

Process Optimization of Supercritical CO₂ Extraction for Enhanced Yield of Sarawak Black Pepper (*Piper Nigrum* L.) Essential Oil

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ABSTRACT

Supercritical fluid extraction (SFE) using carbon dioxide has become a green and efficient alternative to traditional methods for extracting essential oils and bioactive compounds from plant materials. However, extraction performance heavily depends on operating conditions, requiring systematic optimization. In this study, a One Factor At a Time (OFAT) approach was used to optimize the supercritical CO₂ extraction of Sarawak black pepper (*Piper nigrum* L.) oil, aiming to maximize oil yield and retain key bioactive compounds. The effects of extraction pressure (200-325 bar), temperature (20-70 °C), CO₂ flow rate (2-10 g min⁻¹), extraction time (30-180 min), and ethanol co-solvent concentration (0-25% w/w) were examined individually while keeping other parameters constant. Oil yield was measured gravimetrically, and piperine content was analyzed using high-performance liquid chromatography. Results indicated that all operating parameters significantly affected extraction efficiency. The optimal conditions were found at 300 bar pressure, 50 °C temperature, 6 g min⁻¹ CO₂ flow rate, 90 min extraction time, and 10% (w/w) ethanol as a co-solvent. Under these conditions, a maximum oil yield of 4.07 ± 0.10% was obtained. The optimized extract had a piperine content of 4.88% (w/w) and physicochemical properties consistent with high-quality, unadulterated black pepper oil. Compared to Sarawak-specific SFE studies without co-solvents, adding ethanol significantly improved extraction efficiency. Overall, this study shows that OFAT-based optimization offers valuable parametric insights and provides a practical, scalable strategy for producing high-quality Sarawak black pepper oil using supercritical CO₂.

INTRODUCTION

Black pepper (*Piper nigrum* L.), often called the “King of Spices,” is one of the most commercially important spices worldwide because of its broad uses in food, pharmaceutical, cosmetic, and nutraceutical industries. The essential oil from black pepper is especially valued for its rich bioactive content, including piperine, monoterpenes, sesquiterpenes, and alkaloids, which show antimicrobial, antioxidant, anti-inflammatory, and therapeutic effects (Morsy and El-Salam, 2017; Kırkın and Güneş, 2018; Khan et al., 2021). These beneficial properties have greatly increased the demand for high-quality black pepper oil in natural food preservation, health supplements, and cosmetic products.

Sarawak, Malaysia, is internationally recognized for producing high-quality black pepper due to its favorable agro-climatic conditions and fertile soil, resulting in peppercorns with high piperine content and a unique flavor profile (Paracampo et al., 2022). Despite strong global demand, extracting essential oil from Sarawak black pepper remains challenging. Conventional methods like steam distillation and solvent extraction are commonly used, but these techniques often involve long processing times, low oil yields, thermal degradation of heat-sensitive compounds, and potential solvent residue contamination. These limitations affect oil quality and raise environmental and safety concerns (Tran et al., 2019; Dosoky et al., 2019).

Supercritical fluid extraction (SFE), especially using supercritical carbon dioxide (SC-CO₂), has become an efficient and eco-friendly alternative to traditional extraction methods. SFE provides high selectivity, improved

extraction efficiency, and operates at relatively low temperatures, which helps minimize thermal degradation and preserves volatile bioactive compounds (Dawid et al., 2012; Vinod et al., 2014). Additionally, CO₂ is non-toxic, non-flammable, and easy to recover, making the process more sustainable compared to conventional solvent-based techniques (Collings et al., 2018; Hakim et al., 2020). Several studies have shown that SFE yields black pepper oil with superior chemical quality and improved retention of bioactive compounds, boosting its functional and commercial value (Uwineza and Waśkiewicz, 2020).

Despite the recognized benefits of SFE, extraction performance is heavily affected by operating parameters such as pressure, temperature, solvent flow rate, and extraction time. Systematic optimization of these parameters is crucial to maximize oil yield and process efficiency (Oalere et al., 2021; Arief et al., 2023). While multivariate optimization techniques are often reported, there are still relatively few studies that offer a clear parametric understanding using a One Factor At a Time (OFAT) approach, especially for Sarawak black pepper oil. OFAT analysis is useful for identifying the individual effect of each operating parameter and defining practical operating ranges for industrial use (Feyzi et al., 2016; Kumar et al., 2021).

Therefore, this study aims to optimize the supercritical CO₂ extraction of Sarawak black pepper oil using a One Factor At a Time (OFAT) approach by systematically examining the effects of pressure, temperature, CO₂ flow rate, and extraction time on oil yield. The findings are expected to offer fundamental insights into process behavior, establish optimal operating conditions, and support the development of efficient, high-quality, and sustainable extraction strategies for industrial-scale black pepper oil production.

MATERIALS AND METHODS

Raw Material and Sample Preparation

Sarawak black pepper (*Piper nigrum* L.) berries were sourced from the Sarawak Pepper Marketing Board (Kuching, Sarawak, Malaysia) to ensure raw material standardisation. Fully matured berries (6 to 8 months) were selected based on uniform size, colour, and maturity, as maturity significantly influences oil yield and piperine content (Tiong and Rosazlina, 2011). The berries were washed to remove surface impurities and dried at 40–50 °C until the moisture content was reduced to ≤10%, minimising thermal degradation of volatile bioactive compounds (Kumar et al., 2021; Gupta and Sharma, 2019).

Dried samples were ground using an electric grinder and sieved to obtain a uniform particle size of approximately 1.0 mm, following established procedures to enhance mass transfer during supercritical extraction (Obek and Saptoru, 2023). The prepared powder was stored in airtight containers at ambient temperature prior to extraction.

Supercritical CO₂ Extraction (SFE) Procedure

Supercritical fluid extraction was carried out using a Speed SFE Zoran extractor (Applied Separations, Allentown, PA, USA), following the method adapted from Luca et al. (2023). The system was equipped with a 100 mL stainless steel extraction vessel. For each experimental run, 40 g of ground black pepper powder was loaded into the vessel. The extraction temperature was controlled between 40 and 60 °C using a thermostatically regulated heating mantle, while pressure was varied between 200 and 325 bar.

High purity carbon dioxide (CO₂) was used as the main extraction solvent. Additionally, ethanol was added as a co-solvent at 5% v/v relative to CO₂ to improve the solubility of polar compounds, especially piperine, following the procedure described by Luca et al. (2023). Ethanol was introduced into the extraction vessel before pressurization and system equilibration to ensure uniform contact with the sample matrix. The 5% v/v concentration was selected based on preliminary studies, which showed that higher amounts did not significantly increase yield and could complicate system operation.

Before extraction, the system was allowed to equilibrate for 10 minutes at the specified temperature and pressure. The process started by opening the outlet valve, and the CO₂ flow rate was controlled with a micrometre valve and monitored via a digital flow meter. Extracted fractions were collected in pre-cooled glass vials every 15 minutes and stored at –20 °C until analysis. The amount of oil extracted was measured gravimetrically to determine the extraction yield.

OFAT Experimental Design

The optimisation of the SFE process was performed using the One Factor At A Time (OFAT) approach, as recommended for establishing the individual influence of operating variables on extraction efficiency (Dutta and Bhattacharjee, 2016). Key parameters investigated included extraction pressure, temperature, CO₂ flow rate, and extraction time, while all other variables were maintained constant.

Although OFAT does not consider potential interaction effects between variables and might need more experimental runs than multivariate statistical designs, it offers a straightforward, transparent, and systematic approach for initial process optimization and understanding mechanisms (Montgomery, 2017). In this study, OFAT was intentionally chosen to evaluate the individual effects and relative importance of key extraction parameters, enhance fundamental process understanding, and identify appropriate operating ranges for future optimization and scale-up efforts.

The OFAT sequence was conducted as follows:

Parameter	Investigated Range	Constant Conditions
Pressure	200 to 325 bar	T = 40 °C, CO ₂ flow = 4 g min ⁻¹ g ⁻¹ , time = 120 min
Temperature	20 to 70 °C	Optimum pressure, CO ₂ flow = 4 g min ⁻¹ g ⁻¹ , time = 120 min
CO ₂ flow rate	2 to 10 g min ⁻¹ g ⁻¹	Optimum pressure and temperature, time = 120 min
Extraction time	30 to 180 min	Optimum pressure, temperature, and flow rate
Ethanol Concentration	0 to 25 % (w/w)	Optimum pressure, temperature, flow rate, extraction time

All experimental runs were performed in triplicate, and mean values with standard deviations were reported.

Yield Determination

The extraction yield was calculated as the ratio of the mass of extracted oil to the mass of the raw material, expressed as a percentage, following the standard approach used in SFE studies (Reverchon and De Marco, 2006; Sovová, 2012):

$$\text{Yield (\%)} = \frac{m_{oil}}{m_{sample}} \times 100 \quad (1)$$

where m_{oil} is the mass of the extracted oil (g), and m_{sample} is the mass of black pepper powder used in the extraction (g).

Chemical and Physicochemical Characterisation

The chemical composition of the extracted black pepper oil was analyzed using gas chromatography–mass spectrometry (GC–MS) following the methods reported by Dutta and Bhattacharjee (2016) and Luca et al. (2023). Piperine content in the extracted oil was quantified by HPLC (UV detection at 341 nm) using a calibration curve prepared with standard piperine ($\geq 98\%$ purity, Sigma-Aldrich). The extracted oil was dissolved in methanol, filtered, and injected in triplicate. The piperine content (% w/w) was calculated as the mass of piperine divided by the mass of the extracted oil, multiplied by 100. Method validation parameters, including linearity, limit of detection, and limit of quantification, were adopted from Setyaningsih et al. (2021) and Shrestha et al. (2020).

Physicochemical properties, including color and refractive index, were assessed to determine oil quality. Color was measured using a Hunter Lab Color Flex EZ colorimeter (Ravindran and Kallapurackal, 2012), viscosity was determined with a Brookfield rotational viscometer following ASTM D445 (Yadav and Singh, 2020), and refractive index was measured using a digital refractometer at 25 °C in accordance with ISO standards (Filippova et al., 2020).

Statistical Analysis

Statistical analysis of the experimental data was carried out using one-way analysis of variance (ANOVA) to assess the influence of process parameters on oil yield, with significance set at $p < 0.05$. All experiments were

performed in triplicate, and the results are reported as mean \pm standard deviation. Tukey’s post-hoc test was used to identify significant differences between factor levels, with $p < 0.05$ as the criterion. In the results table, means with the same letter are not significantly different, while different letters indicate statistically significant differences.

RESULTS AND DISCUSSION

Effect of Pressure on Extraction Yield

Pressure is one of the most influential parameters in supercritical fluid extraction, as it directly affects the density and solvating power of supercritical CO₂. As pressure increases, CO₂ density rises, enhancing solvent penetration into the plant matrix and improving the solubility of lipophilic compounds such as piperine, β -caryophyllene, and limonene (Silva et al., 2016; Liu et al., 2021).

In this study, increasing pressure from 200 to 300 bar resulted in a steady rise in oil yield, reaching a maximum of 2.08 ± 0.08 % at 300 bar. A further increase to 325 bar caused a slight reduction in yield (1.87 ± 0.075 %), indicating a saturation effect and reduced mass-transfer gradients at higher solvent densities. Similar behaviour has been reported for black pepper oil and other spice matrices, where excessive pressure promotes co-extraction of heavier compounds and limits effective solute diffusion (Campone et al., 2018; Khaw et al., 2017).

These findings agree with Lin et al. (2013) and Salea et al. (2014), who identified 250 to 300 bar as an optimal pressure range for balancing extraction efficiency and oil quality. The results confirm that 300 bar represents an optimal pressure for Sarawak black pepper oil when evaluated using the OFAT approach.

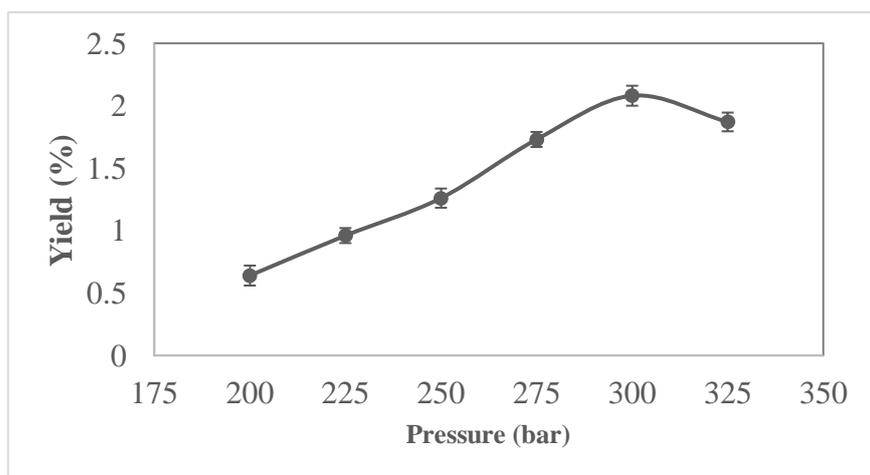


Figure 3.1. Effect of extraction pressure on the yield of Sarawak black pepper oil obtained by supercritical CO₂ extraction.

Effect of Temperature on Extraction Yield

Temperature influences extraction performance through competing effects on CO₂ density and solute vapor pressure. Moderate temperature increases enhance molecular diffusivity and solute volatility, while excessive temperatures reduce solvent density and may degrade thermolabile compounds (Kumoro et al., 2010; Li et al., 2022).

As shown in Figure 3. 3.2, the oil yield steadily increased from 1. 33% at 20 ° C to $2. 76 \pm 0. 11\%$ at 40 ° C, reaching a maximum of $2. 98 \pm 0. 097\%$ at 50 ° C, before sharply declining to $1. 87 \pm 0. 18\%$ at 70 ° C. These results indicate that 50 ° C is the optimal extraction temperature under the studied conditions. Furthermore, the 40–50 ° C range can be considered an optimal operational window, where high yield and extraction efficiency are achieved simultaneously.

An operational window is a range of conditions that ensures stable, reproducible, and near-maximum extraction, rather than relying on a single optimum that is difficult to maintain (Reverchon & De Marco, 2006;

Martínez, 2007). Operating at 40–50 ° C provides sufficient CO₂ solvating power and better mass transfer, while avoiding excessive thermal stress that could damage thermolabile compounds. This improves process robustness, product quality, and economic viability (Herrero et al., 2013). Within this range, moderate CO₂ density reduction and increased solute vapor pressure promote optimal solubility and diffusion, but beyond 50 ° C, decreased solvent density and thermal degradation of piperine and terpenes reduce efficiency and yield (Sovová, 2012; Li et al., 2022). Similar temperature-dependent behavior is seen in spice and essential oil extraction, where moderate temperatures maximize yield and preserve compound integrity (Brunner, 2010).

This trend aligns with previous studies that identified optimal extraction temperatures for black pepper oil within the 40–50 ° C range (Yang et al., 2015; He et al., 2024). The consistency between current results and existing literature further confirms that maintaining a controlled operational window, rather than depending on a single optimum point, offers a more practical and a reliable and scalable approach for efficient supercritical fluid extraction of black pepper oil.

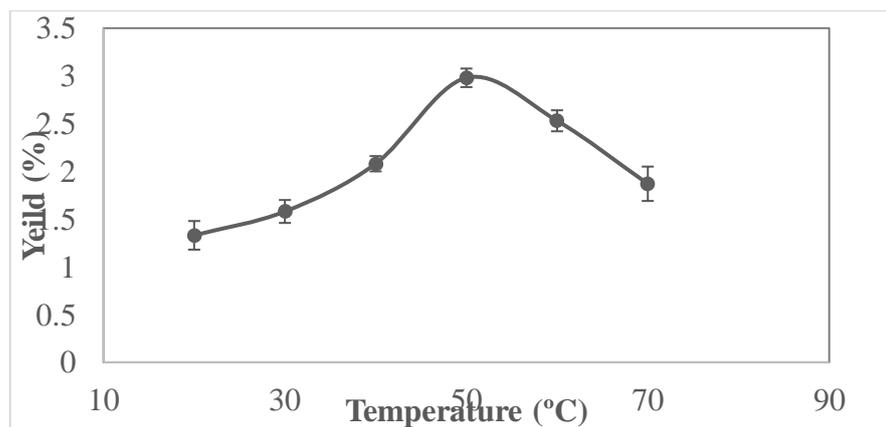


Figure 3.2. Effect of extraction temperature on the yield of Sarawak black pepper oil during supercritical CO₂ extraction.

Effect of CO₂ Flow Rate

CO₂ flow rate affects solvent availability and contact time with the matrix. Increasing flow rate initially improves mass transfer, but too high flow rates decrease residence time and disrupt extraction equilibrium (Han and Kang, 2015; Ruslan et al., 2020).

In the present study, yield increased with flow rate from 2 to 6 g min⁻¹, reaching a maximum of 3.43 ± 0.14 %. Further increases to 8 and 10 g min⁻¹ resulted in lower yields (2.65 ± 0.15 % and 2.36 ± 0.084 %, respectively). This bell-shaped profile is typical of SFE systems and confirms that 6 g min⁻¹ offers the best solvent utilization.

Similar behavior has been observed in black pepper and spice extraction studies, where an optimal flow rate maximizes mass transfer without reducing solvent-solute interaction time (Salea et al., 2014; Maran et al., 2014).

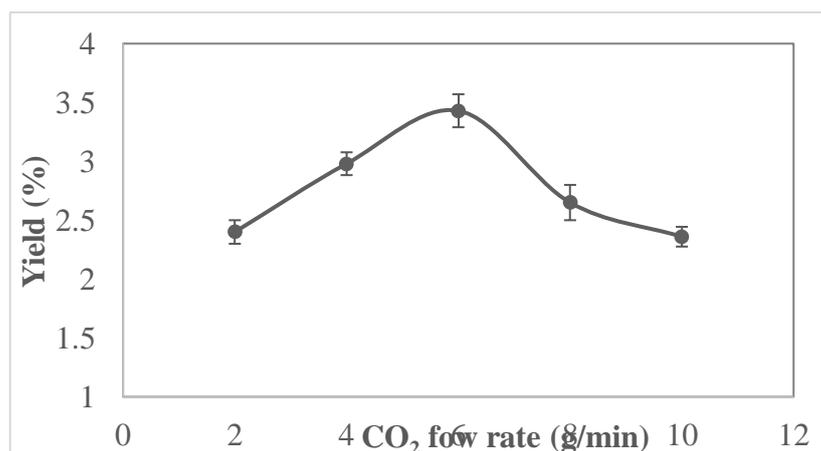


Figure 3.3. Influence of CO₂ flow rate on the extraction yield of Sarawak black pepper oil using supercritical CO₂.

Effect of Extraction Time

Extraction time affects oil recovery kinetics and energy efficiency. The extraction curve usually features a quick constant extraction rate phase, followed by slower diffusion-controlled stages (Sovová, 2012).

The extraction yield increased rapidly during the first 90 minutes, reaching $3.43 \pm 0.15\%$, and remained fairly steady up to 150 minutes before slightly decreasing at 180 minutes. This shows that most of the extractable oil was obtained within 90–120 minutes, as extended extraction afterward yielded little additional oil.

These findings are consistent with studies by Jamel et al. (2012) and Elmaaty et al. (2023), which reported limited benefits from extended extraction due to diffusion constraints and higher energy use.

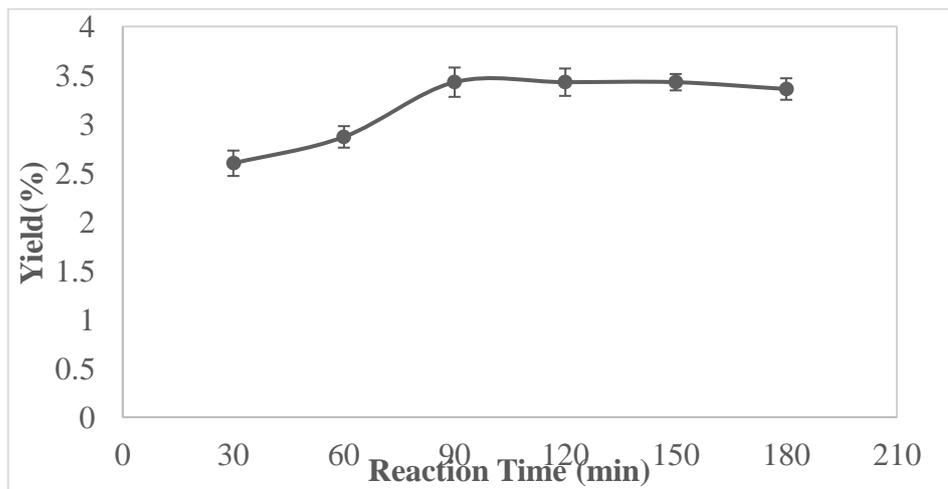


Figure 3.4. Effect of extraction time on the yield of Sarawak black pepper oil obtained by supercritical fluid extraction.

Effect of Ethanol Co-solvent Concentration

Ethanol modifies the polarity of supercritical CO₂, enhancing the solubility of moderately polar compounds such as piperine. In this study, the yield increased with ethanol concentration up to 10 % (w/w), reaching a maximum of $4.07 \pm 0.10\%$, before declining at higher concentrations.

Excessive ethanol likely reduced CO₂ density and promoted the co-extraction of undesired compounds, thereby lowering extraction efficiency. Similar optimal ethanol concentrations (5–10 %) have been reported for black pepper oil extraction (Lin et al., 2013; Nagavekar and Singhal, 2017; Salea et al., 2014).

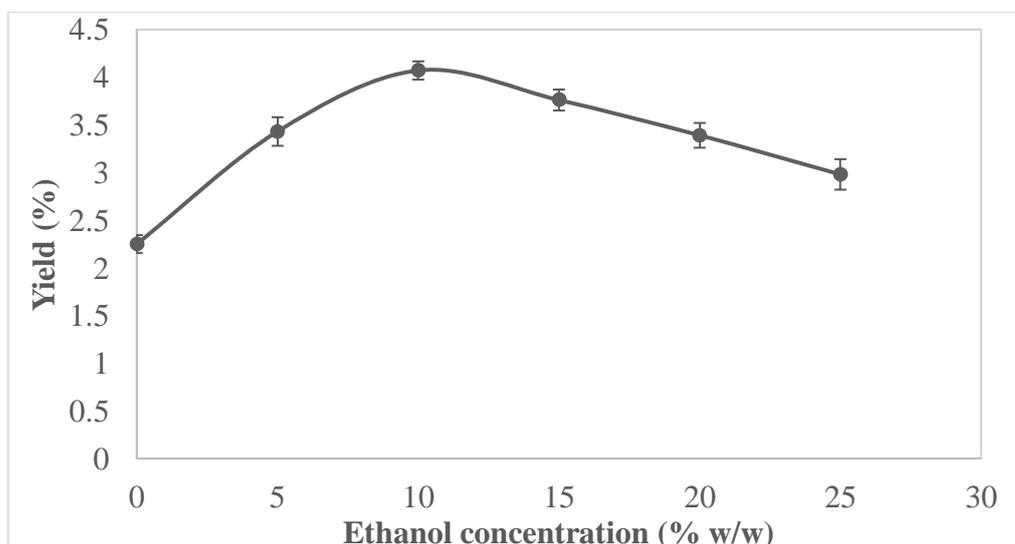


Figure 3.5. Effect of ethanol co-solvent concentration on the yield of Sarawak black pepper oil during supercritical CO₂ extraction.

Optimised Extraction Conditions and Comparison with Literature

Based on OFAT optimisation, the optimal SFE conditions for Sarawak black pepper oil were identified as:

- Pressure: 300 bar
- Temperature: 50 °C
- CO₂ flow rate: 6 g min⁻¹
- Extraction time: 90 min
- Ethanol co-solvent: 10 % (w/w)

The results show that all the factors examined, including pressure, temperature, CO₂ flow rate, extraction time, and ethanol co-solvent concentration, significantly affected extraction performance ($p < 0.05$). The optimal conditions were determined to be 300 bar, 50 °C, 6 g/min CO₂ flow rate, 90 minutes extraction time, and 10% ethanol, yielding $4.07 \pm 0.10\%$, a result significantly higher than most other parameter levels (Tukey's post-hoc test, $p < 0.05$). These findings confirm the effectiveness of the OFAT-based optimization and highlight ethanol's role as a co-solvent in improving mass transfer and bioactive compound recovery.

This maximum yield significantly exceeds those reported in earlier Sarawak-specific studies that did not use co-solvent (Kumoro et al., 2010; Lin et al., 2013) and is comparable to yields from optimized SFE studies with ethanol (Nagavekar & Singhal, 2017), confirming both practical and literature-based relevance of the optimized conditions.

Piperine Content and Oil Quality

The piperine content of the extracted oil was 4.88% (w/w), which falls within the reported range for SFE-derived black pepper oil (Shrestha et al., 2020; Sweata et al., 2023). This value exceeds typical commercial black pepper essential oils (Kapoor et al., 2009) and confirms the effectiveness of OFAT-optimized SFE in producing high-quality extracts.

The physicochemical properties of the extracted Sarawak black pepper oil were within expected ranges for unadulterated essential oil: light to golden-yellow in colour (L 51, a* 2, b* 24), low viscosity (1.087 ± 0.041 mPa·s at 25 °C), refractive index of 1.4793 ± 0.10 and specific gravity of 0.862 ± 0.032 /mL. These values confirm that the extraction conditions preserved oil quality, minimized thermal degradation, and produced a stable, high-quality essential oil consistent with literature reports (Ravindran & Kallapurackal, 2012; Gupta & Sharma, 2019).

CONCLUSION

This study successfully used a One Factor At a Time (OFAT) approach to optimize the supercritical carbon dioxide (SC-CO₂) extraction of Sarawak black pepper oil, focusing on maximizing oil yield and maintaining bioactive quality. The results clearly show that extraction pressure, temperature, CO₂ flow rate, extraction time, and ethanol co-solvent concentration all significantly affect extraction performance.

Among the parameters studied, the optimal conditions were found to be a pressure of 300 bar, temperature of 50 °C, CO₂ flow rate of 6 g/min, extraction time of 90 minutes, and ethanol concentration of 10% (w/w). Under these conditions, the highest oil yield of $4.07 \pm 0.10\%$ was achieved, which was notably higher than yields at lower or higher pressures, temperatures, flow rates, extraction times, or ethanol concentrations (one-way ANOVA, Tukey's post-hoc, $p < 0.05$). Adding ethanol significantly increased solvent polarity and enhanced mass transfer, leading to better recovery of piperine and other key bioactive compounds ($p = 0.003$ compared to 0% ethanol).

Quantitative analysis confirmed a piperine content of 4.88% (w/w) in the extracted oil, which falls within the upper range reported for SFE-derived black pepper oil and exceeds typical commercial essential oil values. Physicochemical properties, including color, viscosity, and refractive index, remained within standard ranges for high-quality and unadulterated black pepper oil, indicating that the optimized SFE process preserves oil integrity and quality.

Overall, the findings confirm that OFAT-optimized SFE is an effective, clean, and sustainable extraction method for Sarawak black pepper oil. The study provides clear parametric insights into the effects of individual operating variables, offering a practical framework for process control and initial scale-up. These results add valuable knowledge for developing efficient, high-quality, and environmentally friendly extraction processes for spice-derived bioactive oils.

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