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Design and control of butyl acrylate reactive distillation column system

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Abstract

In this study, the design and control of a reactive distillation column system for the production of butyl acrylate has been investigated. The proposed design is quite simple including only one reactive distillation column and an overhead decanter. The optimal design is selected based on the minimization of total annual cost (TAC) for the overall system. At this optimized flowsheet condition, output multiplicity was found with reboiler duty or feed ratio as the bifurcation parameter. The highest purity stable steady state was selected as the base case condition for the control study. The overall control of this system can be achieved with no on-line composition measurements. Simple single-point tray temperature control loop is designed to infer final product purity. From results of dynamic simulation, the proposed control strategy performs very well in rejecting various disturbances while maintaining butyl acrylate product at high purity. One of the important finding in this paper is that it is better to operate this reactive distillation column not at the exact feed stoichiometric balance point for better operability reason. The control performances of the proposed operating point and the operating condition right at the exact stoichiometric balance point will be compared.

Keywords: Butyl acrylate; Reactive distillation; Operability; Design and control

1. Introduction

Reactive distillation has demonstrated its potential for capital productivity improvements, selectivity improvements, reduced energy use, and the reduction or elimination of solvents in the process (cf. Malone and Doherty, 2000). From this review paper, there are a total of 562 publications of reactive distillation for the period of 1970–1999. According to a chart in this review paper, exponential rate of growth in the literature is exhibited. For the 5-year period (2000–2004) after that review paper has been published, according to a title search using engineering database Compendex®, there are another 253 publications in the research area of reactive distillation or catalytic distillation. This shows the rapid progress of this technology sector in recent years.

Also, in a book presenting the status and future directions of reactive distillation (cf. Sundmacher and Kienle, 2003), a survey of chemical reaction schemes that performed successfully in reactive distillation columns is given. In Tables 1.1 and 1.2 of this book, over 100 industrially or potentially important reactions for reactive distillation applications are given. This illustrates the importance of this technology in industrial applications.

Butyl acrylate is widely used in industry as a precursor for varnishes, adhesive, and finishes of papers and textiles. This important ester can be produced directly from *n*-butanol and acrylic acid via esterification reaction with the presence of acid ion exchange resin as catalyst. Even though in recent year there are many papers investigating design and control aspects of reactive distillation columns, no paper on design and control of this system has been reported in open literature to the best of our knowledge. The only related paper we can find in the open literature is by Schwarzer and Hoffmann (2002) discussing the experimental reaction equilibrium and kinetics of this system. In that paper, a possible process flowsheet which consists of a catalytic tube reactor and a reactive distillation column is also simulated. However, the butyl acrylate product stream is not pure enough (only up to 93 mol%) for industrial usage.

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In this paper, design and control of a reactive distillation column system for the production of butyl acrylate will be investigated. Section 2 shows the proposed simple design of the overall system with only one reactive distillation column and an overhead decanter. The optimal design flowsheet is determined by minimizing TAC of the overall system. In Section 3, output multiplicity of this system is illustrated with reboiler duty or feed ratio as the bifurcation parameter. The stable steady state with the highest purity is selected as the base case condition for the control study. Another base case condition with two feeds at exact stoichiometric balance is also selected for control performance comparison purpose. Section 4 shows the overall control strategy development of this system. Only single-point tray temperature (not on-line composition analyzer) is needed in the overall control strategy. The closed-loop dynamic simulations of the proposed control strategy in the face of various disturbances including throughput change, feed composition variations, and catalyst decay disturbance are illustrated in this section. Finally, some concluding remarks are drawn in Section 5.

2. Proposed process design

The reaction taking place in the reactive distillation column can be seen as below:

$$CH_3(CH_2)_3OH + CH_2CHCOOH \leftrightarrow CH_2CHCOOC_4H_9 + H_2O$$

(BuOH) (AA) (BA)

The kinetics of this reaction was assumed to behave according to a Langmuir Hinshelwood Hougen Watson (LHHW) mechanism. The kinetics parameters from Schwarzer and Hoffmann (2002) are used in this study. The reaction rate expression is shown below:

$$r_{AA(kmol/m^3\,s)} = K_1 \exp\left(-\frac{K_2}{RT}\right) \times \frac{a_{BuOH}a_{AA} - (1/K_a)a_{BA}a_{H_2O}}{(K_3 a_{BuOH} + a_{AA} + K_4 a_{H_2O})^2} \quad (1)$$

with

$$K_{1(kmol/m^3\,s)} = 8.12 \times 10^9, \quad K_{2(J/mol)} = 8.37 \times 10^4,$$
$$K_3 = 1.864, \quad K_4 = 1.308,$$

$$\ln K_a = -8.805 + 0.05743\,(T/K) - 6.429 \times 10^{-5}\,(T/K)^2 + 3.821 \times 10^{-9}\,(T/K)^3$$

In Schwarzer and Hoffmann's paper, possible polymerization of acrylic acid was not mentioned when they were doing the equilibrium and kinetic experiments of this system. However, from another paper (Witczak et al., 2004) as they were studying the kinetics of reactions producing methyl and ethyl acrylates, an inhibitor hydroquinon was mentioned to prevent polymerization of acrylic acid. In our paper, this polymerization inhibitor is also assumed to be added in the system to prevent polymerization of acylic acid. Since the concentration of the inhibitor is so low (less than 0.2 wt%), this component was not included in the following computer simulations.

The NRTL–HOC thermodynamic model is used to describe vapor–liquid and vapor–liquid–liquid equilibrium with liquid activity coefficients calculated by non-random-two-liquid (NRTL) model and vapor association of AA due to dimerization included by using the second virial coefficient of the Hayden and O'Connell (1975) in the vapor phase. Aspen Plus (2001a) built-in thermodynamic model parameters are used in the simulation. For the two model parameter pairs (BA–AA and BA–H_2O) that do not have the Aspen Plus built-in NRTL parameters, the Dortmund modified UNIFAC group contribution estimation method (Weidlich and Gmehling, 1987; Gmehling et al., 1993) was used to obtain the remaining thermodynamic model parameters.

This system includes two homogeneous azeotropes (AA–BuOH and BA–AA) and two two-component heterogeneous azeotropes (BA–H_2O and BuOH–H_2O) and one three-component heterogenous azeotrope (BA–BuOH–H_2O). The normal boiling point of pure components and the azeotropic temperature and composition of all the azeotropes as predicted by the NRTL–HOC thermodynamic model parameters are given in Table 1.

The proposed design of the reactive distillation column is to produce high purity butyl acrylate product (>99.5 mol%) at the column bottom and to have top vapor composition near three-component heterogeneous azeotrope. This top vapor after condensation can be formed into two liquid phases to be separated in a decanter. The organic phase containing mixture of butanol, butyl acrylate and some water can be refluxed back to the reactive distillation column. The aqueous phase containing high-purity water (over 95 mol%) can be discharged out of the system. The feasibility of the design can be illustrated by the following two residue curve maps (RCMs). Fig. 1 shows the BuOH–BA–H_2O three-component RCM which can be used toward the top of the reactive distillation column where acrylic acid is negligible. From the figure, one can observe that the lowest temperature is the BA–BuOH–H_2O three-component azeotrope and this point is inside of two-liquid boundary, thus the above conceptual design toward the top of the column is feasible. Fig. 2 shows the AA–BuOH–BA three-component RCM which can be used toward the bottom of the reactive distillation column where water is negligible. Although the pure BA corner of this figure is not a stable node, it is possible to properly design the column so that the bottom product is at this saddle point. The proposed design of the overall system including a reactive distillation column and an overhead decanter can be seen in Fig. 3.

The design and operating variables that need to be optimized are total stages of the reactive distillation column, acrylic acid feed stage, butanol feed stage, reboiler heat duty, and acrylic acid feed flow rate. The decanter is assumed to be operated at atmospheric pressure and top of the column operated at 1.1 atm. Because possible polymerization of acrylic acid may happen at higher operating temperature, raising of the column operating pressure in favor of kinetics and possibly in favor of relative volatilities was not considered in this paper.