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Understanding the impact of reactive holdup on process intensification in the design of reactive distillation column

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ABSTRACT

The commercial viability of reactive distillation as a front-runner industrial process intensification technique is limited by slow reactions. It is because the diameter established by the vapor-loading method restricts large holdup volumes (or catalyst amount) to accomplish the essential conversion. However, placing a large amount of catalyst on column trays necessitates either a high tray weir height (limited to excessive column pressure drop) or a large column diameter. This study aims to investigate an alternative design approach by increasing the column diameter beyond that required for vapor loading to retain a large holdup volume. Several combinations of tray weir heights and column diameters were studied and demonstrated through case studies for three industrial processes, and their optimal designs have been reported. High catalyst holdup volume enhanced energy efficiency and overcame hydraulic limitations despite requiring large diameter vessels. These design configurations with optimized catalyst holdup also resulted in improved process economics. Fabricating a wide column diameter is a good conservative engineering procedure considering safety aspects and better design control.

1. Introduction

Advancement in industrial chemical processing depends on sustainable development. Beyond profit and quality, the search for an efficient process design should also consider its effect on the environment and society [1]. The realization of sustainable development goals can be aided by the application of process intensification (PI) theory and design practices [2]. Reactive distillation (RD) is an excellent choice as a PI tool for enhancing the performance of the conventional reaction and separation processes [3–5]. It enables simultaneous separation during a chemical reaction enhancing selectivity, improving reactant conversion, reducing energy consumption, and eliminating the requirement for solvents [6,7]. However, its operational applicability is limited, and it is not always beneficial to improve the conventional processes with RD. An important constraint of RD is its less effective application in reactions with low specific rates, limiting its commercial viability. The diameter established via vapor-loading cannot accommodate the necessary reactive holdup volume for slow reactions. The reactive holdup volume (or catalyst amount) is a crucial aspect of RD as it affects component formation through temperature, composition, and residence time [8]. In an

RD process, determining the reactive holdup is a crucial factor, which involves the column diameter, effective tray volume, and liquid level on the tray. The effective area is usually 90% of the cross-sectional area of the tray, while a liquid height larger than 10–15 cm (practically the lower and upper bounds of columns) is undesirable because of hydraulic restrictions (excessive pressure drop) [9].

For enhancing the effectiveness of RD columns, it is imperative to augment the reactive holdup on the trays. In comparison to tray columns, packed columns generally exhibit much lower holdups in the column. Therefore, for homogeneous RD processes, columns with trays are considered more favorable. While for heterogeneous RD processes, the internal hardware of the column depends upon the size of catalyst particles. Due to the presence of intra-particle mass transfer limitations in larger catalyst particles, wire gauze is utilized to prevent flooding limitations. Typically, catalyst envelopes are packed inside the column; however, in certain instances, they may be placed on the trays [10].

The initial diameter established is uncertain as it depends on the vapor velocity that can be determined after satisfying column specifications. An estimated diameter is first used to calculate tray holdup volume, followed by column optimization. Finally, the diameter obtained from tray sizing is compared to the estimated diameter. This

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Nomenclature	
RD	reactive distillation
PI	process intensification
H	height of weir on tray
ID	internal diameter
TAC	total annual cost
V_r	reactive holdup volume
D	diameter of vessel
R_{DC}	downcomer ratio
MA	methyl acetate
LA	lactic acid
MMH	methoxy-methyl heptane
N_{rxn}	number of reactive trays
P	pressure of column
RR	reflux ratio

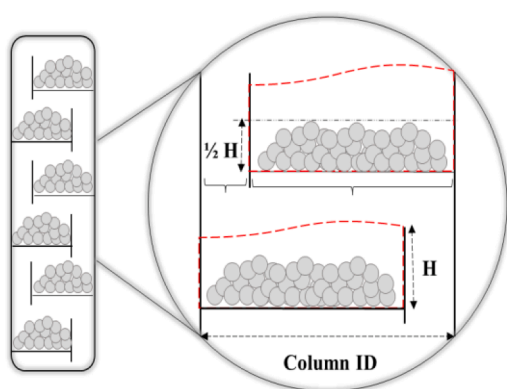


Fig. 1. Association between the column diameter, liquid holdup volume, and weir height.

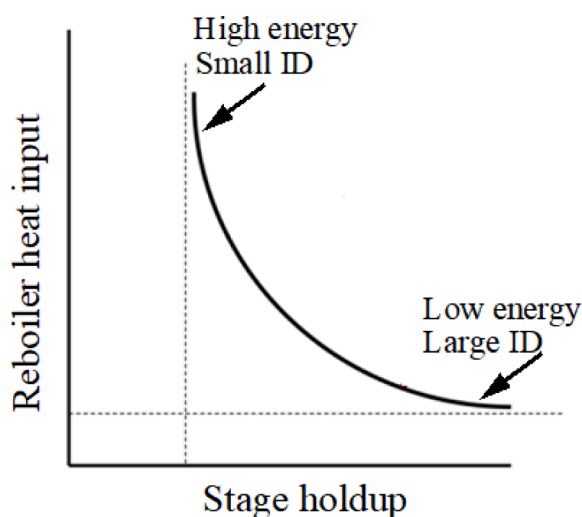


Fig. 2. Reboiler heat load as a function of the catalyst on trays.

procedure is repeated until the difference between the estimated and the computed diameter is below a given tolerance. Specifically, column geometry plays a vital role in the reactive holdup volume selection and cannot be decided arbitrarily. The correlation between the column diameter, catalyst holdup volume, and the weir height on the tray is

depicted in Fig. 1. This procedure restricts the specified amount of catalyst available on individual trays for a particular column diameter. The required tray holdup volume (catalyst) will not be accommodated within the diameter established by vapor loading for slow reactions.

A well-known heuristic while designing a reactive distillation column is that a larger holdup improves reaction performances in terms of conversion, yield, and/or selectivity. The fact is that inadequate catalyst holdup volume reduces the residence time which eventually gives poor conversion. Moreover, adding a large holdup volume (or catalyst amount) on reactive trays extends the residence time of reactions with low specific reaction rates in conventional RD systems. However, a large holdup volume would necessitate either a large diameter column or an elevated height of the liquid on reactive plates.

Utilizing the side-reactor or external reactor concept is one way to solve the incompatibility in the requirements of the high catalyst holdups [11]. This approach involves creating separate reactors that are positioned adjacent to the distillation column. By ensuring that the reaction has sufficient time to occur, the equilibrium conversion can be achieved in the side reactor. However, the side-reactor concept is particularly attractive when the conversion requirements are not stringent [10]. An alternative to accommodate a large reactive holdup volume, resulting in a higher capital cost, is to increase the column diameter beyond that required for vapor loading [12]. Additionally, fabricating a wide-diameter column is prudent engineering considering the safety consequences of the design. In certain cases, a decrement in tray spacing or the installation of additional control devices would increase the number of trays in a column. However, altering the diameter necessitates creating a brand-new vessel [13]. The diameter of separation vessels used in industrial applications ranges from about 0.65 m for small vessels to about 6 m and more for big vessels, and even up to 15 m in some applications [14].

An increase in the catalyst holdup volume on the tray decreases the reflux and vapor boilup rates steadily to achieve specified conversion according to the heuristic approach of designing an RD column [8,15]. A large tray holdup volume results in reduced energy consumption, resulting in reduced greenhouse gas emissions, and improved sustainability. Moreover, a very large increase in reactive holdup improves controllability keeping in view the hydraulic constraints [16]. The traditional plot of the reboiler energy load vs stage holdup has a hyperbolic form with two asymptotes. The first is at the lowest energy consumption with the largest stage holdup volume. The second is at the minimal stage holdup volume with maximum energy consumption. Fig. 2 presents a typical well-known plot of the reboiler heat input versus stage holdup.

By considering a case study of a homogenous RD column, Krishna has demonstrated some of the tray hardware design and sizing challenges [17]. To achieve 98.5% product (methyl acetate) purity in a homogenous RD column, he considers various combinations of tray weir heights and column diameters. He pointed out that target conversion and purity cannot be attained at low liquid hold-up values. For homogenous RD, he suggested operating the tray column at very low superficial vapor velocities in the bubbly flow regime by keeping the weir height at a high value. Additionally, he suggested bubble cap trays, which have a higher liquid holding capacity than sieve trays. For the heterogeneous column, Baur and Krishna investigate an alternative configuration to the RD concept, namely a side-reactor column (SRC) to solve the incompatibility in the requirements of the high catalyst holdups and wider column diameter [18]. They discover that an RD column performs similarly at greater pump-around ratios, which correspond to the five side reactors. The side-reactor configuration column is projected to have a smaller diameter than the RD column based on their comparison of column dimensions. They concluded, nevertheless, that the side-reactor concept is most appealing when the conversion requirements are not strict.

In a different investigation, Luyben simulated two RD columns for sizing calculation [19]. For the case study of ethylene glycol, he uses the

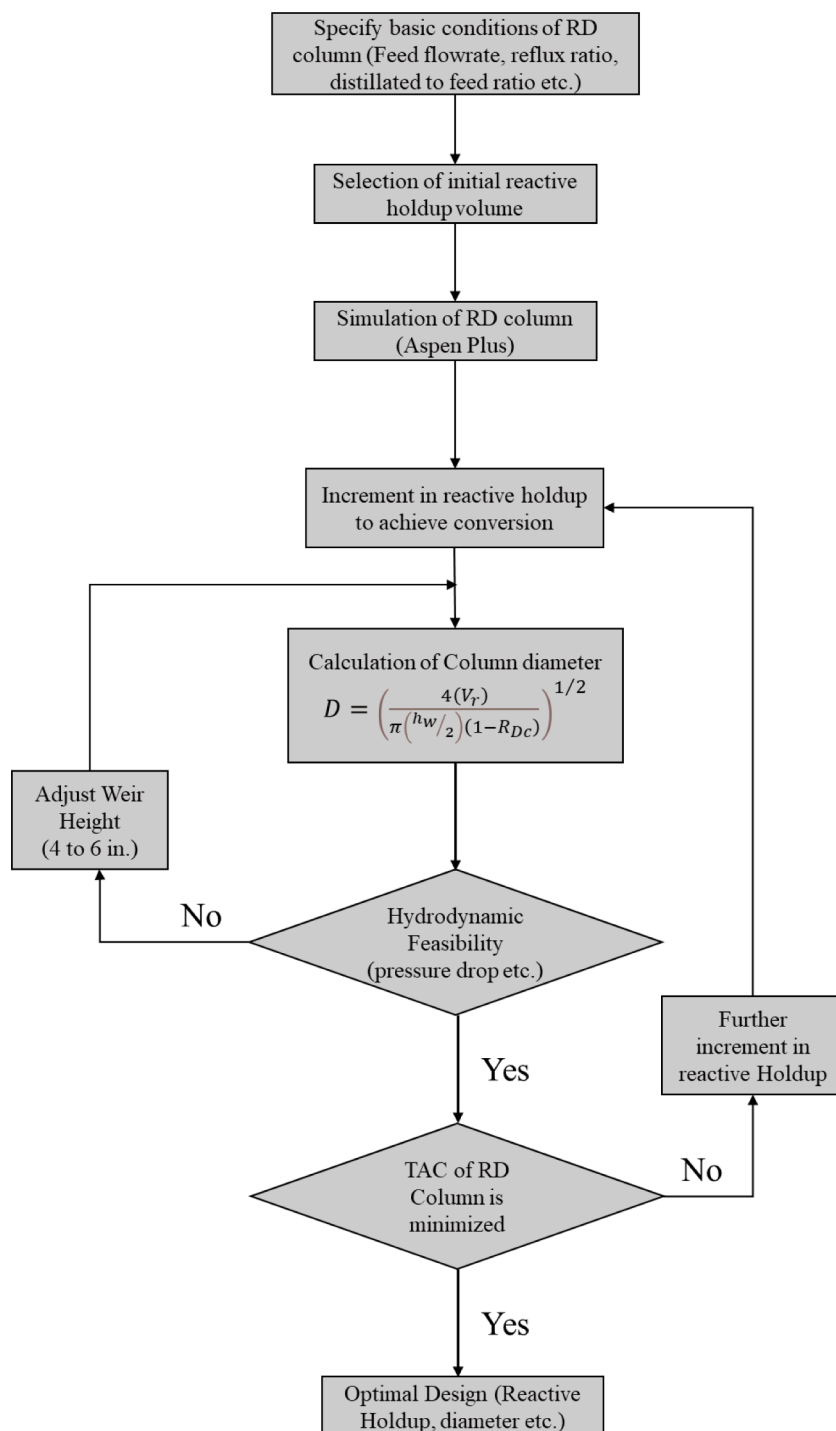


Fig. 3. Methodology for optimization of reactive holdup in RD Column.

upper limit of liquid height (6 inches) and guessed the initial diameter for evaluating the tray holdup using the Eq. (1).

$$Holdup = \left(\frac{\pi(D^2)}{4} \right) (h_w) \quad (1)$$

The diameter determined by the vapor loading is compared with the holdup findings obtained using an assumed column diameter. This sizing procedure is repeated until the diameter established by the maximum vapor velocity closely matches the guessed value. For the methyl acetate synthesis, the column was simulated for a tray holdup of 3-m^3 based on the constraint of the kinetic data. To have a 3-m^3 liquid holdup, the weir

height is estimated to be 0.366 m which corresponds to a large column pressure drop of about 2.8 atmospheres over 42 trays. Luyben suggested an alternative to having this large weir height would be to increase the diameter of the column.

The method for sizing reactive columns for the synthesis of methyl acetate, butyl acetate, and isopropyl acetate was studied by Lee and Hsiao [20]. They eliminated the downcomer section volume and adjusted the catalyst loading to 50% full in the tray for sizing. The required holdup is then determined using the cross-sectional area, $(\pi D^2/4)$, based on the diameter anticipated in the tray sizing of Aspen Plus. They did not examine how diameter fluctuates with various

combinations of weir heights; instead, they only considered the lower range of weir height (0.1016-m) in their sizing analysis.

Accommodating more catalysts on reactive trays necessitates a wider diameter column which reflects column capital cost, even though column energy cost can be reduced. These striking facts demonstrate the importance of a deep process intensification via the selection of a suitable catalyst holdup volume for this alternative design. The authors believe that this is the first study to expand the boundaries of RD operating windows by thoroughly investigating these trade-offs. Several industrial systems that demonstrate the effect of the accommodation of large catalyst holdups on the economics of the RD process are presented in this study.

The goal and novelty of this study are to re-examine the RD design by building a wider column diameter beyond what is established by vapor loading. Operating the RD column at a high holdup volume can improve energy efficiency and overcome hydraulic limitations despite requiring large diameter vessels. Optimizing reactive holdup from an economic perspective ensures the advantages of process intensification.

2. Methodology

This section demonstrates the RD column evaluation process to study the impact of retaining large catalyst holdup volumes on the economic performance of RD systems. Steady-state simulations were performed by varying catalyst amounts on reactive trays for the required conversion and/or yield specification. The impact of increasing reactive holdup volumes on column energy requirement and reactant conversion was investigated, followed by the calculation of the column diameter for different catalyst loading. Several combinations of tray weir heights between 10 and 15 cm (typically representing the lower and upper limits of actual columns) and column diameters were studied and demonstrated through case studies for three industrial RD processes. Their optimal designs were also studied.

The following assumptions were made for column sizing: (1) liquid depths varied from 4 to 6 in, (2) half of a reactive tray was filled with a solid catalyst, (3) 10% of the tray area was occupied by the downcomer section, and (4) the reactive holdup volume per tray is uniform along the reactive section. Using these statements, the column diameter was calculated by applying Eq. (2) to keep the catalyst loading on plates [20].

$$D = \left(\frac{4(V_r)}{\pi(h_w/2)(1 - R_{DC})} \right)^{1/2} \quad (2)$$

where V_r represents reactive/catalyst holdup volume (m^3); D denotes the diameter of the RD column (m); h_w displays the weir height on the tray (m); and R_{DC} is the ratio of the downcomer section of the plate.

Most RD design studies do not consider the hydraulic limitations of columns associated with the reactive holdup volume. Instead, researchers directly specify the holdup volume of the tray or the reaction volume, which is inconsistent with the actual design. The design and optimization method of the reactive holdup volume on a tray is limited by the physical space of the column and is also related to the height of the weir on a tray. Hence, reactive holdup volume cannot be specified arbitrarily. Moreover, meeting the reactive holdup volume requirements to achieve conversion does not guarantee the feasibility of the tower operation (i.e., hydraulic feasibility). So, the aspects of weir height and physical space of the column (diameter) must be considered to obtain an optimal reactive holdup volume.

This study proposes a design methodology for optimizing reactive holdup volume while considering the hydraulic feasibility (weir height on a tray) and diameter of the column rather than fixing the stage holdup volume. The methodology adopted was universal as shown in Fig. 3 and can be implemented in every RD process.

This research methodology employs a comprehensive approach to optimize the reactive holdup volume for the RD column. An initial

reactive holdup is specified along with other initial parameters for the RD column. The RADFRAC model of Aspen Plus is then employed to simulate the RD column, while design specifications are applied to ensure desired product purity. Based on the reactive holdup volume and weir height on a tray (kept within practical range to avoid excessive pressure drop), the diameter of RD column is determined using Eq. (1). The hydrodynamic feasibility of the column is ensured by adjusting the weir height. The iterative process is continued by increasing the reactive holdup volume at regular intervals and monitoring the decrease in the TAC of the column due to a significant reduction in reboiler heat load until the minimum TAC is achieved against the optimal volume of the reactive holdup. The systematic approach exhibits the importance of determining an optimal reactive holdup and provides valuable insights for designing similar RD processes. The methodology diagram is shown below in Fig. 3.

The current study considered RD schemes of methyl acetate (MA), methyl lactate (ML), and methoxy methyl heptane (MMH) processes. Steady-state simulations were performed using the RADFRAC model of Aspen Plus® involving equilibrium-based rigorous calculations. The UNIQUAC thermodynamic model was applied for MA synthesis to define the nonideality of the liquid phase, and deviations from ideal behavior in the vapor mixture were studied using the Hyden-O'Connell equation of state [20]. Binary interaction parameters and reaction kinetics of MA synthesis were taken from Pöpkén et al., 2000 [21]. The UNIQUAC thermodynamic model was used for specifying the nonideality of the liquid phase in the ML process, and deviations from ideal behavior in the vapor phase were studied by applying the Hayden-O'Connell equation of state. The built-in binary interaction parameters of Aspen Plus® were used for simulation. The reaction kinetics for ML synthesis were taken from Krooze et al. [8]. The detailed information is available in supplementary material. The Aspen Plus® built-in UNIQUAC thermodynamic model was used to deal with the nonideality of the liquid phase for MMH synthesis, as recommended [22].

For the quantitative assessment of large catalyst holdup volume impacts on the RD design, the total annual cost (TAC), the well-known economic indicator, was considered. The plant capital expenditure over its payback time and operational expenses were included in the equation for calculating the TAC as stated in Eq. (3).

$$TAC = \left(\frac{\text{Capital cost}}{\text{Payback period}} \right) + \text{Operating cost} \quad (3)$$

The capital investment for the plant incorporated the expenses associated with the column shell, heat exchangers, and reactors, while energy costs corresponded to the heat duty required in the reboiler. A payback period of 3 years was considered. The economic fundamentals and equipment sizing specifications Eqs. (4) and (5) were taken from textbooks by Douglas [23] and Turton [24]. The total number of trays with a spacing of 2 ft with an additional height of 20% was used to determine their height.

$$\text{Column / Reactor capital cost} = 17640(D)^{1.066}(L)^{0.802} \quad (4)$$

For calculating the capital investment of heat transfer equipment (condensers and reboilers) Eq. (4) was utilized. 568 and $852 \text{ Wm}^{-2} \text{ K}^{-1}$ are heat-transfer coefficients used for the reboiler and condenser, respectively.

$$\text{Exchanger capital cost} = 7296(\text{area})^{0.65} \quad (5)$$

The following expenses of cooling and heating utilities typical of US-based chemical plants [25] were used: LP steam (6 bar, 160 °C), MP steam (11 bar, 184 °C), and HP steam (42 bar, 254 °C) were 7.78, 8.22, and 9.88 \$/GJ, respectively.

3. Case studies

Three case studies were investigated to demonstrate the impact of

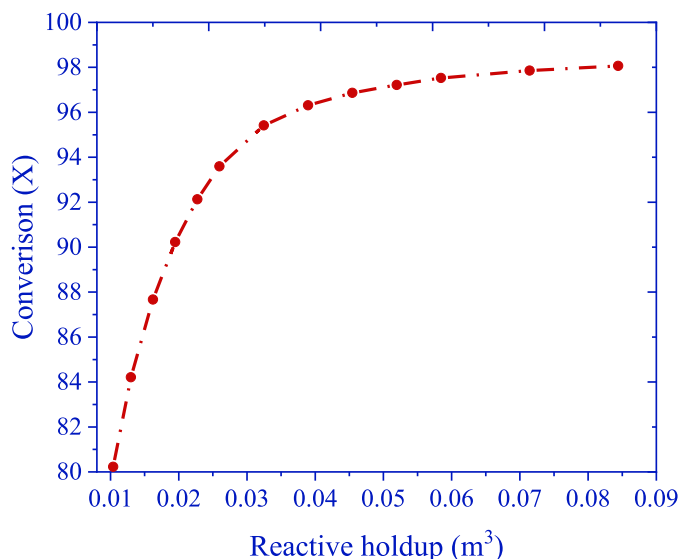
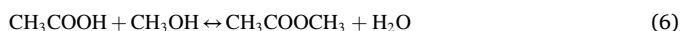


Fig. 4. Reactive holdup volume vs conversion of reactants.

the large holdup volume on RD economics.

3.1. Synthesis of methyl acetate

MA (methyl acetate), a vital raw material in chemical industries, is extensively used to produce coating materials, plasticizers, and fatty acids [26,27]. MA synthesis has a major contribution to the development of the RD process because MA synthesis serves as a model process for RD research and is commercially important. After the successful commercialization of MA production using RD by Eastman Chemical Company, it is now considered a standard model for testing new RD designs developed for chemical equilibrium limited reactions [28]. MA is synthesized by the reaction of acetic acid and methanol in liquid phases in the presence of Amberlyst 15, a solid ion-exchange acid catalyst, as illustrated in Eq. (6).



MA-based RD column analysis conducted by Luyben et al. served as the fundamental reference for the design [29]. The total number of

column stages were 40 including the reboiler as the last stage. There were 34 trays in the reactive section, ranging from the second to the 35th stage. A molar reflux ratio of 1.966 was specified. An equimolar feed containing methanol and acetic acid at flow rates of 100 Kmol hr^{-1} each was introduced at 27th and 4th stage respectively under ambient conditions to the MA-based RD column. A total condenser and kettle-type reboiler were used for simulation, while the method of convergence was strongly nonideal liquid.

3.2. Hydrolysis of methyl lactate

Lactic acid (LA) is a raw organic chemical widely used in pharmaceuticals, food, pesticides, and polymer industries [30]. The conventional production of LA is performed by the bacterial fermentation of biomass, but LA segregation from the fermenter is problematic [31,32]. LA production with high purity and thermal stability is still a challenge because LA self-polymerizes and has a high affinity towards water [33]. An efficient method for producing highly pure LA is esterification with appropriate alcohol, followed by the hydrolysis of the purified ester. ML hydrolysis is performed in a RD column because the spontaneous removal of products can surpass equilibrium limitations [34]. ML hydrolysis requires a large residence time for better conversion, for which some studies have suggested the batch RD process. The ML RD system is selected as conversion can be met by placing large catalyst holdup on trays. Moreover, the conventional processes of ML production involves more capital investment and energy cost in comparison with the RD process [35]. ML hydrolysis catalyzed by a solid acidic ion-exchange resin results in the synthesis of LA, while methanol is formed as a byproduct. ML hydrolysis can be characterized by following the reversible liquid phase reaction as illustrated in Eq. (7).



The reactants of ML hydrolysis are intermediate boiling components, while both products are components with extreme boiling points [36]. RD analysis for hydrolysis of ML performed by Mo et al. was considered as the base case for the design [37]. The total number of column stages were 44 including the reboiler as the last stage. There were 40 trays in the reactive section, ranging from the 5th to the 44th stage. A mass reflux ratio of 5.437 was specified. A feed stream at a rate of 50 Kmol hr^{-1} containing a 90% mass fraction of water and a 10% mass fraction of ML was introduced to the 8th stage of ML-based RD column operating under ambient conditions. A total condenser and kettle-type reboiler

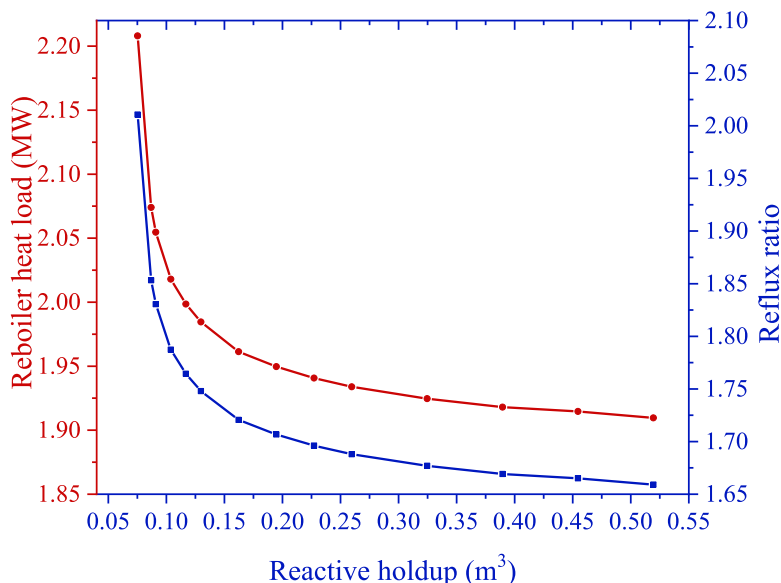


Fig. 5. Influence of reactive holdup on the reflux ratio and reboiler load in the MA-based RD column.

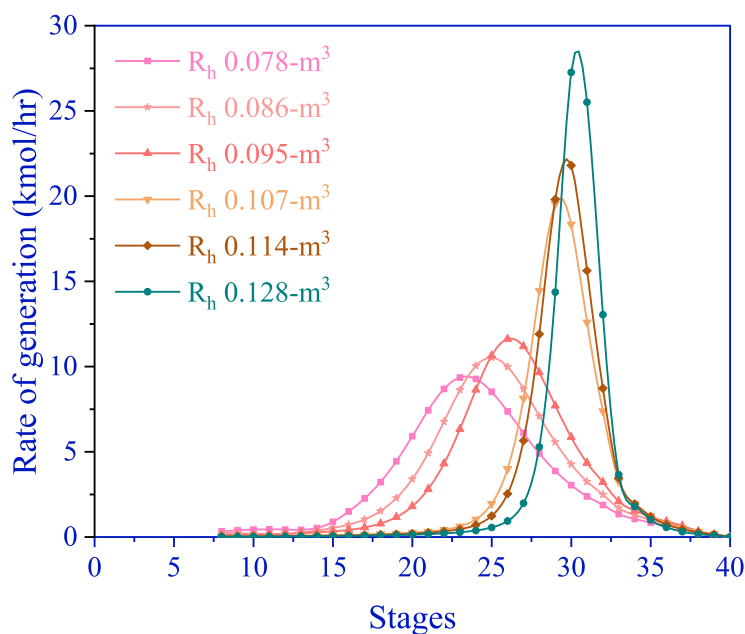


Fig. 6. Effect of the reactive holdup volume on the generation rate of the product in MA-based RD column.

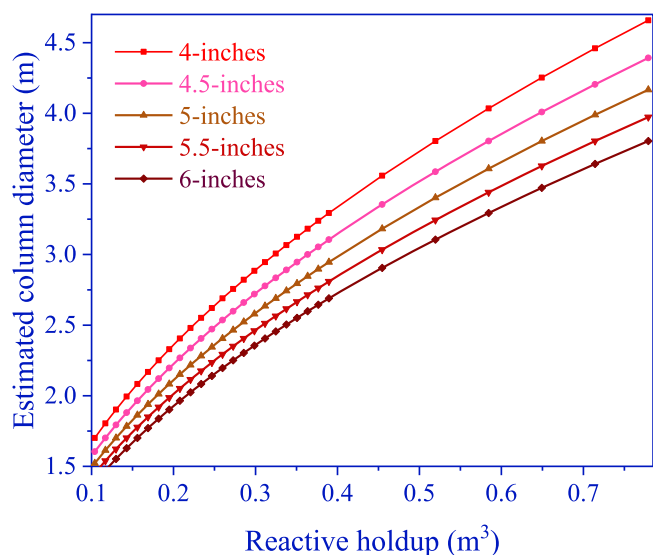


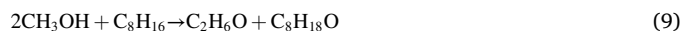
Fig. 7. Impact of the stage holdup volume and the weir height on the diameter of MA RD column.

were used for simulations. The convergence method was strongly nonideal liquid.

3.3. Methoxy methyl heptane synthesis

Methyl tertiary butyl ether (MTBE), a fuel additive, is used to enhance the performance of gasoline [38]. However, the use of MTBE has decreased in recent times based on environmental concerns as it contaminates groundwater [39,40]. Therefore, exploring ethers with high molecular mass and reduced water solubility is an exciting research arena. Methoxy-methylheptane (MMH) has been proposed as an alternative fuel additive with less solubility in water to replace MTBE [22,41, 42]. Although MMH is synthesized using conventional methods, recent advancements reveal that synthesizing it through RD is an effective and economically efficient approach [43]. MMH RD study is picked because of its slow reaction kinetics, as placing large catalyst can overcome

constraint of slow reaction rate. MMH is synthesized by a reversible etherification reaction of methanol with 2-methyl-1-heptene (MH) as shown in Eq. (8). However, an undesirable irreversible side reaction occurs to form 2-methyl-2-heptanol (MHOH) and dimethyl ether (DME) as byproducts as illustrated in Eq. (9).



MMH RD column configuration performed by Hussain et al. was considered as base case for design [41]. The total number of column stages were 35 including the reboiler as the last stage. There were 11 trays in the reactive section, ranging from the second to the 12th stage. A molar reflux ratio of 3.785 was specified. The feed stream containing MeOH at a rate of 50 kmolhr^{-1} and MH stream at a rate of $129.5 \text{ kmolhr}^{-1}$ was introduced at 12th and second stage correspondingly in the MMH-based RD column. The operating pressure of MMH-based RD column was 1.77 atm. A total condenser and kettle-type reboiler were used for MMH-based RD column simulation, while the method of convergence was strongly nonideal liquid.

4. Results and discussion

4.1. Synthesis of methyl acetate

4.1.1. Effect of catalyst holdup on conversion

The conversion of reactants or product formation in an RD process is significantly influenced by catalyst loading on reactive trays. A large reactive holdup volume leads to high reactant conversion. The effect of an increase in the reactive holdup volume on the conversion profile in the MA-based RD process was studied through simulations by varying the overall catalyst loading on reactive trays iteratively. The simulation results depicted in Fig. 4 demonstrate a clear correlation between the amount of reactive holdup and conversion.

4.1.2. Effect of the catalyst holdup volume on heat load

The effect of catalyst holdup on reboiler's heat load to maintain product specification was examined. A large holdup volume steadily decreases the vapor boilup rate and reflux ratio to achieve a specified conversion. The influence of the reactive holdup volume on reboiler heat

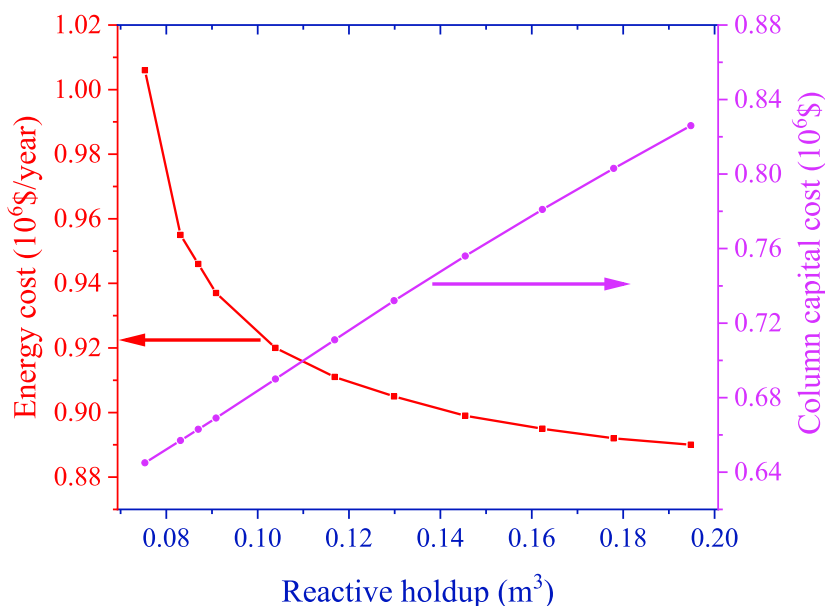


Fig. 8. The impact of reactive holdup volume on the economics of MA-based RD column.

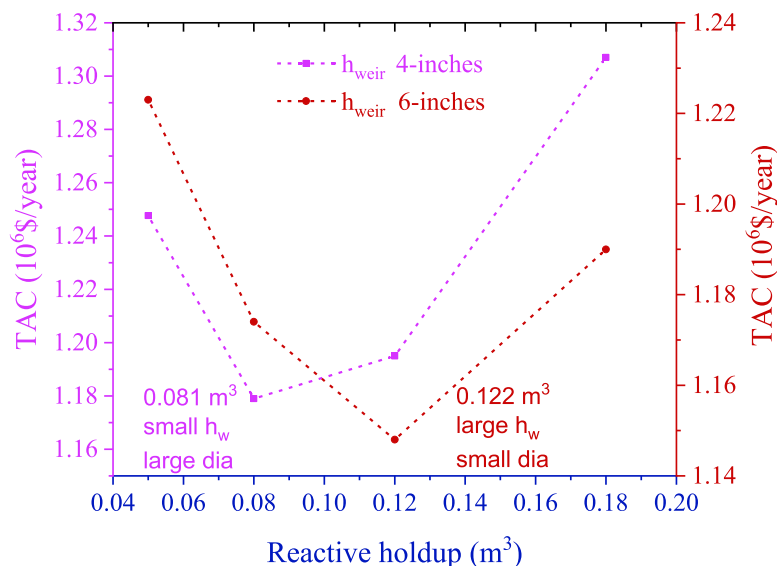


Fig. 9. MA-based RD column: TAC as a function of reactive holdup and weir height.

Table 1

TAC calculations of MA-based RD column.

Parameters	Reactive holdup (m ³)						
	0.075	0.079	0.087	0.104	0.122	0.162	0.195
ID (m)	1.183	1.212	1.271	1.389	1.493	1.736	1.902
Reboiler duty QR (MW)	2.208	2.181	2.074	2.018	1.998	1.961	1.950
Condenser duty QC (MW)	2.579	2.552	2.445	2.389	2.370	2.333	2.321
Operating cost (10 ⁶ \$/y)	1.006	0.994	0.946	0.920	0.911	0.895	0.890
Capital cost (10 ⁶ \$)	0.651	0.657	0.663	0.690	0.711	0.781	0.826
Total annual cost (10 ⁶ \$/y)	1.223	1.213	1.167	1.150	1.148	1.155	1.165

duty was analyzed by varying reactive holdup volumes. Fig. 5 illustrates the specified reflux ratio and the corresponding heat load of the reboiler change as the reactive holdup volume is increased. To achieve the required product purity at a high reactive holdup volume, a low reflux ratio is required reflecting the reduced reboiler heat duty of the MA-based RD column and vice versa.

To further understand the philosophy behind the impact of the reactive holdup volume on the reboiler heat load, the net generation profiles of MA are plotted against various reactive holdup volumes as illustrated in Fig. 6. The reaction takes place in the lower portion of the reactive section when the catalyst holdup volume is increased providing better internal heat integration due to the utilization of reaction heat

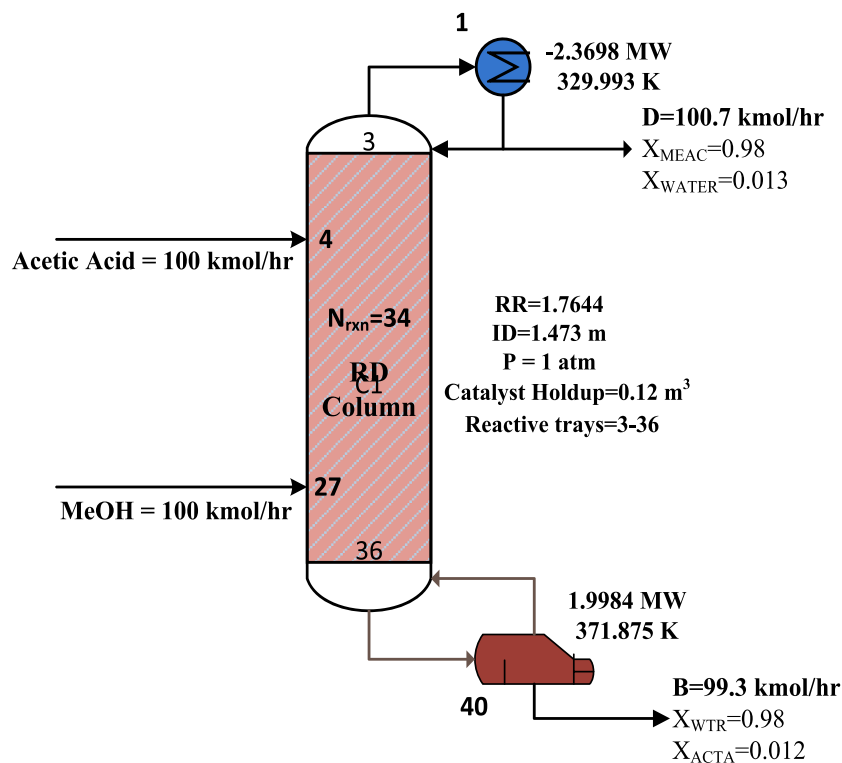


Fig. 10. Configuration of MA-based RD column.

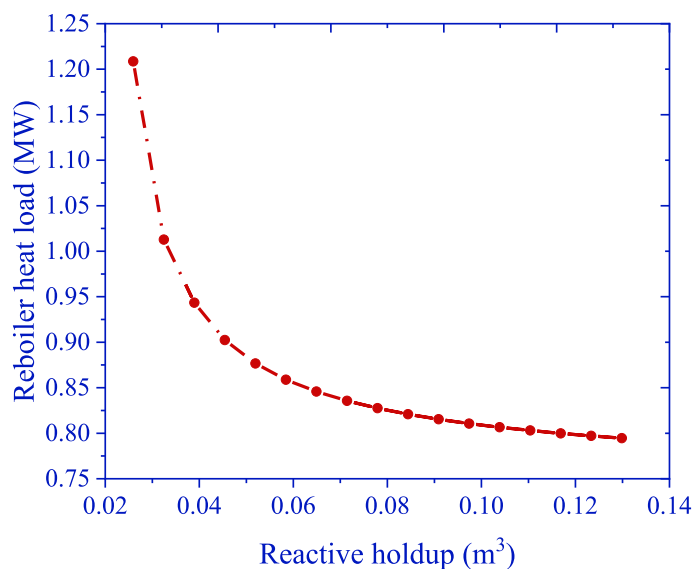


Fig. 11. Influence of reactive holdup on the reboiler load in the ML-based RD column.

between conversion and separation processes. In the RD operation, heat of reactions can be used effectively as a heat source to drive the separation operation in the stripping section, reducing the reboiler heat load. Moreover, the amounts of catalyst holdup serve as a critical design variable in strengthening the internal heat integration of a reactive distillation column involving a kinetically controlled conversion [44–46].

4.1.3. Economic analysis of the MA-based RD process

The maximum vapor velocity, reactive holdup volume, and weir height are governing parameters to establish the diameter of an RD column [47]. The impact of catalyst loading on the reaction is studied,

and the most optimal reactive holdup volume is selected through the simultaneous design based on the weir height, column diameter, and economic feasibility. The column diameter is determined from the sizing relationship for different catalyst holdup volumes and corresponding weir heights as presented in Eq. (1). Fig. 7 shows the changes in the column width with stage holdup volume variation with different tray weir heights. The estimated diameter is 1.70 m when the tray weir height is at its lowest limit of 4 in and the holdup volume is 0.10 m³. However, the estimated diameter increases to 4.65 m when the catalyst holdup volume is 0.78 m³. A holdup volume of 0.10 m³ corresponds to a diameter of 1.38 m when the weir height is at its upper limit of 6 in while a holdup volume of 0.78 m³ results in an estimated diameter of 3.80 m.

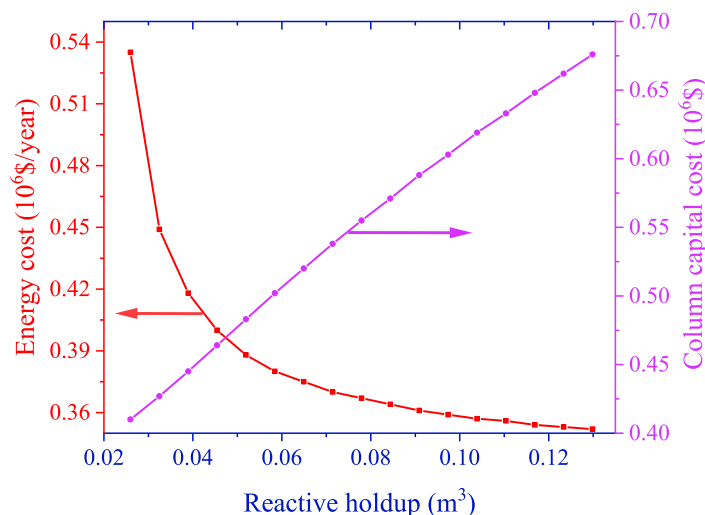


Fig. 12. The impact of reactive holdup volume on economics of ML-based RD column.

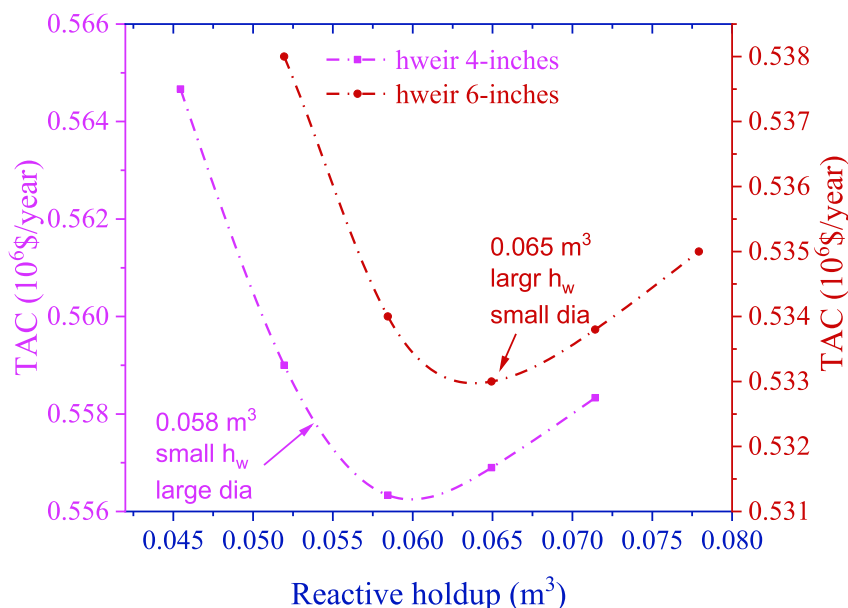


Fig. 13. ML-based RD column: TAC as a function of reactive holdup volume and weir height.

These results depict that a RD column with an upper limit of tray weir height can accommodate a large reactive holdup volume, reducing the required RD vessel diameter. Inversely, a small weir height on the tray necessitates a big diameter of the RD column to accommodate a similar catalyst holdup volume.

The impact of increasing reactive holdup volume on the economics of an MA-based RD column is examined. The energy cost and capital investment of the MA-based RD column are represented as a function of catalyst holdup to find an optimum reactive holdup volume. The dependency of column capital and energy cost on catalyst holdup volume is shown in Fig. 8. At higher reactive holdup, the reboiler heat load decreases due to better internal heat integration within the MA-based RD column. This proves that a high reactive holdup volume necessitates a large diameter of the RD column. Inversely, a low reactive holdup volume corresponds to a small column capital investment, but the energy cost is elevated. So, there exists an optimum design of MA-based RD column with respect to catalyst holdup volume that strikes a balance between energy and capital costs.

Using this data, the TAC is calculated by combining annual capital

and energy costs with a payback period of three years. The sizing relationship and economic constraints presented in Eq. (2) are utilized for obtaining the optimum design of the MA-based RD column.

The design combinations of tray weir heights and reactive holdup volumes exist for achieving the required 98 mol% purities of both MA and water. Two design alternatives associated with the upper and lower bound of practical weir height on the tray are further investigated for the catalyst holdup volume and their economic impact is presented in Fig. 9. The minimum TAC is linked to the column diameter of 1.577 m corresponding to an optimal catalyst holdup volume of 0.81 m³ against the lower limit of the weir height. The minimum TAC corresponds to a column diameter of 1.473 m with an optimal catalyst holdup volume of 0.122 m³ for the upper limit of weir height. The MA RD design by William Luyben and Hao Ye Lee has reported the diameter of 1.0327 m for MA-RD based on a fixed holdup of 0.038 m³ at flow rate of 50 Kmol/hr for each feed stream. The detailed TAC analysis of MA-based RD synthesis is further presented in Table 1 and the obtained design configuration for optimized catalyst holdup is shown in Fig. 10. The red highlighted area from second to 35th tray indicates the reactive section

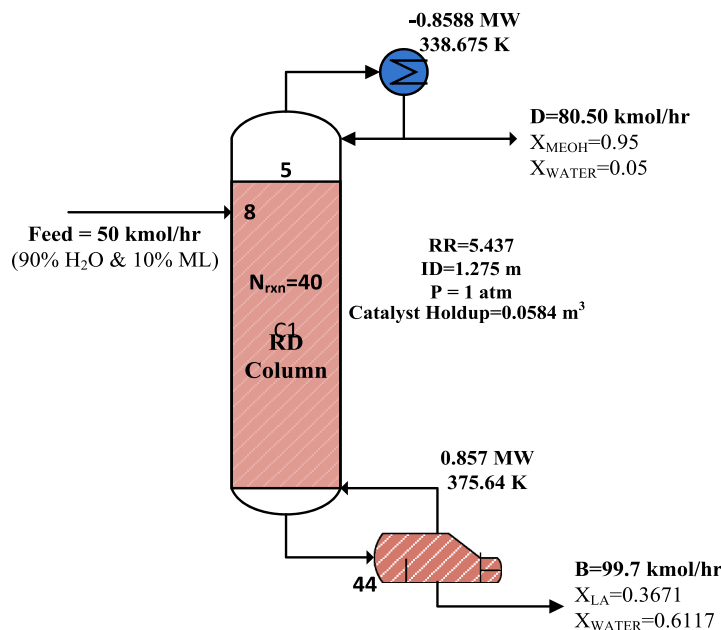


Fig. 14. Configuration ML RD Column.

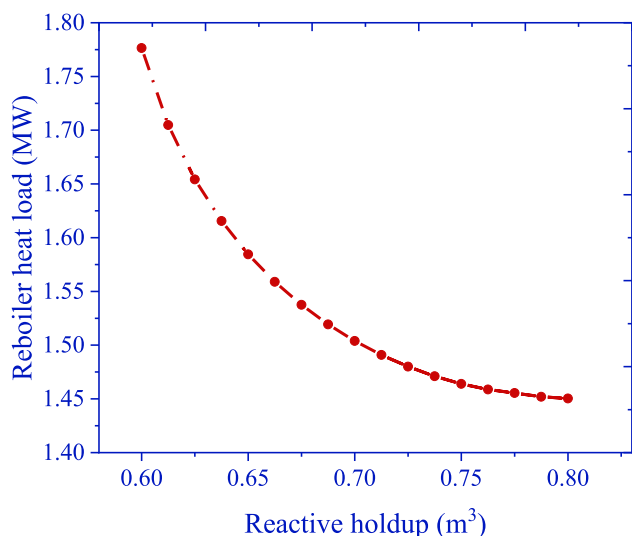


Fig. 15. Influence of reactive holdup on the reboiler load in the MMH-based RD column.

of MA RD column.

4.2. Hydrolysis of methyl lactate

The impact of reactive holdup volume on the reboiler heat duty of ML-based RD column was investigated to achieve a desired product specification. The influence of reactive holdup on reboiler heat duty was analyzed by increasing the catalyst holdup volume. Results indicated that a higher reactive holdup volume led to a gradual decrease in reboiler heat load for achieving a specified conversion, resulting in improved energy efficiency. Fig. 11 demonstrates the association between reactive holdup volume and the required reboiler heat duty of a ML-based RD column.

The impact of increasing the reactive holdup volume on economics of ML-based RD column is investigated. The energy cost and capital investment of the ML-based RD column is plotted as a function of catalyst holdup to obtain an optimal reactive holdup volume. The dependency of

column capital and energy cost on reactive holdup volume is evident in Fig. 12. At higher reactive holdup volume, the reboiler heat load decreases sharply due to improved internal heat integration, necessitating a larger diameter for the RD column. Inversely, a smaller reactive holdup volume results in a lower capital investment but higher energy costs. Consequently, there exists an optimum design of ML-based RD column with respect to catalyst holdup volume that strikes a balance between energy and capital costs.

The design combinations of the tray weir height and reactive holdup volume exist for computing the TAC of the ML-based RD process to obtain appropriate product purity. Two design variants associated with the upper and lower bounds of the realistic weir height on the tray for the ML-based RD column are further analyzed in terms of reactive holdup volumes and their economic impact are represented in Fig. 13. The minimum TAC is linked with the reactive holdup volume of 0.058 m³ against the lower limit of weir height. The minimum TAC corresponds to a reactive holdup volume of 0.065 m³ for the upper limit of weir height, while Mo et al. has reported a diameter of 0.59 m for a fixed catalyst holdup of 0.0157 m³ at a feed flow rate of 1 kg s⁻¹. The obtained design configuration for optimized catalyst holdup of the ML-based RD column with operating specifications is shown in Fig. 14.

4.2. Methoxy methyl heptane synthesis

For MMH-based RD column, the influence of reactive holdup volume on the reboiler heat duty was examined to achieve a desired product specification. By increasing the volume of catalyst holdup, the impact of reactive holdup on reboiler heat duty was examined. Results indicated that a higher reactive holdup volume led to a gradual decrease in reboiler heat load for achieving a specified conversion, resulting in improved energy efficiency. The correlation between the reboiler heat duty and the volume of reactive holdup for an MMH-based RD column is illustrated in Fig. 15.

The economic impact of increasing the reactive holdup volume on an MMH-based RD column is investigated. The energy cost and capital investment of the column is plotted as a function of catalyst holdup to obtain an optimal reactive holdup volume. The dependency of column capital and energy cost on reactive holdup volume is shown in Fig. 16. At higher reactive holdup volume, the reboiler heat load decreases due to improved internal heat integration, necessitating a larger diameter for

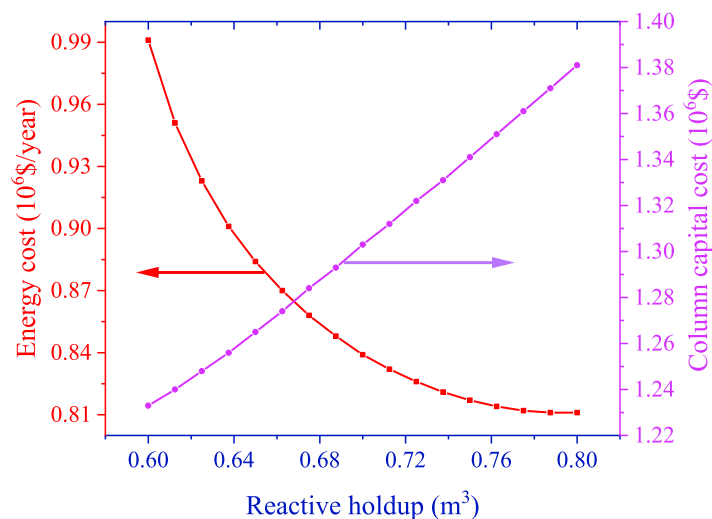


Fig. 16. The impact of reactive holdup volume on economics of MMH-based RD column.

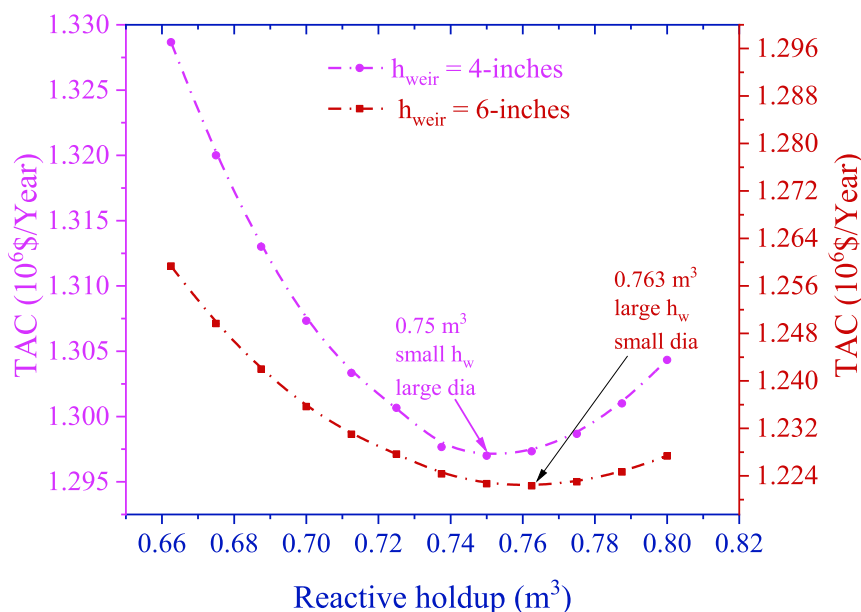


Fig. 17. MMH RD column: TAC as a function of reactive holdup volumes and weir heights.

the RD column. Conversely, a smaller reactive holdup volume results in a lower capital investment but higher energy costs. Therefore, there exists an optimum design of MMH-based RD column with respect to catalyst holdup volume that strikes a balance between energy and capital costs.

The analysis of the technoeconomic aspects of the MMH-based RD process reveals specific design combinations between the tray weir height and reactive holdup volume to achieve the target product purity. Two different column design options corresponding to the upper and lower limits of feasible weir heights on the tray are analyzed for the reactive holdup volume. The economic implication of both design alternatives is shown in Fig. 17. The minimum TAC is linked with a reactive holdup volume of 0.75 m^3 against the lower limit of the tray weir height. The minimum TAC corresponds to a reactive holdup volume of 0.763 m^3 for the upper limit of weir height, while Hussain et al. has reported a diameter of 3.90 m for MMH RD column a fixed catalyst holdup. Fig. 18 presents the obtained design configuration for optimized catalyst holdup of MMH-based RD column with operating specifications.

5. Conclusions

Although RD appears to be a promising concept, its applicability is quite restricted, particularly with heterogeneous catalysts. Slow reactions necessitate large catalyst holdup volumes, leading to significant hydraulic constraints within the column diameter established by vapor loading. The large catalyst holdup volume accelerates the attainment of desired conversion. As the catalyst holdup volume on trays is increased, the reboiler heat load decreases steadily, as the heat of reaction can be utilized efficiently to drive the separation process in the stripping section of RD column. However, a large reactive holdup volume requires either a large column diameter or a high tray weir height, subject to the constraint of excessive column pressure drop, to accommodate the catalyst. The present study investigated an alternative design methodology to accommodate a large catalyst holdup volume by increasing the column diameter beyond that which is established by vapor loading. Several combinations of tray weir heights (typically representing the lower and upper limits of actual columns) and column diameters were studied and demonstrated through case studies for three industrial

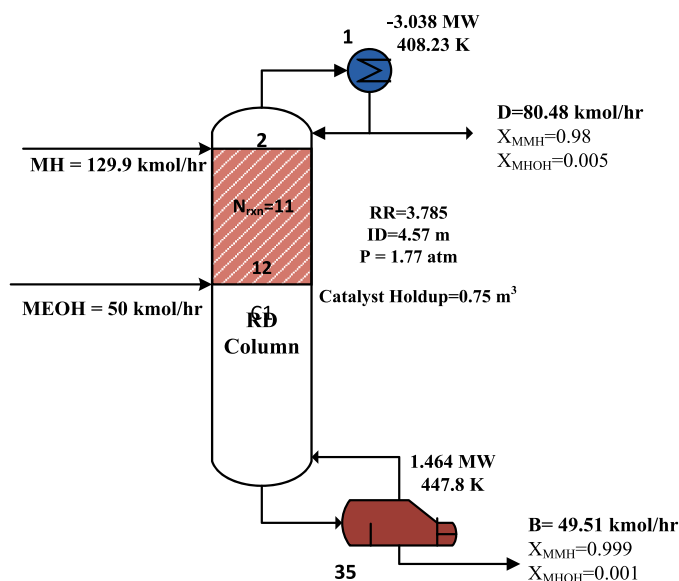


Fig. 18. Optimal MMH-based RD column.

processes. The design configuration of RD processes with optimized catalyst holdup resulted in improved economics. Process intensification via the proposed design can accommodate large catalyst holdup volumes, overcome hydraulic limitations, and improve energy efficiency, despite requiring large diameter vessels. Fabricating a wide column diameter is a good conservative engineering procedure considering safety aspects and better design control.

CRedit authorship contribution statement

Abubakar Saleem: Conceptualization, Methodology, Writing – original draft, Software. **Umar Farooq:** Investigation, Data curation, Writing – original draft. **Amjad Riaz:** Investigation, Validation, Supervision, Writing – review & editing. **Faisal Ahmed:** Formal analysis, Visualization. **Arif Hussain:** Conceptualization, Writing – review & editing, Project administration, Supervision. **Moonyong Lee:** Funding acquisition, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.cep.2023.109440](https://doi.org/10.1016/j.cep.2023.109440).

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