly into the solid sample which contribute to the rise in the solubility. Whereas for temperature, it affects the solubility in terms of vapour pressure of solute in the sample in which the energy for the sample to travel into the SC-CO_2 is increased and thus increasing in the solubility.

2. MATERIALS AND METHODS

2.1 Selection of operating conditions

The values of extraction parameters such as temperature, pressure, extraction time and flowrate of SC-CO_2 to extract oil from *Chromoalaena odorata* leaves were important as it determined efficiency of the extraction yield. The study conducted by Yamini et al. on oil extraction of *Salvia mirzayanii* was done within the range of temperature 35-70°C and pressure 1464-5149 psi for a duration of 15-35 minutes [23]. Hamdan et al. have conducted extraction of cardamom oil by SC-CO_2 at the range of temperature 35-50°C and pressure in between 1450-4351 psi [24]. Recently, Zeković et al. [25] have done an investigation on the optimization of coriander seed using SC-CO_2 at temperatures of 40, 55 and 70°C and pressure of 1450, 2176 and 2900 psi. Hence, it was determined that the extraction temperature and pressure used for the oil extraction of *Chromoalaena odorata* were in the range of 35-70°C and 1450-5200 psi. Thus in this study, the temperature used for extraction of *Chromoalaena odorata* were 40, 45 and 50°C and pressure used were 3000, 3500, 4000, 4500 and 5000 psi.

2.2 Material preparation

The leaves of *Chromoalaena odorata* used were obtained from locally near UiTM Pulau Pinang. The leaves were cleaned meticulously with water to eliminate any impurities. Then, the leaves were oven-dried to less than 10% moisture content and then ground using Vertical Grinder and then sieved using sieve shaker Model Retsch AS 200 to obtain *Chromoalaena odorata* leaves in powdered formed with the approximate size of 0.25mm.

2.3 Supercritical carbon dioxide (SC-CO_2) extraction

SC-CO_2 extraction of *Chromolaena odorata* leaves was carried out using Thar SFC from Thar Process (USA) available in Faculty of Chemical Engineering, UiTM Pulau Pinang. The SC-CO_2 extraction are carried out at operating temperature of 40, 45 and 50°C and pressure of 3000, 3500, 4000, 4500 and 5000 psi with 60 minutes extraction time, constant CO_2 flowrate of 24 mL/min and ethanol flowrate 2.4mL/min. The maximum mass of sample that could be loaded in the extraction vessel is 10g. The Thar SFC equipment was designed specifically to operate at maximum temperature of 90°C and maximum pressure of 5800psi. The oil extracts were collected at extraction time 10, 20, 30, 40, 50 and 60 minutes for every extraction parameters.

2.4 Rotary evaporator

The extracted *Chromolaena odorata* oil from SC-CO_2 was evaporated using Buchi Rotavapor R-210. The heating bath filled with distilled water was set at 76°C which is less 2°C from the boiling point of ethanol of 78.5°C [26]. The vacuum and rotation were set to 320mbar and 30rpm respectively. The evaporating flask was lowered until the extracted oil was half submerged in the heating bath and the evaporation was stopped when ethanol flow into the receiving flask started to slow down.

2.5 Determination of solubility

The solubility was determine by calculating the slope based on the mass of extracted oil per mass of CO_2 consumed [27-28]. The mass of CO_2 consumed can be determined by using Eq. 1:

$$\text{Mass } CO_2 \text{ used} = t(\text{min}) \times Q\left(\frac{\text{mL}}{\text{min}}\right) \times \rho_{CO_2}\left(\frac{\text{g}}{\text{mL}}\right) \quad (1)$$

where t is the extraction time (min), Q is CO_2 flowrate (mL/min) and ρ_{co2} is density of CO_2 (g/mL). The mass of extracted oil was obtained from oil collected at extraction time 10, 20, 30, 40, 50 and 60 minutes and the density of CO_2 at certain pressure (3000, 3500, 4000, 4500 and

5000psi) and temperature (40, 45 and 50°C) were referred from Gupta and Shim [29].

3. RESULTS AND DISCUSSION

In this study, the effect of different pressures 3000, 3500, 4000, 4500 and 5000psi and different temperature of 40, 45 and 50°C was investigate. The oil extract was collected at extraction time of 10, 20, 30, 40, 50 and 60 minutes. As stated by Sapkale et. al [12], the extraction time using SFE is fast which is mostly less than 60 minutes. The solubility determination graphs were provided in Figure 1 (a) to (e). From Figure 1 (a) to (e), the solubility were determined by the slope of each curve. The effects of pressure and temperature on the solubility were shown in Figure 2 and Figure 3.

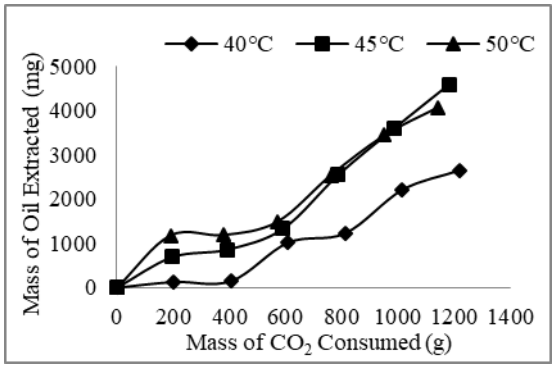

Figure 1(a): The solubility of oil determination curve for 3000 psi

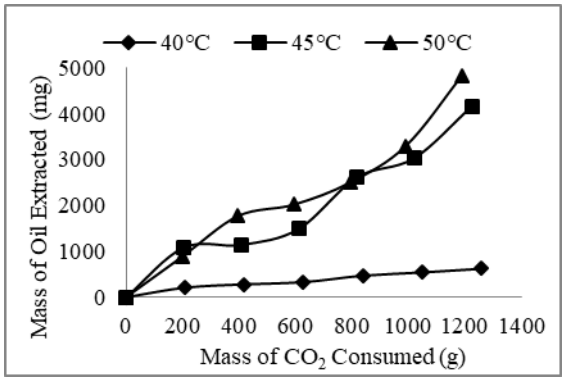

Figure 1(b): The solubility of oil determination curve for 3500 psi

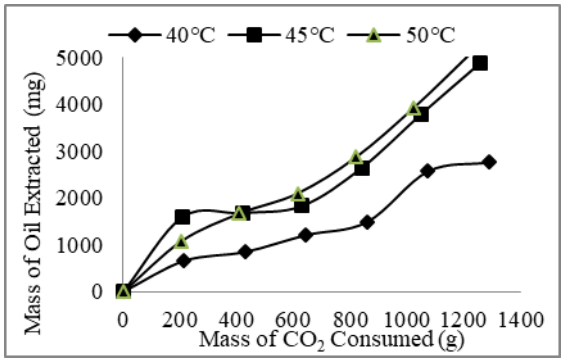

Figure 1(b): The solubility of oil determination curve for 4000 psi

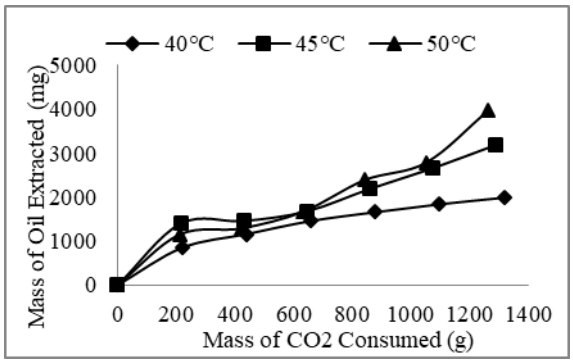

Figure 1(d): The solubility determination curve for 4500 psi

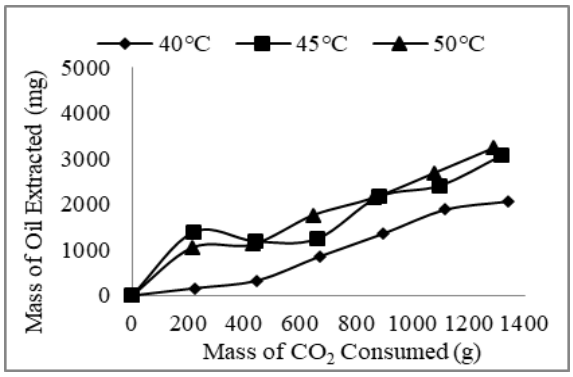

Figure 1(e): The solubility of oil determination curve for 5000 psi

From the Figure. 2, it could be observed that the solubility increased with pressure due to an improvement in the solvent power when the solvent density increased in. On the other hand, the solubility drops at pressure 4500 and 5000psi due to the decreased in mass of oil extracted at that point as shown in Table 2. The highest solubility was 7.619 mg oil/g CO_2 at temperature 50°C and pressure 4000psi.

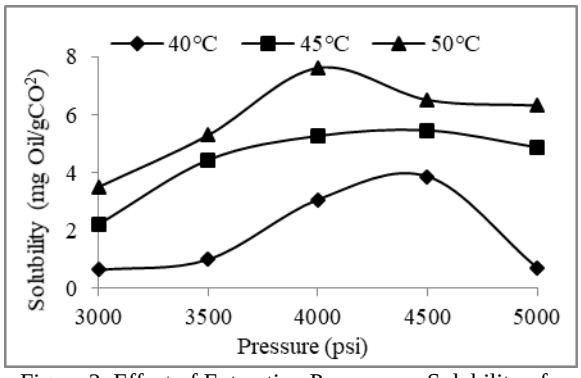

Figure 2: Effect of Extraction Pressure on Solubility of *Chromolaena Odorata* Oil at Constant Temperature (40°C, 45°C and 50°C)

Figure 3 illustrated the effect of extraction temperature on solubility of *Chromolaena odorata* oil at constant pressure (3000, 3500, 4000, 4500 and 5000psi). From the figure, it can be seen that the solubility increased from temperature 40°C to 50°C. It showed that solubility increased with increased in temperature due to increase in the energy of the solid sample to travel into the SC-CO_2 which leads to the increased in solubility. It indicated that the highest solubility at all pressure were 3.871 mg oil/g CO_2, 5.464 mg oil/g CO_2 and 7.619 mg oil/g CO_2 at temperature 40°C, 45°C and 50°C

respectively. Therefore, the density SC-CO_2 increased as the pressure increased which give rise to the solvent power of CO_2 [14, 19].

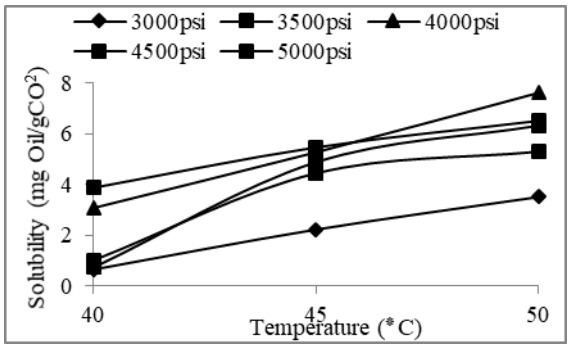

Figure. 3: Effect of Extraction Temperature on Solubility of *Chromolaena odorata* Oil at Constant Pressure (3000, 3500, 4000, 4500 and 5000psi)

4 CONCLUSION

The aim of this research was to determine the highest solubility of *Chromolaena odorata* leaves oil extracted using Supercritical Carbon Dioxide (SC-CO_2) at constant CO_2 flowrate of 24 mL/min and 60 minutes extraction time. The effect of the temperatures of 40, 45 and 50°C, and pressure at the range of 3000, 3500, 4000, 4500 and 5000psi were investigated. Using the parameters to extract *Chromolaena odorata* leaves for oil, the highest solubility was 7.619 mg oil/g CO_2 which was obtained at temperature 50°C and pressure of 4000psi. It was observed that the solubility increased as the pressure until 4000psi as increased. The solubility of extracted oil also increased rapidly with temperature. However, the oil yield and solubility dropped at pressure 4500 and 5000psi. This may be due to the oil achieving the limit to be extracted since at the early stage of extraction the oil yield increased rapidly. At high temperature and pressure, the yield and solubility of the oil extracted increased as it is affected by the balance between solvent (SC-CO_2) density and vapour pressure. Therefore, the density SC-CO_2 increased as the pressure increased which give rise to the solvent power of CO_2.

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