Economic Feasibility of an Integrated Semi-Batch Reactive Distillation Operation for the Production of Methyl Decanoate

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Abstract

The formation of methyl decanoate (MeDC) by esterification reaction of decanoic acid with methanol through batch/continuous reactive distillation columns is operationally challenging, energy intensive and thus cost intensive operation due to complex thermodynamic behaviour of the reaction scheme. Aiming to overcome the equilibrium restriction of the decanoic acid (DeC) esterification operation, to improve the process efficiency and to reduce the total annualised cost (TAC), the semi-batch distillation column (SBD) and the recently proposed integrated semi-batch distillation column configuration (i-SBD) are investigated here.

The performances of each of these column operations are evaluated in terms of minimum TAC for a given separation task. In both column configurations, additional operating constraints are considered into the optimization problem to prevent flooding of still pot due to the continuous charging of methanol into it. This study shows the superiority of i-SBD mode of operation over SBD operation in terms of TAC.

Also, the optimization results for a defined separation task indicate that the performance of multi-interval control policy is overwhelmingly superior to the single-interval control operation in terms of operating batch time, and overall annual cost in the i-SBD system providing about a time saving of 82.75%, and cost (TAC) saving of 36.61% for a DeC (product) concentration of 0.945 molefraction.

Keywords: Integrated Semi-Batch Distillation; Modelling; Optimization; Batch Operation, Methyl Decanoate, Total Annualised Cost

1. Introduction

Batch distillation processing is receiving renewed consideration in recent years (Reddy et al., 2017; Marquez-Ruiz et al., 2019; Ferchichi et al., 2020; Fonseca et al., 2020; and May-Vázquez et al., 2020). With the increased environmental and economic requirements, the optimization techniques become more and more significant as any inappropriate choice can lead to higher total cost and energy penalties. It is well known energy demand in the world will continue to increase. It is expected that there will be a 48% of increase in energy demand in 2040 compared to that in 2012. Nowadays, about 86% (of which, 32% crude oil, 30% coal, and 24% natural gas) of the worldwide energy requirements is satisfied by fossil fuels (Cui et al., 2017; Parhi et al., 2019a, 2019b; Gor et al., 2020).

The dreadful significance of using fossil fuels results in air and water pollutions, and global warming and thus climate change. To combat these problems, more considerations must be given in improving the energy efficiency (thus cost) of industrial operations including distillation which is inherently an energy intensive process (Mujtaba et al.,2012).

Methyl decanoate (MeDC) as biodiesel fuel, is known as methyl caprate, can be manufactured via the esterification reaction of decanoic acid (DeC) and methanol (Steinigeweg and Gmehling, 2003; Hernandez et al., 2010; Nguyen and Demirel, 2011; Machado et al., 2011; Noshadi et al., 2012; Lamba et al., 2018). Aqar et al. (2017a, 2017c, 2018a) used conventional batch distillation (CBD), integrated conventional batch distillation (i-CBD), semi-batch distillation (SBD) and integrated divided wall (i-DWC) columns for the synthesize of MeDC.

The economic feasibility in terms of total annualised cost (TAC) for batch distillation operation for some reaction schemes has been discussed by a number of investigators in the open literature (Jana and Maiti, 2013; Maiti et al., 2013; Maiti and Jana, 2013; Kao and Ward, 2016). As an example, Jana and Maiti (2013) developed an ideal internal heat integrated apparatus for batch distillation operation for the synthesize of n-butyl acetate. The thermally integrated batch configuration includes a rectifying column equipped with a total condenser at the top and concentrically bounded by a reboiler from the bottom. Maiti and Jana (2013) proposed a novel combination of internal and external heat integrations for batch distillation column for the synthesize of n-butyl acetate. Their hybrid thermally integrated scheme was found economically superior to its conventional batch distillation counterpart in terms of lower annualised cost. Maiti et al. (2013) presented a novel heat integrated batch reactive distillation having a jacketed reboiler to reduce the energy consumption rate and thus the total annualised cost for the production of ethyl acetate.

Recently, Kao and Ward (2016) investigated the simultaneous optimization of design (e.g. a total number of trays and vapour boil-up rate) and operating variables (reflux ratio and catalyst loading) of batch distillation operation for two reaction systems: formation of lactic acid and formation of methyl formate.

The influence of equipment design and operating variables on the total annualised cost problem was studied. In this work, the application of integrated semi-batch distillation operation (i-SBD) recently developed by Aqar et al. (2016a) but applied to methyl lactate system is employed and the performance of i-SBD column is evaluated by minimizing the total annualized cost via minimizing the processing batch time. The aim of this work is (1) to investigate different i-SBD scenarios by simulation in a case study, (2) to compare the specific total annual costs of the different policies for a reaction system, (3) to re-use recently established i-SBD process for this system and (4) to study the influence of multi-interval control policy on the overall performance.

A detailed dynamic process model based on mass and energy balances is used within the optimization framework built in gPROMS (general PROcess Modeling System, 2015) software. The dynamic optimization problem is converted to a nonlinear programming (NLP) problem, which is solved by using Control Vector Parameterization (CPV) method using successive quadratic programming (SQP) technique within gPROMS (more details about this technique can be found in Mujtaba, 2004).

2. Column Configuration and Process Modelling

2.1 Integrated Semi-batch distillation Operation (i-SBD) with methanol recycle

Figure 1a shows SBD column without methanol recovery and recycle option. In the i-SBD configuration, the distillate accumulator from the SBD (Figure 1a) is further proceeded in a CBD mode to separate methanol at a desired concentration (say, 0.950 molefraction) and combined with make-up methanol of the same concentration before being fed to the next batch of SBD (Batch 3 onward) in the i-SBD process.

The process strategy for i-SBD configuration is presented in Figure 1b, which is translated in Figure 1c for easy understanding. The CBD of batch 1 is allowed to have the same operation time as SBD of batch 2, so that the methanol rich distillate from CBD of batch 1 can be fed to the SBD of batch 3. Thus, after few batches a quasi-steady state operation is achieved (see Mujtaba and Macchietto, 1992) for the concept of quasi-steady state operation. Note, t_S is the joint setup batch time for SBD and CBD operations. For modelling this column configuration, the following assumptions are made:

- Perfect mixing on each stage.
- Vapor phase is ideal and Liquid phase is non-ideal.
- Fast energy dynamics.

- Negligible vapor holdup.
- Negligible heat loss to surroundings.
- Constant tray pressure drop and tray efficiency (vapor-phase Murphree efficiency 100%).
- Total condensation (no sub-cooling).
- Reaction on the stages, in the total condenser, and in the reboiler.

Further details about these column configurations and the mathematical dynamic models (except the reaction rate and phase equilibria for MeDC esterification process) for these two configurations can be found in detail in Aqar et al (2016a, b, c).

2.2 Chemical kinetic and thermodynamic data

The modified kinetic model (LH) for the synthesis of methyl decanoate (MeDC) through the esterification reaction of decanoic acid and methanol (MeOH) by using the heterogeneous catalyst (Amberlyst-15) was explored experimentally by Steinigeweg and Gmehling (2003). The rate expression of the modified Langmuir−Hinshelwood (LH) kinetic model is:

$$-r_{DeC}= m_{cat}\left\{\frac{3.1819\times10^{6}\exp\left(\frac{-72230}{RT}\right)a_{DeC}\,a_{MeOH}}{(2.766\,a_{H_2O})^{2}}-\frac{3.5505\times10^{5}\exp\left(\frac{-71900}{RT}\right)a_{MeDC}}{(2.766\,a_{H_2O})}\right\} \quad (1)$$

Where, $-r_{DeC}$ is the rate of reaction, m_{cat} is the total amount of catalysis loaded, a_i is the activity of each component, and T is the temperature. For thermodynamic model (VLE), the studied system has four components, including two reactants (DeC and MeOH) and two products (MeDC and H_2O). The nonrandom two-liquid (NRTL) model was employed to compute the liquid activity constants (the vapour-liquid equilibrium) of each component. The NRTL model binary interaction parameters are obtained from Aspen HYSYS V8.8 package, which are same as those presented in the previous work (Aqar et al., 2018a). Vapour phase is assumed to be ideal.

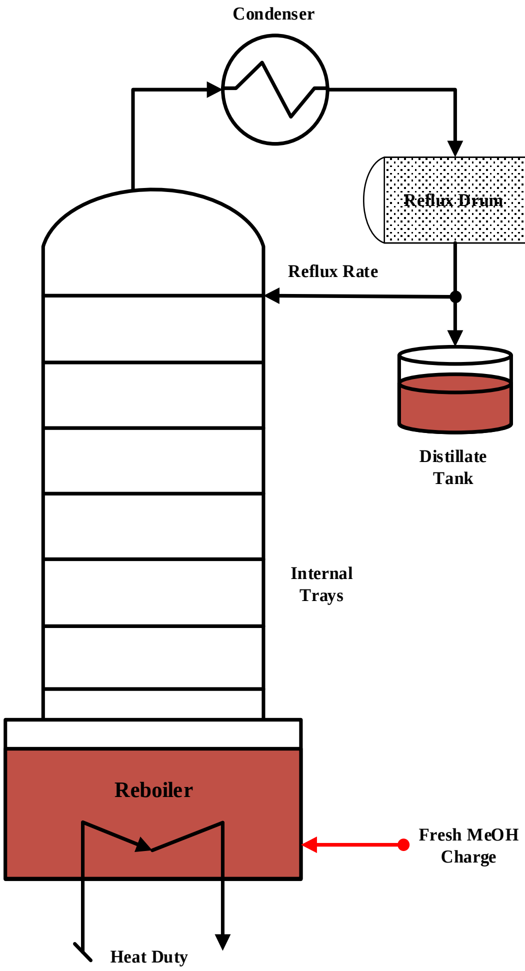

Figure 1a: Scheme of the single semi-batch distillation (SBD) column.

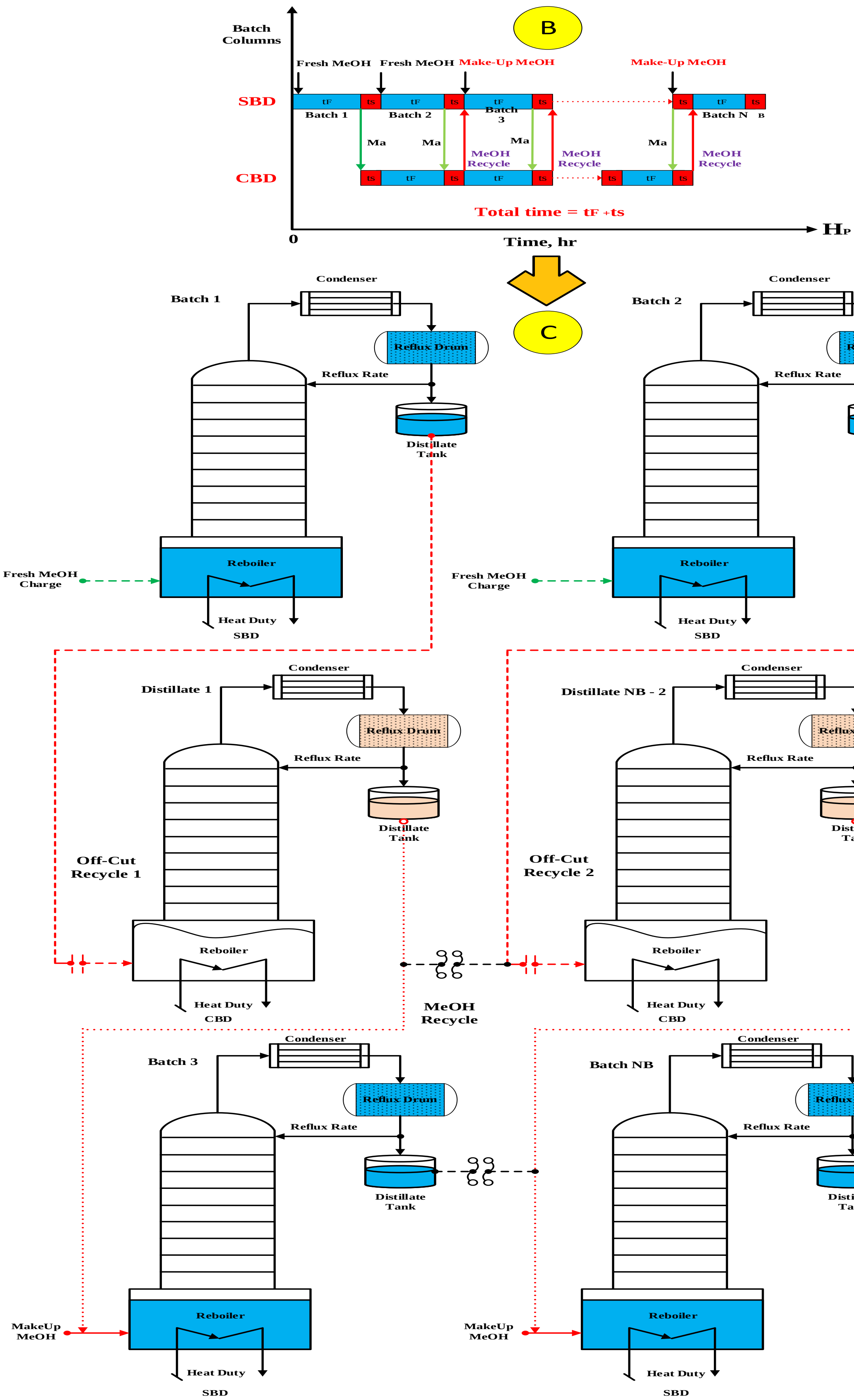

Figure 1b, c: The operating policy for SBD and i-SBD operation.

3. Economic evaluation

The Total Annualised Cost (TAC) is employed as an economic indicator for determining the economic feasibility of i-SBD process. In this work, TAC is evaluated based on minimizing the operating batch time (t_F) while the operational parameters such as reflux ratio (R_{CBD}) and methanol feed rate (F_{MeOH}) are optimised and therefore a single objective optimisation problem is considered rather than multi-objective optimisation (e.g. simultaneously maximising the amount of product and minimising the TAC). The cost equations for TAC analysis are employed (Douglas, 1988) as shown in the following form:

$$\text{TAC} = \text{Annual operational cost} + \frac{\text{Annual capital cost}}{\text{Payback period}} \qquad (2)$$

Where the operational cost is the summation of the reboiler energy cost, the cooling water cost, the catalyst costs, as well cost of methanol (fresh or make-up) charged for both SBD/i-SBD operations. For both (SBD and CBD) columns, the annual capital cost includes the cost of the columns, stages, condenser and reboiler. Note, as the number of stages reflects the capital cost, it should also be optimised if the objective function is to minimise the TAC. However, in this work column configuration (including number of stages) is already considered fixed.

In this work, a payback period of 5 years is assumed to be used and a catalyst is assumed to be replaced every 3 months. Note, the cost of methanol charged at the purity of 0.950 molefraction is 3.07 ($/kmol) which is taken from (Alibaba Trade, 2020).

It can be noted that total annual cost (TAC) and the process duration (t_F) with the distillate amount (D) are totally dependent because (t_F) includes the quantity of charged methanol, D_{MeOH} includes the amount of make-up methanol that are used for the computing of total operational cost (OC). Therefore, the minimization of operation time means indirectly minimizing the total annualized cost at the fixed batch setup time for each batch cycle. The cost equations for computing the TAC are given in detail in the Appendix.

4. Formulation of optimization problem

In this work, the performance of the column configurations (SBD/i-SBD) is evaluated in terms of minimum batch time under single and multi-reflux intervals modes for a given bottom product amount and desired purity. The reduction in the operating batch time leads to a decrease the cost of methanol (fresh or make-up) charged (see equations 18 and 19) and thus the annual operating cost and total annualized cost.

The optimization problem can be described as follows:

4.1 Minimum Operating time Problem for SBD Column (of i-SBD process)

Given:	The SBD column configuration, the feed concentration, the condenser Vapour rate, desired amount of MeDC product and composition constraint.
Optimize:	The reflux ratio (R_{SBD}), and the MeOH feed rate (F_{MeOH}) profiles
So as to:	Minimize the operating batch time (t_F)
Subject to:	Process constraints (still pot overfilling, etc.), Model equations

Mathematically, the optimization problem (OP1) can be represented as follows:

OP1 Min t_F

$R_{SBD}(t)$, $F_{MeOH}(t)$ (3)

Subject to :

$f(t, \dot{x}(t), x(t), u(t), v) = 0$; $[0 \quad t_F]$ (Process model, equality constraint)

$B_{MeDC} \geq B^*_{MeDC}$ (Inequality Constraints)

$x_{MeDC} \geq x^*_{MeDC}$ (Inequality Constraints)

Linear bound on $R_{SBD}(t)$, $F_{MeOH}(t)$ (Inequality constraints)

$f(t, \dot{x}(t), x(t), u(t), v) = 0$, represents the process model shown in section 2, where t denotes the independent variable (operating batch time), x(t) gives the set of all differential and algebraic variables, $\dot{x}(t)$ represents the derivative of differential variables with respect to batch time, u(t) denotes the control variables, and v denotes the set of the design variables (fixed parameters). The switching time of interest is $[0 \quad t_F]$, and the function f: is assumed to be continuously differentiable with respect to all its arguments (Ekpo and Mujtaba, 2007, Aqar et al., 2017b, 2018b, 2018c). B_{MeDC} and x_{MeDC} are the amount of bottom product and quality of MeDC at end batch time (t_F) in the reboiler, (denotes that the $B^*_{MeDC} \times x^*_{MeDC}$ are specified). R_{SBD} (t) is the time dependent reflux ratio, F_{MeOH} (t) is the methanol feed rate profiles, which are optimized into the optimization problem. Note, the process models for SBD/i-SBD columns are described by many non-linear differential and algebraic equations (DAEs) that act as the inequality constraints to the dynamic optimization problem.

4.2 Maximum Distillate Amount Problem for CBD Column (of i-SBD process)

As the CBD column of the i-SBD system has the same batch time as SBD (described in section 2), the performance of CBD column is evaluated in terms of maximum distillate amount of methanol recycled for a given final operating time and purity of recycle methanol and the total energy consumption is calculated.

Given:	The CBD column configuration, the feed composition, the condenser vapour load, purity of the distillate product, fixed batch time.

Optimize: The reflux ratio (R_{CBD})

So as to: Maximize the distillate amount of product (D_{MeOH})

Subject to: Process constraints, Model equations

Mathematically, the optimization problem (OP2) can be represented as follows:

OP2 Max D_{MeOH}

$R_{CBD}(t)$ (4)

Subject to :

$t_F = t_F^*$ (Inequality Constraints)

$x_{MeOH} \geq x_{MeOH}^*$ (Inequality Constraints)

Linear bound on R_{CBD} (t) (Inequality constraints)

Where, D_{MeOH} represents the quantity of the distillate methanol, R_{CBD} (t) is the reflux ratio as a function of time (t), x_{MeOH} represents the composition of recycled methanol at the final batch time (t_F), (t_F^*, x_{MeOH}^*) are the specified the end operating time and the specified purity of methanol recycle (which is recycled to SBD of the next batch). Note, the process models of i-SBD column are described by a set of highly non-linear differential and algebraic equations (DAEs), which perform as equality constraints to the optimization layout. Also, the operational constraints approach in terms of the overflowing of still pot for the i-SBD system is same as that was suggested by Aqar et al. (2016 b).

5. Results and Discussions

5.1 The performance of semi-batch distillation column (SBD)

The production of methyl decanoate (MeDC) (the main product) is carried out in 10 a theoretical plates-batch column involving (total condenser and still) with a 2.5 kmol/hr of the condenser vapour load and a 5 kmol of fresh feed charged in the still pot with the following concentration in molefraction: 0.5 decanoic acid, 0.5 methanol, 0.0 methyl decanoate, and 0.0 water. The total number of trays for the batch distillation columns are computed from top-down, tray 1 being the condenser and tray N the reboiler. Four percent of initial feed amount is the total column holdup, which 50% of it is placed in the total condenser and the rest is distributed on all trays (equally shared). Note that, the total number of plates refers to the number of theoretical or ideal trays. The pressure drop of the column can be fixed, which results in approximately fixed vapour load to the condenser, and hence constant condenser load is considered here. This assumption is made in by several works in the literature (Furlonge, et al., 1999; Low and Sørensen, 2002) In this work the top column pressure is 100 kpa (1 bar) with a pressure drop of 0.2 kpa per stage.

The concentrations of total condenser and column stages are initialized to the feed composition at the beginning of process. In the start-up procedure, the distillation column runs in total reflux mode for some time until the column reaches the steady state and then concentration profiles of column are consequently established.

After that, the production procedure for all cases begins from this point (designated as t = 0) onward. Three case studies are investigated here, composing of one-reflux interval, two-reflux intervals and three-reflux intervals operations. The purity of methanol charged to the reboiler drum is 0.95 molefraction. The quality of MeDC product in the still drum is changed from (0.915 to 0.945 molefraction) as considered earlier by Aqar et al. (2017a) but for different column configurations, while the amount of product in the still is kept constant at 2.5 kmol for all cases presented below.

5.1.1 Case 1: Optimal Operation using Single Interval Control (NCI=1)

The operation results (including optimal feed rate, optimal reflux ratio, maximum reflux ratio, total amount of feed, final batch time, and total number of batches, as well as minimum energy demand) and total annual cost details (in terms of capital annual and operating costs as well the total annualized cost) for different product purity constraints for the SBD process using single reflux control interval are summarized in Table 1. As it can be noted, the optimal feed rate of methanol, the final production time with the total energy consumption rate, and the total quantity of methanol fed as well as the total annualized cost gradually increases with increasing MeDC purity specification (see Equation (18)).

As expected, the increase in batch time decreases the number of batches according to Equation (20). Note, the column and trays costs reduce progressively as the diameter of column is decreased upon optimization. In addition, as can be seen, as the total amount of methanol fed over the batch time increases resulted in increase in the cost of methanol charge, which has a significant impact on the total operating cost and the overall annual cost as well.

It is obvious from Table 1 that the higher amount of methanol charge, and the higher operating time requires the higher energy demand at 0.945 molefraction of MeDC concentration compared to others to avoid moving of MeDC up to the distillate receiver and to satisfy the bottom product composition specification. Also, it can be noticed that the total annualized cost of the SBD system is increased (by almost 12.59%) from 45141 to 51645 ($/yr). This the reflection of the increase in the product quality from 0.915 to 0.945.

Note, for all the MeDC composition cases, the maximum reflux ratio (R_{Max}) is larger than the optimal values of actual reflux ratio avoiding the overload of the still pot tank. Also note, in all cases R_{max} is calculated for different values of feed rate of MeOH as shown below.

$$R_{Max} = \left(1 - \frac{F_{MeOH}}{V_C}\right) \tag{5}$$

The mixture profiles in the collector tank and reboiler drum are illustrated in Figures 2, and 3 for the MeDC composition (x^*_{MeDC}= 0.945 molefraction) for single-control operation.

Table 1. Optimal Operation and Design for Total Annual Cost Details for Different Product Purities for SBD using one interval control (NCI = 1).

Product Purity (x^*_{MeDC})	0.915	0.925	0.935	0.945
Optimal MeOH Rate, F_{MeOH} (kmol/hr)	1.62	1.64	1.85	1.88
Optimal Reflux Ratio, R_{SBD}	0.275	0.272	0.196	0.189
Maximum Reflux Ratio, R_{Max}	0.351	0.345	0.261	0.248
Total Amount Fed, F_{tot}	19.56	20.66	26.19	29.58
Final Batch Time, t_F (hr)	12.06	12.62	14.17	15.73
Number of Batches (batch/yr)	649	621	555	502
Energy Demand (MJ)	1.122	1.172	1.310	1.448
Column Diameter, D_C (m)	0.155	0.152	0.149	0.149
Column Cost ($)	16146	16056	15720	15649
Plate Cost ($)	741	735	712	708
Condenser Cost ($)	148	151	160	162
Reboiler Cost ($)	146	146	146	145
Capital Cost ($/yr)	**3436**	**3417**	**3348**	**3333**
Energy Cost ($/yr)	2585	2580	2566	2557
MeOH Charge Cost ($/yr)	39029	39466	44723	45665
Cooling Water Cost ($/yr)	84	84	84	84
Operating Cost ($/yr)	**41705**	**42137**	**47380**	**48312**
Total Annual Cost ($/yr)	**45141**	**45555**	**50727**	**51645**

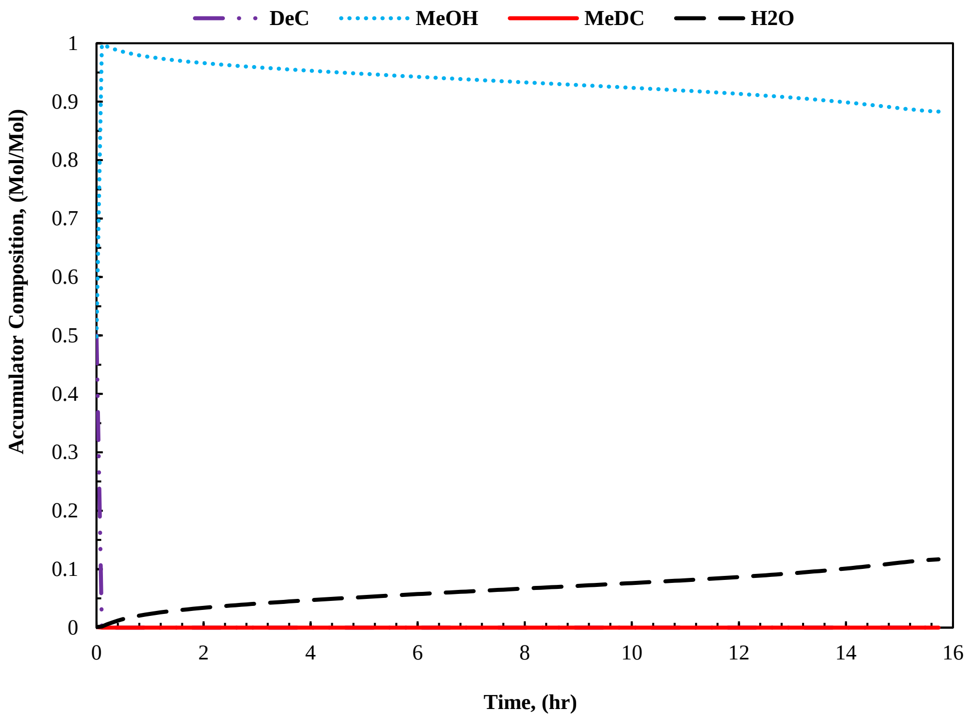

Figure 2: The accumulator composition profile for SBD, Single-Interval Control (x^*_{MeDC}= 0.945).

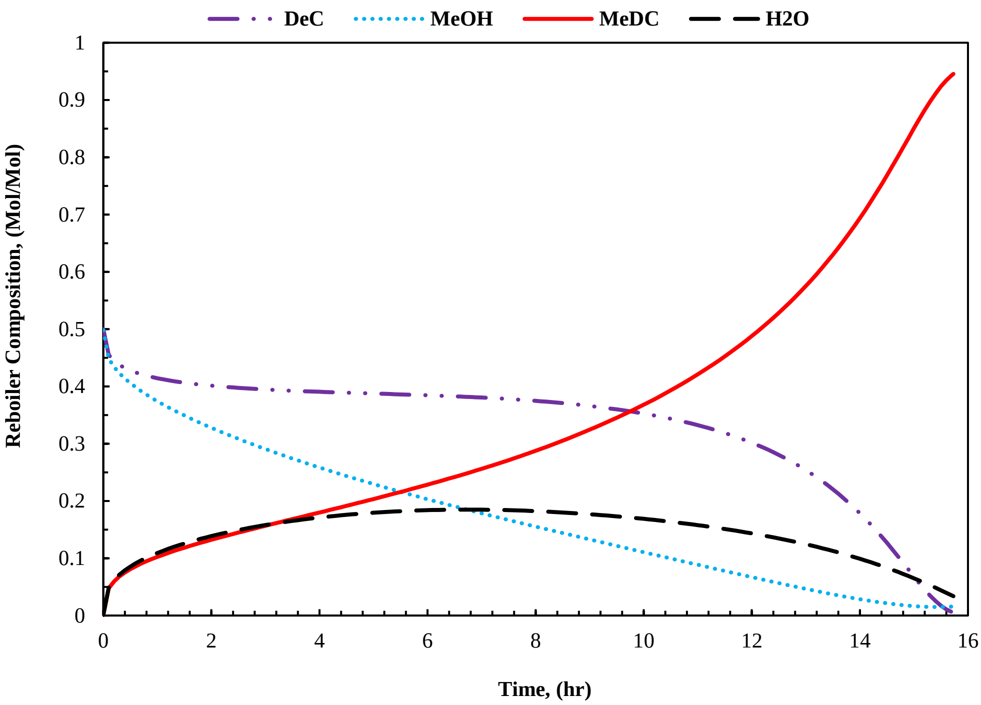

Figure 3: The reboiler composition profile for SBD, Single Interval Control (x^*_{MeDC} = 0.945).

5.1.2 Case 2: Optimal Operation using Two Intervals Control (NCI=2)

To accomplish the desired product concentration, multi- interval control strategy leads to offer better performance (in terms of operating batch time and energy consumption rate) than single-interval control strategy. This is because varied reflux ratio based multi-control interval operation gets updated regularly, where the single-interval control one works with a fixed reflux ratio (Aqar et al. 2019). Table 2 shows the optimizations results in terms of minimum batch time, the optimal methanol feed rate, and reflux ratio for each operating time, total amount fed, optimal switching intervals, final production time, total number of batches, the total energy consumption, the diameter of batch column, capital and utility costs, and total annual cost using two reflux ratio intervals to achieve the bottom product purity (Case 2). It was found that increasing the total amount of methanol charge leads to an increase in the methanol charge cost and thus the annual operating cost. Thus, increasing the total fed quantity can increase the total annual cost of SBD process. Increasing energy consumption also increases the overall annual cost. It is obvious from Table 2 that the use of two-control strategy resulted considerable savings in the batch time and energy required, and the total annualized cost compared to the single-control operation. For all MeDC purities, the total annual cost (TAC) for the two-control strategy is much better than that of single-control strategy (Table 1). This visibly indicates the benefit of employing two-reflux policy. It is observed from Table 2 that significant reductions in the processing batch time (by about 67.79%), the total energy consumption (by almost 64.99%), and the methanol charge cost (by about 19.35%), as well as the total investment cost (by almost 16.87%) using two-reflux intervals as compared to single-reflux interval SBD column (Case 1). It can be seen from Table 2 that at the lower MeDC purity constraint, the SBD process operates with higher reflux ratio for the first-time interval to push water up to the distillate receiver and then runs at lower reflux ratio in the second-time interval to keep both reactant species (DeC and MeOH) in the reaction region to allow more chemical reaction to fulfil the specified quality requirement. Whilst, at higher MeDC concentration, the distillation column runs at lower reflux ratio in the first time and running at higher reflux ratio in the second time. For all cases, the optimal values of actual reflux ratios (R_1 and R_2) are found to be lower than the values of R_{Max} making sure there is no overloading of reboiler drum. Figures 4 and 5 present the concentration profiles in the accumulator tank and still pot at the MeDC concentration specification (x^*_{MeDC} = 0.945) for the two-control process. The methyl decanoate (MeDC) attained the desired product concentration lower operating time for the two-control operation than one-control policy (Figure 3).

Table 2. Optimal Operation and Design for Total Annual Cost Details for Different Product Purities for SBD using two intervals control (NCI = 2).

Product Purity (x^{*}_{MeDC})	0.915	0.925	0.935	0.945
Optimal Feed Rates F_1, F_2 (kmol/hr)	0, 2.01	1.08, 1.55	1.62, 1.48	1.62, 1.61
Optimal Reflux Ratios R_1, R_2	0.482, 0.180	0.202, 0.329	0.077, 0.395	0.114, 0.329
Total Amount Fed, F_{tot}	5.06	5.97	7.18	8.18
Time Intervals, t_1, t_2 (hr)	1.70, 2.51	2.18, 2.34	3.28, 1.26	3.69, 1.38
Final Batch Time, t_F (hr)	4.22	4.52	4.54	5.08
Number of Batches (batch/yr)	1729	1624	1617	1464
Energy Demand (MJ)	0.434	0.460	0.463	0.507
Column Diameter, D_C (m)	0.149	0.152	0.143	0.146
Column Cost ($)	15525	15821	14952	15177
Plate Cost ($)	699	719	662	677
Condenser Cost ($)	168	150	150	157
Reboiler Cost ($)	156	155	155	153
Capital Cost ($/yr)	**3310**	**3369**	**3184**	**3237**
Energy Cost ($/yr)	2862	2826	2830	2779
MeOH Charge Cost ($/yr)	26867	29809	35682	36829
Cooling Water Cost ($/yr)	87	87	87	86
Operating Cost ($/yr)	**29822**	**32729**	**38606**	**39700**
Total Annual Cost ($/yr)	**33132**	**36098**	**41790**	**42933**

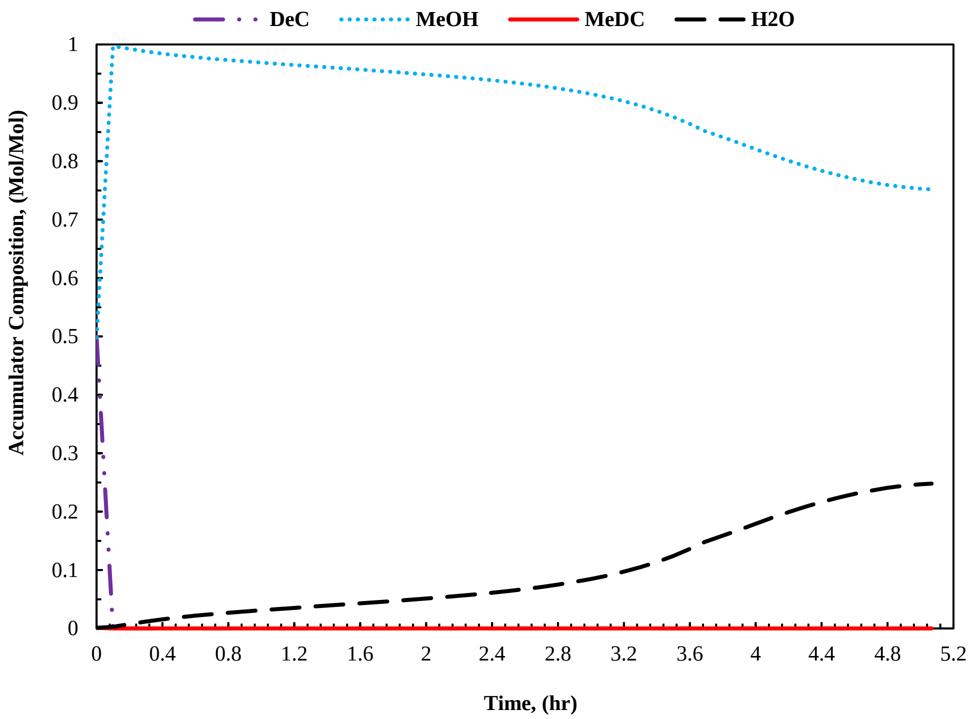

Figure 4: The accumulator composition profile for SBD, Two Intervals Control (x^*_{MeDC} = 0.945).

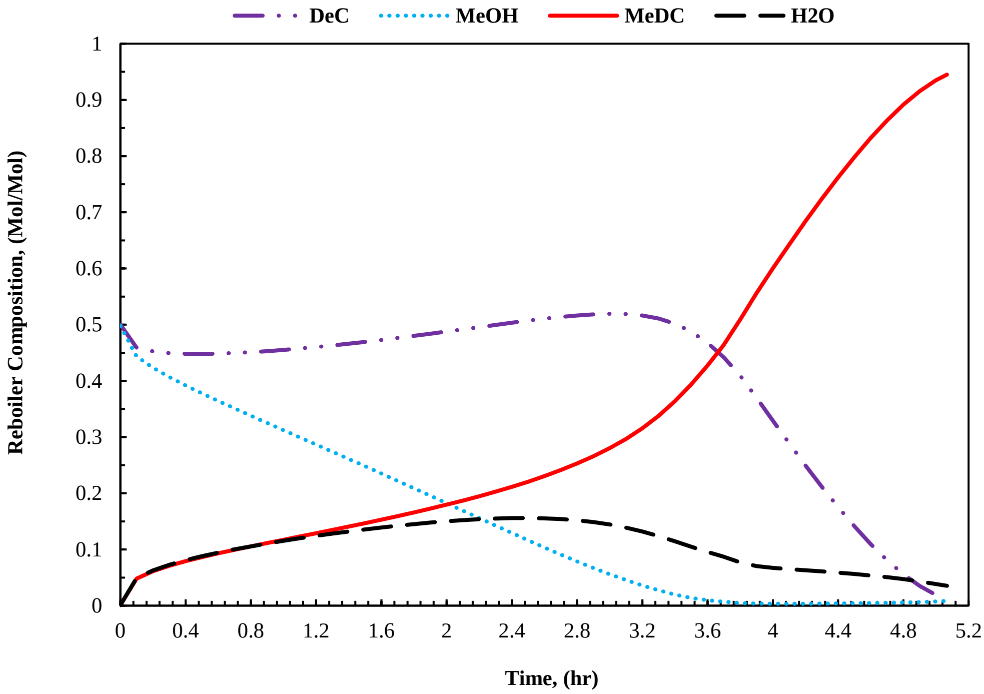

Figure 5: The reboiler composition profile for SBD, Two Intervals Control (x^*_{MeDC} = 0.945).

5.1.3 Case 3: Optimal Operation using Three Intervals Control (NCI= 3)

For the four compositions considered, the optimal operation results (such as the methanol feed rates, and the reflux ratios for three time intervals, the amount of methanol charge, the time intervals, the final production time, the total number of batches, and the minimum energy required) and the corresponding cost details (the parameters of total capital investment, and total operational costs, as well as total annual cost) for 3- intervals control policy are listed in Table 3.

As mentioned before, the optimization results in Table 3 clearly indicate that as the composition of MeDC increases from 0.915 to 0.945 molefraction, all the batch time, the total energy consumption, the total amount of MeOH fed, the cost of methanol charge, and the operational cost increase gradually together with the overall annualized cost (TAC). Note also, it is found from Table 3 that the use of three-intervals control operation can significantly improve the process efficiency and secure potential minimizations in batch time thermal energy consumption rate, and methanol charge price, as well as the total annualized cost (TAC%) compared to the use of two-control SBD operation to achieve the desired product purity constraints.

For example, the overall operating time reduction of 46.45% and the thermal heat expense reduction of 39.41%, and the cost of methanol charge saving of 41.18%, and the minimum annual cost of SBD mode reduction of 34.62% at MeDC composition of 0.945 molar-mass as compared to those obtained by using the two-reflux operation (Case 2). The saving in operating time and energy demand cannot only result a more effective and economical operation but also can produce a cleaner atmosphere by reducing the flue gas emissions.

It can be noticed from Table 3 that the SBD column operates at zero reflux ratio and zero methanol feed rate for the first-time interval for each MeDC composition condition to drive the water (one of the reaction product) up to the distillate tank and to shift the chemical reaction to the right in the reboiler. Whilst, higher reflux ratio and higher feed rate of methanol are needed to retain both chemical reactants (DeC and MeOH) in the reactive zone to allow further reaction and to accomplish the specified MeDC purity requirement in both second, and third time intervals. Note also, for all MeDC composition requirements, the cost of catalyst loading in the SBD operation remained the same (6.48 $/yr) for all cases presented earlier.

Table 3. Optimal Operation and Design for Total Annual Cost Details for Different Product Purities for SBD using three intervals control (NCI = 3).

Product Purity (x^*_{MeDC})	0.915	0.925	0.935	0.945
Optimal Feed Rates F_1, F_2, F_3 (kmol/hr)	0, 1.55, 1.30	0, 1.59, 1.45	0, 1.59, 1.48	0, 1.48, 1.48
Optimal Reflux Ratios R_1, R_2, R_3	0, 0.368, 0.460	0, 0.351, 0.409	0, 0.319, 0.399	0, 0.247, 0.405
Total Amount Fed, F_{tot}	2.48	2.55	2.62	2.78
Time Intervals, t_1, t_2, t_3 (hr)	0.90, 1.50, 0.12	0.90, 1.06, 0.60	0.89, 0.60, 1.13	0.83, 0.50, 1.38
Final Batch Time, t_F (hr)	2.52	2.55	2.61	2.71
Number of Batches (batch/yr)	2702	2669	2622	2536
Energy Demand (MJ)	0.288	0.292	0.297	0.307
Column Diameter, D_C (m)	0.146	0.146	0.146	0.143
Column Cost ($)	15074	15224	15070	14863
Plate Cost ($)	670	680	670	657
Condenser Cost ($)	145	148	153	157
Reboiler Cost ($)	168	167	167	166
Capital Cost ($/yr)	**3211**	**3244**	**3212**	**3169**
Energy Cost ($/yr)	3183	3176	3166	3144
MeOH Charge Cost ($/yr)	20597	20892	21142	21662
Cooling Water Cost ($/yr)	90	90	90	90
Operating Cost ($/yr)	**23876**	**24164**	**24405**	**24903**
Total Annual Cost ($/yr)	**27088**	**27408**	**27617**	**28071**

It can be observed from all cases presented in Tables 1, 2, and 3 that although multiple-intervals control strategies are considered in the SBD operation, the values of total yearly cost are still more higher due to an increase in the cost of total quantity of charged methanol and this makes SBD uncompetitive operation in all cases and hence the proposed i-SBD operation. The mixture profiles in the accumulator tank and the reboiler drum of three-control SBD operation at product purity requirement (x^*_{MeDC}= 0.945) are displayed in Figure 6 and 7. Also, as can be seen from Figure 7 that the MeDC reached the desired product purity at the lower batch time for three-reflux strategy than the single and the two-reflux SBD cases (Figures 3 and 5).

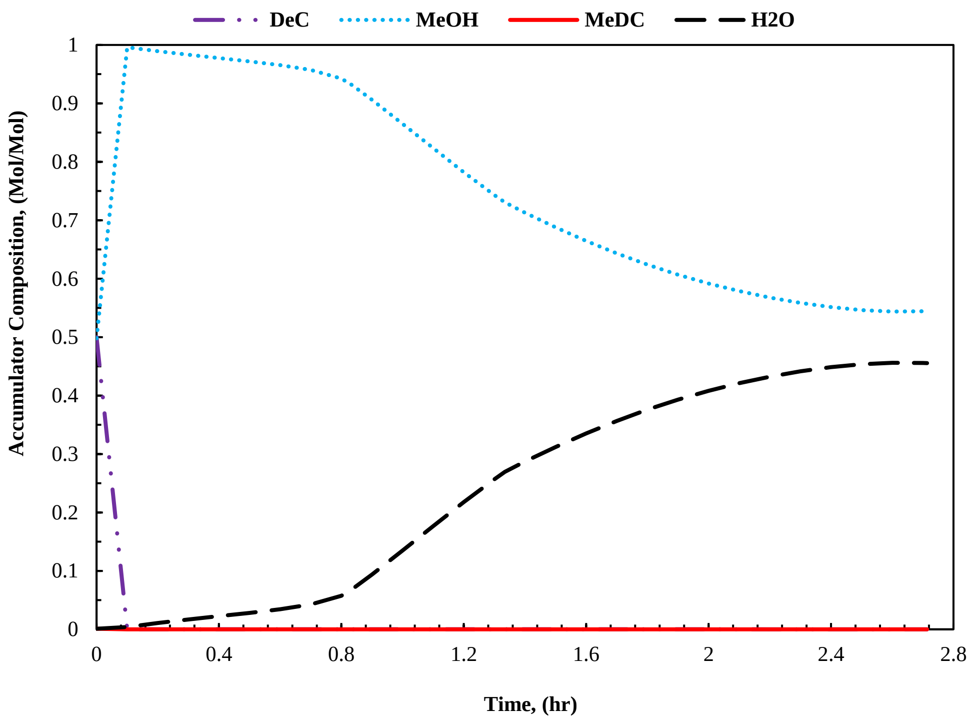

Figure 6: The accumulator composition profile for SBD, Three Intervals Control (x^*_{MeDC} = 0.945).

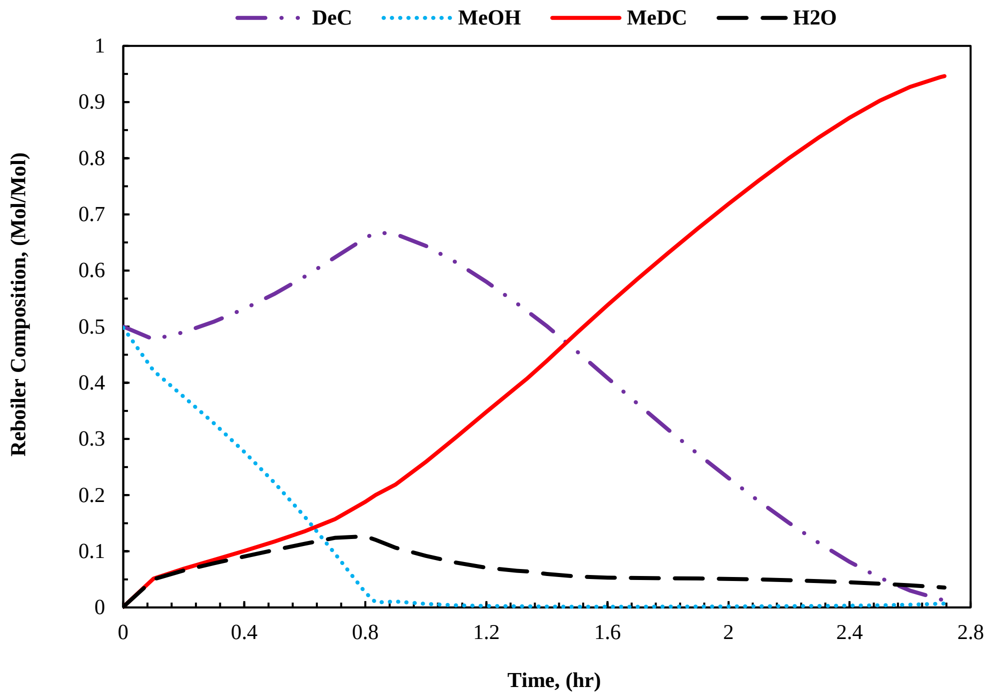

Figure 7: The reboiler composition profile for SBD, Three Intervals Control (x^{*}_{MeDC} = 0.945).

5.2 The performance of i-SBD Operation

The novel integrated semi-batch and a conventional batch distillation process (i-SBD), where methanol is fed continuously to the still, while the conventional batch distillation column recovers methanol from the distillate of the first column, which is recently proposed by Aqar et al. (2016b) but tested for different chemical reaction systems, is utilized in this current work for the production of MeDC as shown in Figure 1. The operational and economic feasibility of the i-SBD process as a potential option for the synthesis of MeDC is examined. The first (SBD) column is used to synthesize methyl decanoate and the second (CBD) column is to recover methanol from the accumulator tank of SBD at 0.95 molefraction and which is fed back into the SBD column together with make-up methanol at same purity of methanol (0.95 molefraction). Note, the column configuration and input data for both columns of i-SBD process are similar to those for SBD process except that the total number of trays (including total condenser and still pot) and the condenser vapour load in the CBD column are 6-plates and 1.5 kmol/hr respectively. Three scenarios are considered here, one (Scenario 1) with one-interval control and the second one (Scenario 2) with two-intervals control and the other (Scenario 3) with three-intervals control policy of operation. As mentioned before, the purity of product (MeDC) is changed from 0.915 to 0.945 molefraction in each scenario with the bottom product amount being set at 2.5 kmol so that comparison of performances of i-SBD operation can be made with SBD in terms of minimum yearly cost.

5.2.1 Scenario 1: Optimal Operation using Single-Interval Control (NCI =1)

The optimization results of i-SBD system are similar to those reported in Table 1. However, the parts of these results are presented in Table 4 together with those achieved for CBD column for convenience. In addition, as the quantity of methanol fed to the CBD increases, the reflux ratio of CBD operation (R_{CBD}) decreases expectedly due to the easy separation of methanol from water. Since the amount of methanol charged to SBD increases, the cost of makeup methanol also increases together with the total operating cost for the i-SBD process.

The results in Table 4 obviously indicate that the using of i-SBD operation is considerably more beneficial than SBD column in terms of using single-reflux strategy (Table 1). As an example, for the scenario with 0.945 molefraction of MeDC the i-SBD system provided about a total annualized cost saving of 58.52% compared to that of the SBD counterpart.

Table 4. Optimal Operation and Design for Total Annual Cost Details for Different Product Purities for i-SBD using one interval control (NCI = 1).

Product Purity (x^*_{MeDC})	0.915	0.925	0.935	0.945
	Semi batch	**Distillation**	**Column**	
Optimal MeOH Rate, F_{MeOH} (kmol/hr)	1.62	1.64	1.85	1.88
Optimal Reflux Ratio, R_{SBD}	0.275	0.272	0.196	0.189
Column Cost ($)	16146	16056	15720	15649
Plate Cost ($)	741	735	712	708
Condenser Cost ($)	148	151	160	162
Reboiler Cost ($)	146	146	146	145
Capital Cost ($/yr)	**3436**	**3417**	**3348**	**3333**
Energy Cost ($/yr)	2585	2580	2566	2557
Cooling Water Cost ($/yr)	84	84	84	84
Operating Cost ($/yr)	**2676**	**2671**	**2657**	**2647**
	Conventional	**Distillation**	**Column**	
Amount of Feed (kmol)	21.86	22.96	28.49	31.88
Optimal Reflux Ratio R_{CBD}	0.167	0.161	0.111	0.089
Distillate Amount, D_{MeOH} (kmol)	15.06	15.89	18.91	21.49
Column Diameter, D_C (m)	0.113	0.113	0.113	0.113
Column Cost, $	6601	6603	6617	6620
Plate Cost, $	226	226	227	227
Condenser Cost, $	120	120	120	120
Reboiler Cost, $	102	102	102	102
Capital Cost ($/yr**)**	**7050**	**7051**	**7066**	**7069**
Energy Cost, $/yr	1488	1488	1487	1487
Cooling Water Cost, $/yr	50	50	50	50
Operating Cost ($/yr)	**1538**	**1538**	**1537**	**1537**
	Integrated	**Semibatch**	**Column**	
Final Batch Time, t_F (hr)	12.06	12.62	14.17	15.73
Number of Batches (batch/yr)	649	621	555	502
Make-Up Cost, Makeup ($/yr)	8969	9122	12433	12489
Total Capital Cost ($/yr**)**	**4846**	**4828**	**4760**	**4747**
Total Operating Cost ($/yr**)**	**13182**	**13330**	**16628**	**16673**
Total Annual Cost, $/yr	**18028**	**18158**	**21388**	**21420**

5.2.2 Scenario 2: Optimal Operation using Two Intervals Control (NCI =2)

The summary of optimization results for two-reflux intervals control policy is illustrated in Table 5, which is similar to Table 4. The trend of the operation results of each column is qualitatively same as to those shown in Table 4. It can be realized form Table 5 that, the cost of makeup methanol achieved is 35.60% at the MeDC quality of 0.915 as compared to that obtained by using the one-control i-SBD operation (Scenario 1). The total annual cost of i-SBD process with two-intervals control is about 16.97% less compared to the single-reflux policy of i-SBD mode due to low amount of methanol makeup charge and operating batch time which are demanded to satisfy the MeDC constraint (0.915 molefraction). While, at higher product purity (0.945 molefraction) there are little reductions in both the cost of methanol makeup and cost of TAC by almost 1.78% and 0.49%, respectively compared to the one-reflux ratio i-SBD operation.

Table 5. Optimal Operation and Design for Total Annual Cost Details for Different Product Purities for i-SBD using two intervals control (NCI = 2).

Product Purity (x^{*}_{MeDC})	0.915	0.925	0.935	0.945
	Semibatch	**Distillation**	**Column**	
Optimal Feed Rates F_1, F_2 (kmol/hr)	0, 2.01	1.08, 1.55	1.62, 1.48	1.62, 1.61
Optimal Reflux Ratios R_1, R_2	0.482, 0.180	0.202, 0.329	0.077, 0.395	0.114, 0.329
Time Intervals, t_1, t_2 (hr)	1.70, 2.51	2.18, 2.34	3.28, 1.26	3.69, 1.38
Column Cost ($)	15525	15821	14952	15177
Plate Cost ($)	699	719	662	677
Condenser Cost ($)	168	150	150	157
Reboiler Cost ($)	156	155	155	153
Capital Cost ($/yr)	**3310**	**3369**	**3184**	**3237**
Energy Cost ($/yr)	2862	2826	2830	2779
Cooling Water Cost ($/yr)	87	87	87	86
Operating Cost ($/yr)	**2956**	**2920**	**2924**	**2872**
	MeOH	**Separation**	**Column**	
Amount of Feed (kmol)	7.35	8.27	9.48	10.48
Optimal Reflux Ratio R_{CBD}	0.372	0.334	0.295	0.282
Distillate Amount, D_{MeOH} (kmol)	3.97	4.52	4.80	5.46
Column Diameter, D_C (m)	0.110	0.113	0.113	0.113
Column Cost, $	6508	6543	6573	6578
Plate Cost, $	222	223	225	225
Condenser Cost, $	120	120	120	120
Reboiler Cost, $	102	102	102	102
Capital Cost ($/yr)	**6951**	**6988**	**7020**	**7025**
Energy Cost, $/yr	1489	1489	1489	1489
Cooling Water Cost, $/yr	50	50	50	50
Operating Cost ($/yr)	**1539**	**1539**	**1539**	**1539**
	Integrated	**Semibatch**	**Column**	
Final Batch Time, t_F (hr)	4.22	4.52	4.54	5.08
Number of Batches (batch/yr)	1729	1624	1617	1464
Make-Up Cost, Makeup ($/yr)	5775	7267	11804	12266
Total Capital Cost ($/yr)	**4700**	**4767**	**4588**	**4638**
Total Operating Cost ($/yr)	**10270**	**11725**	**16266**	**16677**
Total Annual Cost, $/yr	**14969**	**16491**	**20854**	**21315**

5.2.3 Scenario 3: Optimal Operation using Three Intervals Control (NCI = 3)

As before, similar to Table 5, Table 6 shows the operation results for three-reflux ratio strategy. Qualitatively, the optimization results of each distillation column have the same trend to those displayed in Table 5. Compared with two intervals control i-SBD system (Table 5), the cost of methanol makeup quantity is significantly cut down by almost 65.32% for MeDC purity of 0.945 molefraction. It is obvious form Table 6 that the three control i-SBD operation caused a decrease in the total yearly cost compared to the two reflux intervals. For 0.945 molefraction concentration the reduction in the minimum annualized cost is about 36.30% compared to two reflux ratio strategy (Scenario 2). Visibly the three-control i-SBD operation (Scenario 3) provides much better operational flexibility and economic performance in terms of minimum annual cost. Note that, although in the present study only one, two, and three-intervals policy are used here, more than three-reflux intervals can be taken into consideration and optimization techniques can be employed (Mujtaba, 2004) to determine appropriate control intervals and further decreases in the batch time and energy demand, as well as the annual cost are expected in such cases. However, incremental benefit for going beyond 3 control intervals will not be high.

Table 6. Optimal Operation and Design for Total Annual Cost Details for Different Product Purities for i-SBD using three intervals control (NCI = 3).

Product Purity (x^*_{MeDC})	0.915	0.925	0.935	0.945
	Semibatch	**Distillation**	**Column**	
Optimal Feed Rates F_1, F_2, F_3 (kmol/hr)	0, 1.55, 1.30	0, 1.59, 1.45	0, 1.59, 1.48	0, 1.48, 1.48
Optimal Reflux Ratios R_1, R_2, R_3	0, 0.368, 0.460	0, 0.351, 0.409	0, 0.319, 0.399	0, 0.247, 0.405
Time Intervals, t_1, t_2, t_3 (hr)	0.90, 1.50, 0.12	0.90, 1.06, 0.60	0.89, 0.60, 1.13	0.83, 0.50, 1.38
Column Cost ($)	15074	15224	15070	14863
Plate Cost ($)	670	680	670	657
Condenser Cost ($)	145	148	153	157
Reboiler Cost ($)	168	167	167	166
Capital Cost ($/yr**)**	**3211**	**3244**	**3212**	**3169**
Energy Cost ($/yr)	3183	3176	3166	3144
Cooling Water Cost ($/yr)	90	90	90	90
Operating Cost ($/yr)	**3280**	**3273**	**3262**	**3240**
	MeOH	**Separation**	**Column**	
Amount of Feed (kmol)	4.78	4.85	4.92	5.08
Optimal Reflux Ratio R_{CBD}	0.452	0.453	0.454	0.452
Distillate Amount, D_{MeOH} (kmol)	2.07	2.10	2.13	2.23
Column Diameter, D_C (m)	0.110	0.110	0.110	0.110
Column Cost, $	6449	6447	6445	6446
Plate Cost, $	219	219	219	219
Condenser Cost, $	119	119	119	119
Reboiler Cost, $	102	102	102	102
Capital Cost ($/yr**)**	**6889**	**6887**	**6885**	**6886**
Energy Cost, $/yr	1487	1487	1487	1487
Cooling Water Cost, $/yr	50	50	50	50
Operating Cost ($/yr)	**1537**	**1537**	**1537**	**1537**
	Integrated	**Semibatch**	**Column**	
Final Batch Time, t_F (hr)	2.52	2.55	2.61	2.71
Number of Batches (batch/yr)	2702	2669	2622	2536
Make-Up Cost, Makeup ($/yr)	3400	3699	3935	4254
Total Capital Cost ($/yr**)**	**4589**	**4621**	**4589**	**4546**
Total Operating Cost ($/yr**)**	**8216**	**8508**	**8734**	**9032**
Total Annual Cost, $/yr	**12805**	**13130**	**13323**	**13577**

6. Conclusions

The synthesis of methyl decanoate (MeDC) via the esterification reaction of decanoic acid and methanol in a batch reactive column is a very challenge and cost intensive operation due to the high complicated thermodynamic behaviour within the reaction scheme. To face these limitations of equilibrium reaction, two alternatives of batch reactive distillation columns are examined here: (1) SBD and (2) i-SBD operation.

The economic performances of those operation configurations are evaluated by minimizing the total annualized cost (TAC $/yr) under one and multiple-intervals control polices. Detailed dynamic models for these columns are built utilizing gPROMS Model Builder 4.2.0 and are embedded within the optimization problem formulation. The dynamic optimization problem is solved for various values of MeDC concentration ranging from 0.915 to 0.945 molefraction. The piecewise constants, including reflux ratio, and methanol feed rate (for both SBD and i-SBD processes) on the production batch time, and the thermal energy expense together with the total yearly cost are estimated. Obviously, the integrated semi-batch distillation mode (i-SBD) is found to outperform the traditional SBD column to meet the bottom product (MeDC) requirements with lower economic annual cost.

Note also, the optimization results for a given separation task demonstrate that the use of multi-intervals control case is more attractive process as compared to the single-interval control case in terms of processing time, and total annual cost reductions in the i-SBD system. As an example, the operating time and TAC cost savings achieved are about 82.75%, and 36.61% at MeDC purity of 0.945 molefraction compared to these obtained by using one-reflux interval strategy.

Nomenclature

a_i	Activity of component i	-
B_{MeDC}	The product amount in the reboiler drum	kmol
CBD	Convectional batch distillation	-
C_{MeOH}	Methanol charge cost	$/kmol
CVP	Control vector parameterisation	-
DAEs	Differential algebraic equations	-
D_{MeOH}	Distillate methanol amount	kmol
F_{MeOH}	Methanol feed rate	kmol/hr
F_1, F_2, F_3	Methanol feed rate in time interval 1, 2, and 3 for SBD	kmol/hr
i-CBD	Integrated conventional batch distillation	-
m_{cat}	The catalyst loading	kg
NCI	Number of intervals control	-
NLP	Nonlinear programming problem	-
OP	Optimisation problem	-
Q_C, Q_R	Condenser duty and reboiler heat duty	kj/hr
R_{CBD}	Reflux ratio for CBD	-

R_{SBD}	Reflux ratio for SBD	-
R_{Max}	Maximum reflux ratio	-
r_A	Reaction rate	kmol/kg.s
SBD	Semi-batch distillation	-
SQP	Successive quadratic programming algorithm	-
t_1, t_2, t_3	Length of interval 1, 2, and 3	hr
t_F	Batch processing time	hr
V_C	Vapour load the condenser	kmol/hr
T	Temperature	^oK

Abbreviations

DeC	Decanoic Acid
H_2O	Water
MeDC	Methyl Decanoate
MeOH	Methanol

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Appendix

TAC calculation

The equipment cost estimation follows the procedure of Douglas (1988) and specific equations of Elliott and Luyben (1996), Su et al. (2013) were employed in this work. A payback period of 5 years is assumed, and a Marshall and Swift (M&S) index of a 1461.3 (2013) is used in the calculation. Materials of construction are stainless steel. The equipment is sized as follows:

1- Compute the reboiler heat transfer (A_R) area (m^2):

$$A_R = \frac{Q_R}{U_R \triangle T_R} \tag{6}$$

Where, the overall heat-transfer coefficient U_R is assumed to be 788.45 W m^{-2} K^{-1} (Cho et al. 2014), and the log mean temperature driving force ΔT_R (K) in the reboiler is assumed to be 20 K. Q_R (kJ/kr) is the reboiler duty.

2- Compute the condenser heat transfer (A_C) area (m^2):

$$A_C = \frac{Q_C}{U_C \triangle T_C} \tag{7}$$

Where, the overall heat-transfer coefficient U_C is assumed to be 473. 07 W m^{-2} K^{-1}(Cho et al. 2014), and

$$\triangle T_C = \frac{(120\text{-}90)}{\ln \frac{(T_C\text{-}90)}{(T_C\text{-}120)}} \tag{8}$$

Where, T_C is the condenser temperature (K), and Q_C (kJ/kr) is the condenser duty.

3- Compute the column (L_C) length (m):

$$L_C = 0.7315 \times N_P \tag{9}$$

Where N_P is number of plates, N_P = 8 for SBD column, and 8 for SBD column, 4 for CBD column of i-SBD policy.

4- Compute the column (D_C) diameter (m):

$$D_C = \sqrt{\frac{1}{900\pi}\left[\frac{MW_V^2}{\rho_V}\right]^{0.5}} \times (V)^{0.5} \tag{10}$$

Where V (kmol/hr) is the maximum vapor flow rate, ρ_V (kg/m^3) is the vapor density in the column, and MW_V [kg/kmol] is the average vapor molecular weight.

5- Compute the column cost ($)

$$\text{Column Cost} = \left(\frac{M\ \&\ S}{280}\right) \times \left(101.9 \times D_C^{1.066} \times L_C^{0.802} \times (2.18 + F_C)\right) \tag{11}$$

Where $F_C = F_m F_p = 3.67 \times 1$

6- Compute the distillation column plate cost ($)

$$\text{Plate Cost} = \left(\frac{\text{M \& S}}{280}\right) \times \left(4.7 \times D_C^{1.55} \times L_C \times F_T\right) \tag{12}$$

Where F_T = 4.5.

7- Compute the reboiler cost ($)

$$\text{Reboiler Cost} = \left(\frac{\text{M \& S}}{280}\right) \times \left(A_R^{0.65} \times (2.29 + F_R)\right) \tag{13}$$

Where $F_R = (F_D + F_P) \times F_m = (1.35 + 0) \times 3.75$

8- Compute the condenser cost ($)

$$\text{Condenser Cost} = \left(\frac{\text{M \& S}}{280}\right) \times \left(A_C^{0.65} \times (2.29 + F_C)\right) \tag{14}$$

Where $F_C = (F_D + F_P) \times F_m = (1 + 0) \times 3.75$

The capital cost, CC ($) is calculated by adding (the column and plate costs, and condenser and reboiler costs).

9- Compute energy cost ($/yr):

$$\text{Energy Cost} = \left(\frac{\$C_S}{453.59\ \text{kg}}\right) \times \left(\frac{Q_R}{\lambda_V}\right) \times (8150\ \text{hr/yr}) \tag{15}$$

Where C_s is the saturated steam price and λ_V is the latent heat of the steam, which depends on the bottom temperature of the column. (We assumed C_s is $2.28 per 1000 lb of high pressure (HP) steam at 260 C with λ_V of 2790 kJ/kg).

10- Compute cooling water cost ($/yr):

$$\text{Cooling Water Cost} = \left(\frac{\$0.03}{3.785\ \text{m}^3}\right) \times \left(\frac{0.001\ \text{m}^3}{\text{kg}}\right) \times \left(\frac{Q_C}{30}\right) \times (8150\ \text{hr/yr}) \tag{16}$$

11- Compute catalyst cost ($/yr) by assuming a catalyst life of 3 months:

$$\text{Catalyst Cost} = m_{Cat}(\text{kg}) \times 7.7162(\$/\text{kg}) \times 4/\text{yr} \tag{17}$$

12- Compute Methanol charge cost ($/yr) for SBD operation:

$$\text{MeOH Charge Cost} = F_{MeOH} \times t_F \times N_B \times C_{MeOH} \times \text{Purity of MeOH} \tag{18}$$

13- Compute Methanol Makeup cost ($/yr) for i-SBD operation:

$$\text{MeOH Makeup Cost} = (F_{MeOH} \times t_F - D_{MeOH}) \times N_B \times C_{MeOH} \times \text{Purity of MeOH} \tag{19}$$

Where the number of batches (Batch/yr) is calculated through the following equation:

$$N_B = \frac{(P_H)}{(t_F + t_s)} \quad (20)$$

Where, N_B (batch/yr) is the total number of batches produced per year, setup time (t_s) = 0.5 hr; Production horizon (P_H) = 8150 hr/yr.

The operating cost, OC ($/yr) is obtained by adding the energy, cooling water, catalyst costs and methanol charge cost (for SBD column) or methanol makeup cost (for i-SBD process).

14- Compute the total annual cost ($/yr):

$$\text{TAC} = \text{OC} + \frac{\text{CC}}{\text{Payback period}} \quad (21)$$