Modelling and Simulation of a Fully Continuous Pilot-Scale SFE Process

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ABSTRACT

A fully continuous supercritical fluid extraction (SFE) process is necessary for the cost-effective treatment of high volumes of solid materials containing valuable compounds. A continuous pilot scale SFE system has been developed using an extractor (with counter current SC-CO_2 and solid-slurry flows) and a separator (to recover the extracted components from SC-CO_2).

Dynamic models of this pilot scale system are important to understand the behavior of the system thus allowing improvements in operating strategies and designing of control systems. In addition, these models ultimately aid in the design and development of subsequent commercial scale systems. The main process variables include pressure, temperature and slurry levels within the extractor as well as extracted compound concentrations. The focus of this work is therefore to develop a hydrodynamic model of the continuous pilot scale SFE system to understand the response of pressure, temperature and slurry levels in the extractor and pressure and temperature in the separator as a function of process parameters such as outlet valve openings and heat input.

The results show that the presence and conditions of the separator impacts the steady state pressure of the extractor, which, in turn, has an effect on the slurry level inside the extractor. The transient response of the extractor is only slightly impacted by the separator conditions. Transient response results were also able to identify which process variables are most appropriate to use in controller design. The results provide insight into the degree of interaction between process variables and the process units. This finding is a key step towards safe operation, optimal extraction and accurate control of this fully continuous SFE process.

INTRODUCTION

Supercritical fluid technology is an effective technique for extracting compounds from solids using supercritical carbon dioxide (SC-CO_2) [1,2]. Research demonstrates successful extraction and high removal efficiencies using mostly batch or semi-batch SFE systems [3,4,5]. Application of a fully continuous SFE process will overcome economic challenges and labour intensive operations associated with batch processes [6]. Therefore, a pilot scale fully continuous SFE process has been designed, constructed and operated at the University of Alberta. This paper reports on the continuing development of a hydrodynamic model associated with this fully continuous process applicable to the extraction of oil from solid slurries.

Mathematical/hydrodynamic modelling of SFE processes has been the subject of only a few studies reported in the literature. Ramachandran *et al.* [7,8] conducted a study on modelling and control of a SFE process for extraction of iPA from water using SC-CO_2. The developed model relied on mass and energy balances, as well as on phase equilibrium. Cesari *et al.* [9] simulated a semi-continuous SFE process applied to fractionation of ethanol from water and the extraction of citral from lemon oil using SC-CO_2. The developed model relied on mass and energy balances, as well as on phase equilibrium and hydrodynamic relationships. Ruivo *et al.* [10] and Fernandes *et al.* [11, 12, 13] conducted a comprehensive study of modelling a SFE process for the fractionation of a binary liquid mixture of squalene and methyl-oleate using SC-CO_2. The modelling involved equations describing mass transfer, thermodynamic phase equilibrium and hydrodynamics.

In the fully continuous pilot scale SFE process, the pressure and slurry level in the extractor are the two main variables that must be controlled throughout an experimental run. This control strategy is important both from a safety and extraction efficiency perspective. Based on previous work [14, 15], the slurry and CO_2 pump flows were selected as the manipulated variables to control the slurry level and pressure in the extractor, respectively. Controlling the pressure and temperature of the separator at certain set points is also important to allow effective separation of the oil from the CO_2, to minimize process upsets in the separator and to minimize the energy cost of CO_2 recirculation in a continuous SFE process.

Previous publications from our group report on the hydrodynamic model development and validation as well as control associated with only the extractor [16, 17]. In this work, the separator has been added to the hydrodynamic model to investigate the impact of its presence on process dynamics specifically pressure, temperature and slurry level in the extractor and pressure and temperature in the separator. Inclusion of the separator is important because of the interactions that exist between the extractor and separator variables. Understanding these interactions and the resulting complexities, aids in effective operation and control of this fully continuous pilot scale SFE process and subsequent fully continuous commercial scale systems. Therefore, the main objective of this study is to develop a hydrodynamic model of a fully continuous pilot scale SFE process including both the extractor and the separator. As part of this objective, simulations are conducted on the developed hydrodynamic model at different operating conditions and process variable responses are validated/analyzed and compared.

PROCESS DESCRIPTION AND MODELLING

Fully continuous SFE process

A schematic diagram of the pilot scale fully continuous SFE process and relevant control volumes used in modelling the process are presented in Figure 1.

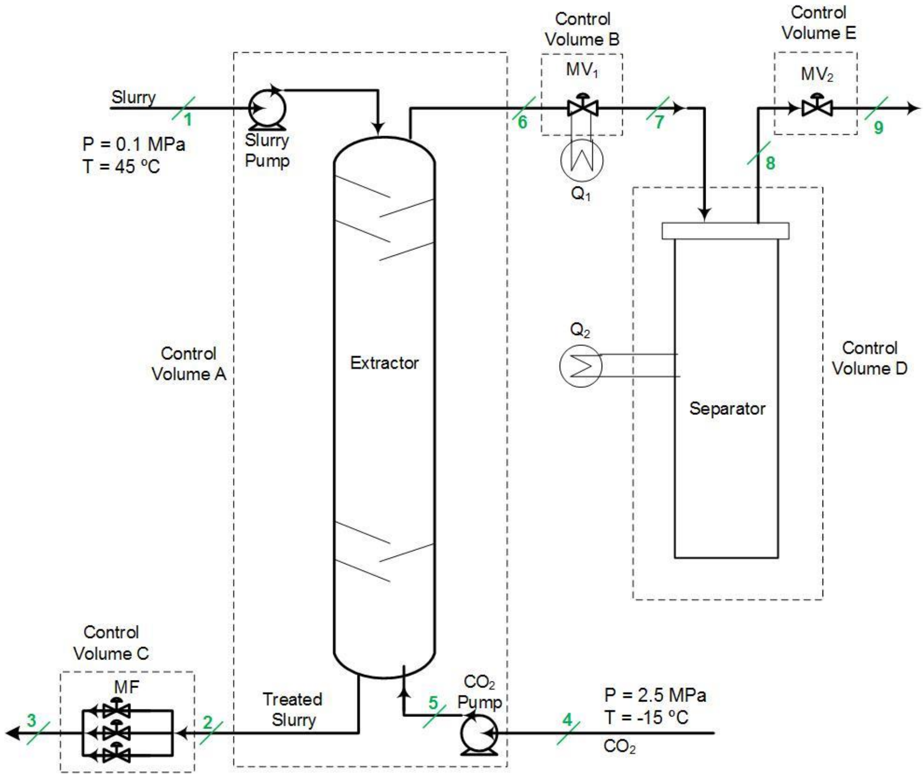

Figure 1. Schematic diagram of the process and corresponding control volumes

The slurry (a mixture of oily solids and water) and SC-CO_2 are pumped into the top and bottom of the extractor, respectively. The two phases come into contact counter currently in the extractor. The SC-CO_2 extracts the oil from the slurry phase and exits from the top of the extractor, where it goes through a metering valve (MV_1) for depressurization before entering the separator. It should be noted that a heating band with an On/Off heating function (Q_1) is located on MV_1. The slurry exits from the bottom of the extractor and flows through a manifold (MF) to depressurize before entering the slurry receiving tank. The extractor and separator have a volume of approximately 12.5 L and 7.5 L, respectively.

In the separator, due to the depressurization of the CO_2 phase, the oil is separated from the CO_2 and accumulates at the bottom of the separator. The CO_2 exits the separator, where it flows through a heated metering valve (MV_2) and is finally vented in to the fume hood. A double pipe heat exchanger (Q_2) provides heat to MV_2. Additional information regarding the continuous pilot scale process is presented in Roodpeyma *et al.* [16].

Modelling methodology

The hydrodynamic model was developed by applying mass and energy balances to the slurry and the SC-CO_2. To calculate CO_2 outlet flowrate from both the extractor and separator (i.e. the flow through MV_1 and MV_2), an empirical equation describing the behaviour of gas flow through the metering valve as provided by the valve literature was used [18]. To calculate the slurry outlet flowrate from the extractor (i.e. the flow through MF), the Darcy-Weisbach equation for pressure drop in a circular pipe was applied. For this purpose, the Swamee-Jain equation [19] was used to calculate the Darcy friction factor. The Span and Wagner fundamental equation of state [20] was used to calculate SC-CO_2 properties, specifically the pressure, enthalpy and constant volume heat capacity of the SC-CO_2 (as a function of the temperature and density). The developed equations were numerically solved using Simulink® coupled with a program written in C to solve the fundamental equation of state relationships.

SIMULATION RESULTS AND DISCUSSION

After the development of the hydrodynamic model, two sets of simulations were conducted. In the first set of simulations, the dynamics of the separator and extractor was investigated as a function of the separator's presence and the status of its heating (Q_2) and valve (MV_2) opening. In the second set of simulations, sensitivity analysis was conducted to investigate the degree of interaction between input and output process variables in the extractor and separator.

Impact of separator presence on process dynamics

Four separator cases (Table 1) were defined, simulated and compared to investigate the impact on the process dynamics. The Base Case represents the hydrodynamic model for the extractor in the absence of a separator the results of which have been previously published [16]. The Base Case conditions were set to result in a constant pressure of 2.5 MPa downstream of MV_1. Figure 2 demonstrates the separator's pressure and temperature dynamics for Cases 1-3.

Table 1. Separator cases for investigating the impact of separator presence

Case	Separator presence	Q_2	MV_2 opening
Base Case	No (Fixed Pressure after MV_1)	N/A	N/A
Case 1	Yes (Dynamic Pressure)	No	2.3 *
Case 2	Yes (Dynamic Pressure)	No	1
Case 3	Yes (Dynamic Pressure)	Yes	1

* This value was needed to achieve the same pressure after MV_1 as in the Base Case

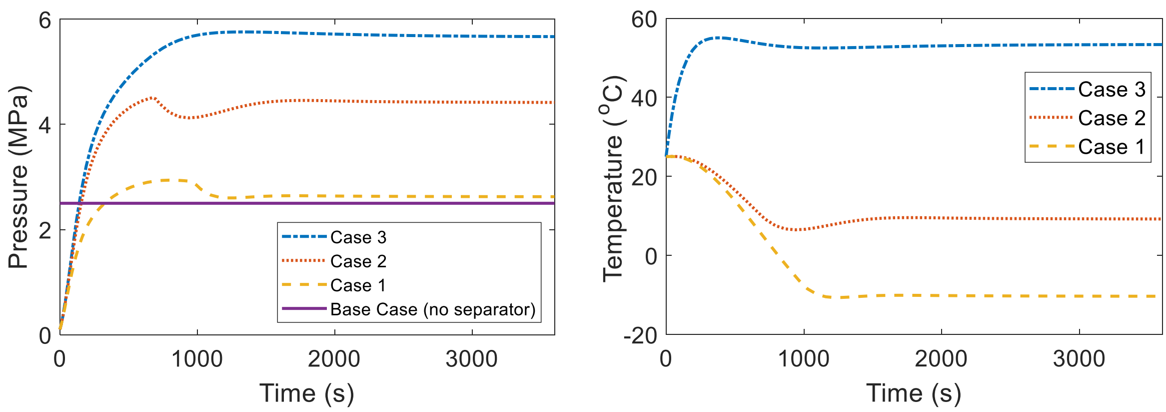

Figure 2. Separator dynamics for Cases 1-3

Case 1, and its MV_2 selected value, was to match the Base Case fixed pressure context at steady state. A MV_2 value of greater than 1 indicates that the addition of a wide-open metering valve is more restrictive to flow than no valve at all. Case 2 considered the metering valve in its wide-open position (a value of 1). In comparing Cases 1 and 2, decreasing MV_2 results in an increase in steady state pressure response from 2.6 MPa to 4.4 MPa. This was expected due to the increase in resistance on the CO_2 outlet line from the separator when decreasing the opening of MV_2.

For Cases 1 and 2, the separator's pressure response is not a smooth approach towards a steady state value. At nearly 1000 s for Case 1 and approximately 700 s for Case 2, the slope in the pressure changes dramatically. In both cases, this rapid slope changes occurs when the CO_2 in the separator begins a phase change from gas to liquid as the temperature in the separator is cooling. The resulting steady state conditions in the separator for Cases 1 and 2 is liquid CO_2. The resulting steady state temperature corresponds to the vapour-liquid equilibrium temperature at the steady state pressure conditions.

Case 3 introduces heating to the separator relative to Case 2. Pressure, in Case 3, makes a smooth transition from the initial condition to the steady state conditions. During Case 3, the CO_2 remains gaseous throughout. Heating the separator leads to a higher steady state pressure (5.7 MPa) and temperature (53 °C) in comparison with Case 2.

Next, the impact of the separator cases (different status of Q_2 and MV_2) on the dynamics of the pressure and slurry level in the extractor was investigated. Figure 3 demonstrates extractor dynamics for the cases.

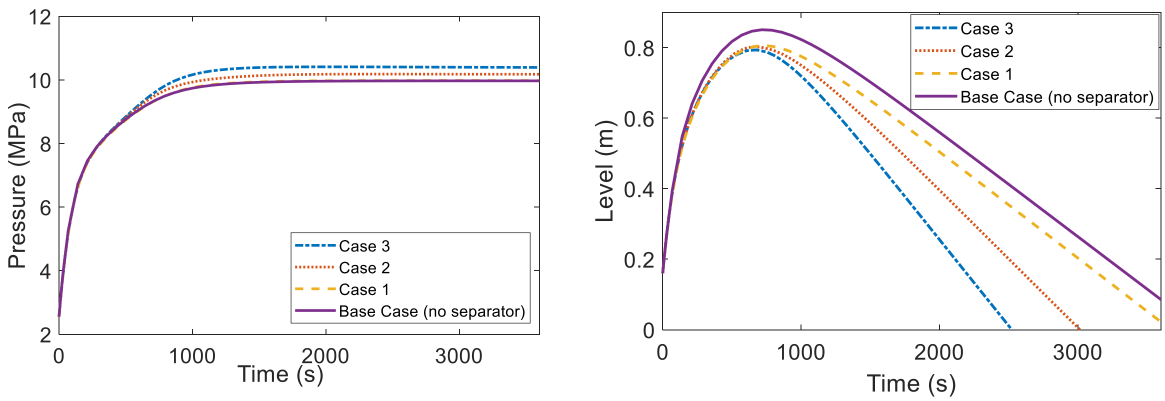

Figure 3. Extractor dynamics for the four defined cases

As seen in Figure 3, in comparing the Base Case and Case 1, the behaviour of the pressure is very similar (i.e. different by only 0.01 MPa). The slightly higher pressure in Case 1 results in a slightly faster level response compared to the Base Case. Therefore, adding the separator (with an MV_2 opening of 2.3 and providing no heat) does not significantly change the dynamics of the extractor compared to the case in which only extractor dynamics are taken into consideration (i.e. Base Case).

In comparing Case 2 and Case 3 with the Base Case, it is concluded that decreasing MV_2 and adding heat to the separator (Q_2), both result in an increase in the steady state pressure of the extractor (2.5 % increase for Case 2 and 4.6 % increase for Case 3). The increase in pressure in turn effects the slurry level i.e. the higher pressure increases the flow rate of slurry out of the extractor. It should be noted that an insignificant change was observed in the extractor temperature between Cases 1-3 and the Base Case. Therefore, the temperature dynamics have not been shown.

The results demonstrate the various impacts that separator presence has on process variables in the extractor and the separator itself. In addition, it was observed that changing MV_2 opening and heat input of Q_2, can significantly shift both the pressure and temperature in the separator.

Sensitivity analysis: Investigating the degree of interaction

Sensitivity analysis has explored the impact of changes in metering valve opening and heating on the system. These results enhance understanding and are important for the development of control strategies. The response of the extractor and the separator on the opening of the two metering valves is explored first. This is followed by the response of the separator and extractor to the two heat input locations.

The impact of changing MV_1 and MV_2 on the extractor and separator dynamics is presented in Figure 4. The left column is associated with the change in MV_1, while the right column is associated with the change in MV_2. Both MV_1 and MV_2 are independently increased and decreased by 10% (MV_1 from 0.32 to 0.42 and 0.22, MV_2 from 1 to 1.1 and 0.9).

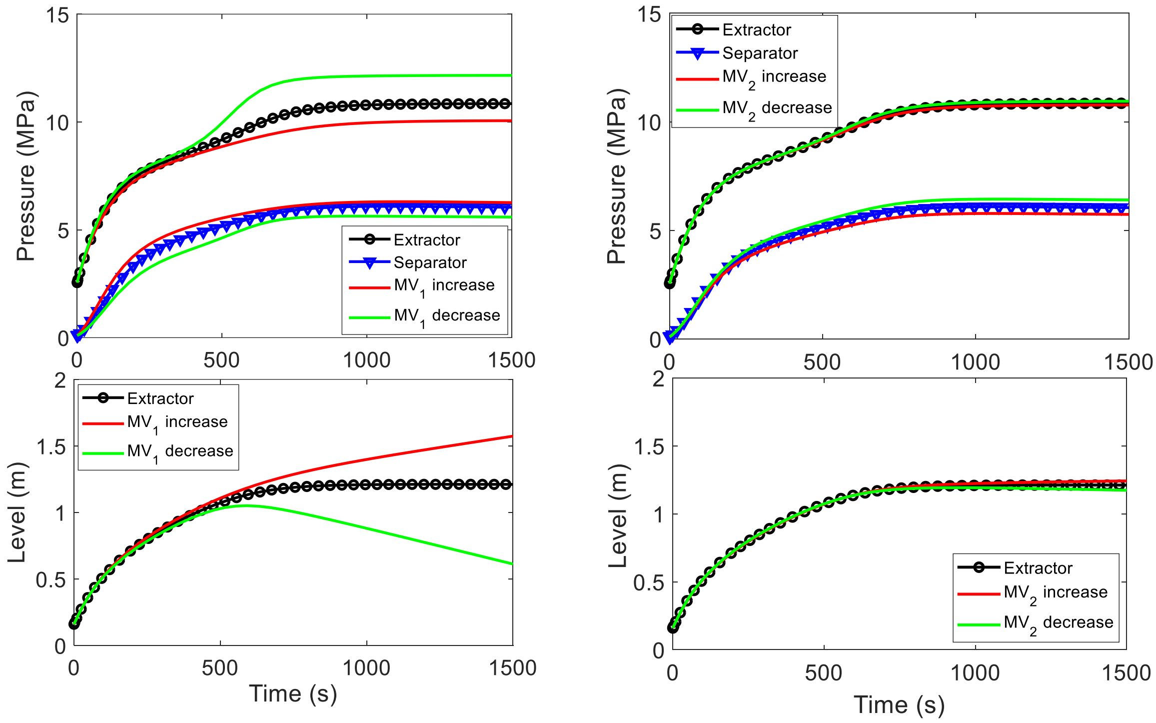

Figure 4. The impact of changing MV_1 (left column) and MV_2 (right column) on extractor and separator dynamics

As seen in Figure 4, the 10% change in MV_1 significantly impacts the pressure and, as a consequence, impacts the slurry level in the extractor. However, the 10% change in MV_2 only slightly impacts pressure and level in the extractor. In terms of the separator pressure, the 10% change in MV_2 has more impact compared to changing MV_1. A change in either MV_1 or MV_2 does not significantly impact temperature in the extractor or the separator, therefore the results have not been shown. To summarize, manipulating MV_2 opening (as compared to MV_1) has more impact on the separator pressure and less impact on the extractor pressure and level. Therefore, MV_2 opening is the preferred manipulated variable towards controlling separator pressure.

A similar approach was conducted to investigate the impact of changing the heat rate of Q_1 and Q_2 on the extractor and the separator dynamics. As previously stated, Q_1 has an on/off function while Q_2 operates as a double pipe heat exchanger. The results are presented in Figure 5. The left column is associated with the change in Q_1, while the right column is associated with the change in Q_2. Both Q_1 and Q_2 are independently changed i.e. Q_1 is turned On (720 W) or Off while, the water temperature associated with Q_2 is increased and decreased by 10 °C (i.e. from 60 °C to 70 °C and 50 °C).

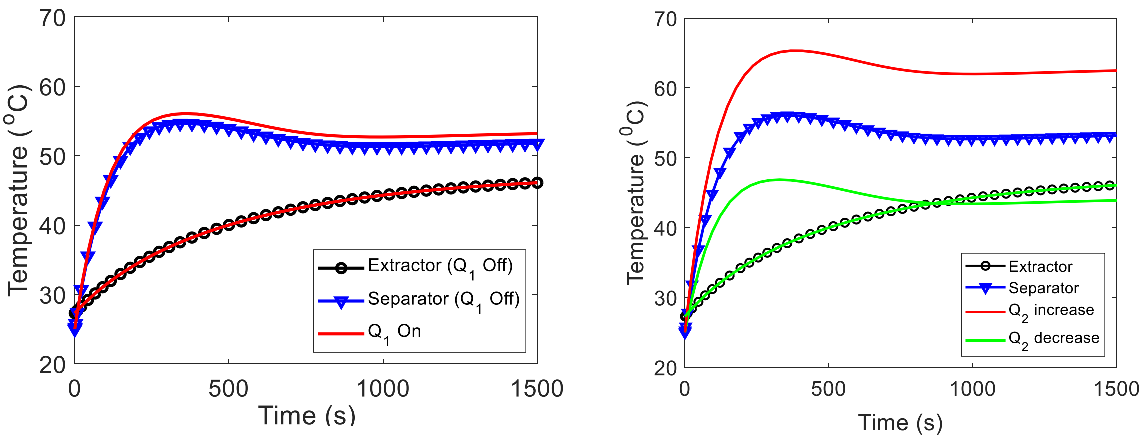

Figure 5. The impact of changing Q_1 (left column) and Q_2 (right column) on extractor and separator dynamics

As seen in Figure 5, applying Q_1 results in an increase of 1 °C in the steady state temperature only in the separator. No change is observed in the extractor temperature. Applying Q_1 also does not shift pressure responses in the separator or in the extractor. Therefore, results have not been presented.

The 10 °C change associated with Q_2, significantly impacts separator temperature, but again no change is observed in the extractor temperature. Conclusively, applying Q_2 has more impact on separator temperature compared to the case in which Q_1 is applied. Therefore, Q_2 is the preferred manipulated variable towards controlling temperature in the separator. This was expected as Q_2 is located on the separator and directly applied to it.

In summary, the results demonstrate that MV_2 and Q_2 should be used towards controlling pressure and temperature in the separator, respectively.

CONCLUSION

A hydrodynamic model characterizing a continuous pilot scale SFE process including the extractor and the separator is developed. The model is based on mass and energy balances and an equation of state for SC-CO_2. Simulations were performed in Simulink® to investigate the impact of the separator presence on process dynamics and to analyze the degree of interaction between the input and output process variables of the extractor and the separator. It was concluded that the separator presence has a slight effect on extractor dynamics and that the conditions of the separator (MV_2 and Q_2) could significantly shift separator responses. The results also demonstrate the importance of investigating the degree of interaction between the main unit operations of the SFE process (the extractor and the separator). Consequently, MV_2 and Q_2 were selected for controlling pressure and temperature in the separator, respectively. The obtained results are valuable for improving operating strategies and completing the control system design of the fully continuous pilot scale SFE process. The results will also prove valuable for subsequent scale-up and commercialization of the SFE process under investigation.

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