**Reviewing: 1**

Comments to the Author:

this manuscript is well written and can fill the research gap in the supercritical fluid extraction. in my opinion, this manuscript can be accepted to canadian journal of chemical engineering.

*Thank you*

**Reviewing: 2**

Comments to the Author:

In this manuscript, authors reported the application of sensitivity analysis to understand how the parameters of the model affect its output. In fact, this research implemented automatic differentiation to derive the sensitivity equations. Local sensitivity analysis techniques take into account only a small region of parameter space, and the conclusions derived from such an analysis are limited to local conditions. This work focuses specifically on the effect of pressure. Authors pointed out that for all the cases a pressure increment enhances mass transfer, leading to a faster mass transfer of solute from the particles. Also, they pointed out that the extraction yield is improved and characterized by positive sensitivities. Authors found that the system responses at low pressures were stronger than those at higher pressures. This behaviour can be described by the rapid changes in solvent properties occurring near the critical point of CO2. Authors also emphasized that local sensitivity analysis results offer valuable information by characterizing which parameters are influential and how these influences change over time. They concluded that the sensitivity analysis results can be utilized to design future experiments by specifying which parameters should be varied and observed. Finally, authors remarked that the parameters which are characterized by low sensitivity can be the subject of model modification.

In general, the manuscript is very attractive and interesting, as well as beneficial for applied purposes. I strongly recommend it for publishing in the Canadian Journal of Chemical Engineering. Meanwhile, the manuscript needs a major revision before its acceptance. I also invite authors to completely and fully address the comments raised here. The comments are explained below.

1. Please explain and highlight the novelties of this research work with respect to the previously published papers, in the manuscript.

*Thank you for your comment. The novelty of this work was included. Please see lines 279-285*

1. The motivations of conducting this research work are described.

*Thank you for your comment. The discussion on the motivation behind this study was elaborate. Please see lines 195-212*

1. English language of manuscript needs a mild revision throughout the manuscript.

*Thank you for your note. The manuscript went through proofreading.*

1. Please double check the equations and formulas of the used in the manuscript.

*Thank you for your comment. The equations and formulas have been double-checked.*

1. As you may know, supercritical fluids such as carbon dioxide (SC-CO2) has many applications in various topics. Authors are requested inserting a proper statement to emphasize the importance of the utilizing of SC-CO2 technology to be familiar for readers. For this purpose, write and insert this statement in the introduction section of the main manuscript along with fully cited references in below. "supercritical fluids like supercritical carbon dioxide (SC-CO2) has shown a great ability in various fields including extraction of

**essential oil [**

**~~https://doi.org/10.1016/j.supflu.2016.11.014~~, https://doi.org/10.1016/j.supflu.2014.07.023,**

**https://doi.org/10.1016/j.supflu.2016.04.006, https://doi.org/10.1016/j.supflu.2011.02.002,**

**https://doi.org/10.1016/j.supflu.2016.05.015, https://doi.org/10.1016/j.supflu.2017.04.007,**

**https://doi.org/10.1016/j.jtice.2015.11.003, ],**

**seed oil [**

**https://doi.org/10.1016/j.supflu.2016.08.019,https://doi.org/10.1016/j.supflu.2015.12.004, https://doi.org/10.1016/j.supflu.2017.12.026],**

**solubility [**

**https://doi.org/10.1002/cben.202200020, https://doi.org/10.1007/s11814-018-0125-6],**

**nanoparticle formation [**

**https://doi.org/10.1016/j.supflu.2017.10.015,**

**~~https://doi.org/10.1016/j.heliyon.2020.e04947~~, https://doi.org/10.1016/j.supflu.2021.105163,**

**~~https://doi.org/10.1016/j.jcou.2021.101799~~, https://doi.org/10.1016/j.supflu.2018.11.007**

**https://doi.org/10.1016/j.cherd.2018.12.020, https://doi.org/10.1016/j.fluid.2018.11.006,**

**https://doi.org/10.1016/j.supflu.2018.06.009],**

**impregnation**

**[https://doi.org/10.1016/j.supflu.2020.104892~~, https://doi.org/10.1016/j.supflu.2022.105674~~],**

**optimization and mathematical modeling [**

**https://doi.org/10.1080/14786419.2017.1361954,**

**https://doi.org/10.1016/j.supflu.2017.04.007],**

**polymer synthesis**

**[https://doi.org/10.1016/j.supflu.2022.105679], etc.**

*Thank you for your comment. The literature review was extended. See lines 31-45, 56-78, 105-127, 247-278*

1. Literature review on the available models could be completed in the introduction section.

*Thank you for this comment. The literature review on the available models can be found in the introduction. Chapter 2 discusses the model used by the authors.*

1. Could you provide quantitative results in the abstract or conclusion sections?

*Thank you for your comment. The discussion on the results was expanded by introducing normalized sensitivity coefficients invariant under the rescaling of model variables and parameters. An integral of the normalized sensitivities was proposed to quantify the cumulative effect of the parameter influence on the system. Please see lines 668-704 and 729-743*

1. Explain clearly the limitations of this study?

*Thank you for your comment. The discussion on the limitations of this work was extended. Please see lines 668-675 and 692-704.*

1. Please provide abbreviations and symbol lists.

*Thank you for your comment. The list of symbols and abbreviations can be found on the last page of the manuscript.*

1. It is recommended removing the last paragraph of conclusion section.

*Thank you for your comment. The last paragraph of the conclusion section was removed.*

1. Please provide consistency between the abstract and conclusion sections.

*Thank you for your comment. The consistency was improved by making changes in both the abstract and conclusion sections.*

1. The number of references could be increased for this valuable research work.

*Thank you for the comment. The number of references increased from 30 to 38*

**Reviewing: 3**

Comments to the Author:

The topic of manuscript is modelling of supercritical extraction of camomile oils. The work is relevant and in generally well written. The structure is logical, starting from a literature review on state-of-art, progressing to mathematical models and finally to simulations. The final goal is sensitivity analysis to get perspective on the effect of various parameters affecting the extraction efficiency. It can be accepted after minor revision. I list some points below:

1. The highlights are somehow very abstract - could they be partially re-written, thinking the understanding of a general reader?

*Thank you for your comment. The highlights have been modified.*

1. In equation (8) the author talk about 'axial diffusion coefficient'. In fact the equation has the 'axial dispersion coefficient' which can be very different from the diffusion coefficient, depending on the degree of backmixing and the presence of turbulent eddies.

*Thank you for your comment. The variable's name was corrected to the 'axial dispersion coefficient.'*

1. Using one (1) pseudo-component is a dramatic simplification, even though understandable. Please provide a comment on this issue. Has something more advanced been published in literature?

*Thank you for your comment. Indeed, using a pseudo-component is a simplification and having a multi-component system would make this problem even more interesting to solve.*

*The main issue is the availability of an appropriate dataset, as this work utilizes the data previously published by other researchers. Our model describes a dynamic system, which means a time series of measurements at different operating conditions is required for parameter estimation. To our knowledge, the number of publications which deliver the time evolution of an extract's composition is rather low. Unfortunately, we did not find a good source of data that presents the time evolution of an extract composition at different operating conditions, including temperature, pressure, and mass flow rate. The primary motivation behind this work is to focus on the influence of operating conditions on the extraction process. We decided to focus on the dataset, which covers multiple operating conditions even at the cost of using a single pseudo-component extract.*

1. The discussion on extraction mechanisms is as such good, but which one to select?

*Thank you for your comment. When external mass transfer dominates, solute transport resistance occurs at the fluid-solid interface and is best described by the* ***Bulley formulation,*** *which emphasizes fluid-phase concentration as the driving force. Conversely, when internal mass transfer controls the process, resistance arises from diffusion or adsorption within the solid phase, making the* ***Reverchon formulation*** *more suitable, as it accounts for deviations in equilibrium through solid-phase concentration. The discussion on the extraction mechanism selection was introduced to the main text. Please see lines 404-417.*

1. The future perspectives could be discussed. Experimental activities in future?

*Thank you for your comment.*

*The future activities involve more computational work. The sensitivity analysis results will be used in our future work on optimal model-based experiment design.*

*Regarding the experimental activities, we work on modifying our equipment, which would allow us to perform more experiments focusing on the influence of operating conditions on extraction efficiency and physical phenomena. To investigate the system better, we added a set of temperature probes inside the extraction chamber, allowing us to recreate the axial and radial profiles. We plan to run a set of experiments where the operating conditions change during the batch and observe how the yield and temperature profiles change over time. Such an experiment will help us to research the mass and heat transfer in supercritical extraction systems.*

**Reviewing: 4**

Comments to the Author:

Title：Local Sensitivity Analysis of a Supercritical Extraction Model

Number: CJCE-24-1047

The authors studied the effect of pressure on the model state and extraction yield. They investigated the process of chamomile oil extraction from chamomile flowers. This work is meaningful and can be published on CJCE after addressed the following concerns.

1. The authors should evaluate the models investigated by including statistical parameters such as Average Absolute Relative Deviation (AARD), R², and Adjusted R² to assess the accuracy and reliability of the models.

*Thank you for your comment. This publication is based on our previously published article (DOI: 10.1002/cjce.25557), which discusses the parameter estimation problem and delivers a set of statistical metrics related to the goodness of fit.*

1. To include more than one material for extraction studies to enhance the scope and generalizability of the results. Currently, the manuscript focuses only on chamomile flowers. Adding additional materials will strengthen the study's comparative analysis.

*Thank you for your comment. We agree that such a generalization of discussion would be very beneficial and allow for the development of a more robust mathematical model. Due to technical limitations, we had to rely on the dataset published by other researchers for model calibration. Unfortunately, there seems to be a limited amount of good-quality datasets considering the extraction yield's time evolution at different operating conditions, including temperature, pressure, and mass flow rate. Regarding the experimental activities, we are modifying our equipment, which will allow us to perform more experiments focusing on the influence of operating conditions on extraction efficiency and physical phenomena. We added temperature probes inside the extraction chamber of our modified equipment to better investigate the system's dynamic. Our subsequent work will use lingonberry press cakes and spruce needled for wax extraction.*

1. Provide a table or figure to present the model analysis, clearly demonstrating the agreement between model predictions and experimental data. This visual representation will help validate the model's performance and improve the clarity of the findings.

*Thank you for your comment. Indeed, the comparison between model prediction and a real system's response would help validate the model and enhance the understanding.*

*Due to technical limitations, we had to rely on the dataset published by other researchers for model calibration. For this reason, we cannot recreate the same experimental conditions, which would be necessary for reliable tests. After we modified our equipment, we could perform a test with chamomile flowers. However, they might not be representative, considering the different equipment setups and original raw materials.*

*The limitations of this study and its relation to the corresponding experiment were discussed. Please see lines 692-704*