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Intensification and performance assessment of the formic acid production process through a dividing wall reactive distillation column with vapor recompression

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ABSTRACT

Distillation operation in the production of formic acid (FA) involves a large amount of investment and has high-energy requirements. Thus, process intensification is crucial in minimizing the energy consumption and investment. Dividing wall column (DWC) is one such way of achieving it. Aim of this study is to design a dividing wall reactive distillation column (DWRDC) with vapor recompression in the production of FA instead of normal reactive distillation – distillation sequence and investigate the performance in terms of energy, cost, and carbon emission. A patented FA production process is chosen as a base case. The intensification of the base case FA production process is then carried out through the application of (i) DWRDC, (ii) vapor recompression in a distillation column, and (iii) vapor recompression in DWRDC. The performance of the new intensified processes is subsequently investigated for multiple performance indexes. It is observed that an impressive saving of 18.33% in utility cost, 8.4% in total annual cost (TAC), 2.55% in cost of manufacturing (COM), and 17.68% in carbon emission can be secured with the proposed new intensification as compared to its conventional counterpart for a payback period of 4 years.

1. Introduction

Formic acid (FA) is a strong organic acid that is principally used in various industries, such as chemical, pharmaceuticals, rubber, textile, agriculture, and leather. FA is readily bio-degradable, hence it is environmentally sustainable, which makes it a popular raw material in many industries for the production of various desirable chemicals [1]. Among several commercial processes for the FA production, namely acidolysis of formate salts, oxidation of hydrocarbons, hydrolysis of formamide, mineral acid catalysis, and the hydrolysis of lower alkyl formates, the hydrolysis of methyl formate (MF) is attractive as it overcomes the limitations of existing counterparts [2–4]. Limitations of existing processes include slow reaction rate, unwanted side-products, high cost of investment, and issues related to the environment. Hydrolysis of MF involves two steps: (1) carboxylation reaction in a

continuous stirred tank reactor (CSTR) using a heterogeneous catalyst, (2) hydrolysis of the intermediate product to obtain formic acid. This process solves the limitations of the conventional process viz. undesirable side products, slow reaction time, environmental complications, and high capital investments [5]. Huang and colleagues [6] designed a process by combining a CSTR and a simple distillation column in the MF hydrolysis-based process into a reactive distillation (RD) unit. The combination of the reaction and separation occurring simultaneously in RD provides many advantages: (1) enhanced yield and selectivity, (2) reduced energy consumption, and (3) avoidance of hot spots. In addition, it is observed that there is a significant reduction in heat duty, which further reduces the operating cost.

Distillation is the most commonly used separation technique in the chemical process industry, worldwide, accounting for 95% of all separations [7]. Sixty per cent of energy use in chemical industry is from

Abbreviations: C1-VR, vapor recompression in C1; CO, carbon monoxide; COM, cost of manufacturing; CSTR, continuous stirred tank reactor; DWC, dividing wall column; DWRDC, dividing wall reactive distillation column; DWRDC-VR, vapor recompression in DWRDC; FA, formic acid; FCI, fixed capital investment; MF, methyl formate; RD, reactive distillation; TAC, total annual cost; TCDC, thermally coupled distillation column; TCDWC, thermally coupled dividing wall columns; VRC, vapor recompression

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the distillation process [8]. A large amount of energy required in FA production is in the distillation process. Process intensification is currently considered as one of the major approaches to reduce the number of pieces of equipment in the plant and even improve process performance. For a distillation column system, thermal coupling provides such an approach to retrofit the existing simple column configurations through the elimination of either the condenser or the reboiler [9] after integrating two columns into one. Thermally coupled distillation column (TCDC) and divided wall column (DWC) in their fully integrated form have attracted strong attention due to the greater need to reduce energy requirement and capital costs in distillation operations [10]. DWC integrates the two columns into a single shell providing an excellent path for process intensification. There are considerable energy savings obtained by this process intensification with a reduction of space for installation by 35–45% due to the replacement of two columns by a single column [11–13]. The benefits of thermally coupled dividing wall columns (TCDWC) are: (1) integrated design with reduced capital investment and footprint, (2) high purity products, (3) increased thermodynamic efficiency due to reduction in remixing effects, (4) lower energy requirements in comparison to conventional separation sequences, and (5) low maintenance costs. Studies suggest that DWC systems can reduce energy requirement by up to 30% over conventional direct and indirect distillation sequences [14]. However, practical implementation and operation of this intensified structure is difficult due to the highly integrated structure. In addition, such highly integrated configurations pose a tough challenge pertaining their optimal design and control.

Energy consumption can further be reduced by a suitable heat integration technology such as vapor recompression (VRC) scheme, where a fraction of the overhead vapor is compressed and used as a heating medium in reboiler. The conventional VRC technique is more energy efficient with small temperature span across the different stages and large heat duty [15]. The VRC scheme, with side heat exchanger configurations has been applied in both conventional distillation and reactive distillations for separating wide boiling mixtures [16–18]. In the intensified configuration, the conventional VRC is added between the overhead and the bottom of DWC to recover the heat of the vapors as much as possible. Luo et al. [19] added a VRC heat pump to further increase the energy savings up to 40% in extractive distillation for the bioethanol purification process. Johri et al. [20] proposed a variable speed vapor recompression in batch reactive distillation, in which a 65.85% energy saving was obtained with a payback period of 4 years [20]. Luyben and coauthors [21] reported that VRC in azeotropic DWC secures a lower total annual cost (TAC) with a payback period of 5 years despite the higher capital costs.

Goal of this study is to suitably reduce energy consumption in the FA production process by performing process intensification through the application of a dividing wall reactive distillation column (DWRDC) with vapor recompression respect to the patented base case process for FA production. At first, the base case process is simulated using Aspen Plus v8.8. Subsequently, the process is studied for the application of DWRDC, vapor recompression in distillation column, and vapor recompression in DWRDC. The merits of the new configurations through the application of above heat integrations are discussed in terms of crucial criteria, namely energy savings, TAC, cost of manufacturing (COM), and carbon emission. To our knowledge, this is the first study in the literature that analyses the application of vapor recompression in DWRDC for the FA synthesis.

2. Process description

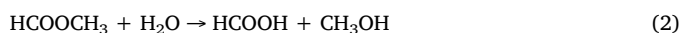
For the present study, we chose the patented configuration from Huang and coauthors [6] for FA production. This process is chosen as it combines a hydrolysis unit (i.e. a reactor) and a separation unit (i.e. a distillation column) into a reactive distillation unit to produce high purity FA, thereby reducing the processing steps and the footprint.

Fig. 1 shows the overall process flow sheet for the base case process.

The FA manufacturing system consists of a reactor (where the carbonyl reaction occurs) and three separation columns, which include one reactive distillation (RD) column for Hydrolysis of Methyl Formate as well as for separation of products. A high pressure adiabatic CSTR is used for the reaction between methanol and carbon-monoxide (CO) (Carbonyl reaction) with sodium methoxide (CH_3ONa) as a heterogeneous catalyst (Eq. (1)).



Fresh carbon monoxide is fed to the CSTR with a flow rate of 70.3 kmol/h. Make up of methanol is fed to the CSTR to maintain the excess of methanol. The reactor is operated at 75 °C and 40.53 bar. The CSTR product is flashed into the phase separator and is then fed to a distillation column to separate MF from the top of the methanol recovery unit (C1). Finally, it is fed to the RD unit. The remaining little amount of CO is purged using a partial-vapor-liquid condenser in the separation column. The total CO emission from all purge streams in this process adds up to only about 90 kg CO/h in base as well as in modified cases. The unconverted methanol obtained from the bottom of the separation column (C1) is recycled back to the CSTR. The production of FA and methanol by the hydrolysis of MF takes place in the RD column along with its separation. Required amount of fresh water is added to RD on 2nd tray (trays are numbered from top to bottom) and MF from C1 enters RD on 33rd tray.



The separation between reactants (i.e. MF and Water) and products (i.e. FA and Methanol) takes place simultaneously in the RD unit, which consists of 35 stages. This configuration helps improving the reaction efficiency as well as the separation efficiency. RD consists of some reactive trays (1st-19th from top) and a stripping zone below the reactive zone. The liquid mixture obtained from RD is fed into the separating column (C2). FA and water are separated from the C2 column as a bottom product, whereas methanol and MF are recycled back from the top. Finally, we obtain about 85% FA as a final product with a flow rate of 92 kmol/h. The distillate from the C2, mainly containing methanol, is recycled to the CSTR to maximize the yield. Important operating data, such as flow rates, temperature, heat duty, and pressure are given in Fig. 1.

3. Formic acid production process modelling and simulation

The UNIQUAC-HOC thermodynamic model of Aspen Plus is selected to deal with solvation of polar compounds and dimerization of the vapor phase [22]. This model is suitable as it accounts for strong association and solvation effects. The UNIQUAC Functional-group Activity Coefficient (UNIFAC) method is applied to estimate the missing parameters. The rigorous RADFRAC model is adopted to simulate a distillation unit explicitly by considering the three phase balances. Fig. 2 depicts T-xy diagram for FA-water at 1.03 and 4 bar. This figure shows a large deviation from ideality and dependence on the pressure. At low pressure, i.e. at 1.03 bar, a negative azeotrope (i.e. maximum boiling) is observed while that azeotrope disappears at higher pressure i.e. at 4 bar. An equilibrium stage model (RADFRAC) is used for the simulation of all distillation and RD columns. The catalyst hold up for the kinetic rate controlled reaction is 0.25 m³ of catalyst on each stage of RD.

The kinetics of the carbonyl reaction occurring in the CSTR is taken from [23]. Kinetics is valid for a temperature of 60–110 °C and pressures of about 2–4 MPa. The CSTR volume is 4084 L.

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