



Retrofitting an isopropanol process based on reactive distillation and propylene-propane separation



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ABSTRACT

In this study, an isopropanol (IPA) process based on reactive distillation (RD) using excess propylene and propylene-propane separation column (splitter) is analyzed for retrofitting. This analysis led to identifying two limitations of this process, namely, excess propylene feed to RD and requirement of high purity IPA product from RD. To overcome these limitations, two retrofit designs are proposed. One modified process introduces a new RD with excess water to replace the propylene-propane splitter; simulation results show 14.1% reduction in manufacturing cost per unit product (MCU, decreases from 0.092 \$/kg to 0.079 \$/kg) for this retrofit design. In the other retrofit design, RD produces an azeotropic mixture of IPA and water, thus requiring extractive distillation and solvent recovery columns. This modified process leads to a slight increase in MCU (from 0.092 \$/kg to 0.095 \$/kg). Hence, the design involving a new RD with excess water is recommended for retrofitting the original process.

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1. Introduction

Process intensification (PI), defined as ‘any chemical engineering development that leads to a substantially smaller, cleaner, safer and more energy efficient technology’ [18], is an effective strategy to achieve capital and/or operating cost savings. In recent years, it is attracting considerable interest from both academia and industry as a promising way for enhancing process performance, product quality and adapting to more stringent environmental regulations. Many PI-based unit operations such as rotating packed bed (also known as HiGee), dividing-wall column (DWC) and multi-functional reactors (e.g. reactive distillation, reactive extraction, reactive absorption, reactive adsorption and reactive crystallization) were studied in the literature [18,17].

One common PI-based unit operation is reactive distillation (RD). Different from that in a conventional chemical plant, where separate equipment are linked together by material and energy streams, in RD, separation and reaction occur in the same vessel [6]. This results in significant reductions in both energy and equipment in process systems having appropriate reaction kinetics and vapor-liquid equilibrium [15]. According to Taylor and Krishna [24], and Sharma and Mahajani [22], an average 20% savings in total annual cost (TAC) is achieved by using RD. Reay et al. [18]

summarized other advantages of RD as follows: improved conversion and selectivity of the main reaction, increased reliability, reduction of byproducts, better heat integration and control of highly exothermic reaction, and reduced problems with hot spots and thermal runaway.

RD is commonly used for esterification and etherification [26]. The first patent to employ RD dates back to 1920s, when RD was proposed for homogeneous catalyzed esterification [20]. The most well-known RD-based esterification is the ‘Eastman Process’ for methyl acetate. Eastman’s first methyl acetate RD column was established in 1980, producing over 200,000 metric tonnes of methyl acetate per year [26]. Owing to its successful implementation in the industry, many researchers used this system for experimental and modeling studies on RD. As a result, different aspects of methyl acetate RD, for instance, characterization of the catalyst used, effect of operating conditions on the performance and hydrolysis, have been well studied (e.g., Kolodziej et al. [10])

For etherification, a typical process involving RD includes synthesis of methyl tertiary butyl ether (MTBE) and ethyl tertiary butyl ether (ETBE). Baur et al. [2] simulated the RD-based MTBE process using both the equilibrium stage (EQ) and non-equilibrium stage (NEQ) models. Their results show that, although both models predict multiple steady states, the window in which these multiplicities occur is significantly reduced in the NEQ model. Another finding from their study is that the hardware design may have a significant influence on the conversion and selectivity of the reactions. This influence is further confirmed by Lee and

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Westerberg [12], who introduced a graphical tray-by-tray method for the same system. This graphical method can help to determine the optimal location for the reaction zone in RD column and also the optimal reflux ratio. Results in Lee and Westerberg [12] are in accordance with the values reported in other papers.

There are some studies on producing multiple products using a RD. For example, Rix et al. [19] reported 99.9% of isobutene as a distillate with 98% pure MTBE as a bottom product from the RD column. Tadó and Tian [23] used a nonlinear inferential model to simulate a 10-stage ETBE RD pilot plant. Their results from the model are in good agreement with the experimental data. Besides esterification and etherification, other prospective applications of RD include liquid-liquid hydrolysis, oxidation and fermentation [18].

The literature reviewed above covers most of the classical examples of RD-based processes. Lutze and Gorak [13] noted that most of the studies on RD assume high purity reactants and analyze one-main-product-one-reaction systems. However, real industrial systems are usually more complex in terms of number of reactions involved and products generated. To address these, recent studies focused on complicated systems. González R. et al. [5] simulated a RD-based tertiary amyl ethyl ether (TAEE) process with around 20 chemical reactions and 100 components. Following the pre-defined criteria (e.g. pure component properties, azeotropes and their presence in reactions), certain components were lumped, and the system reduces to 20 components and 6 chemical reactions. Even then, good agreement between experimental data and simulation results was found by González R. et al. [5].

Keller et al. [9] applied RD to a multiple reactions system, where diethyl carbonate and ethyl methyl carbonate are simultaneously produced. Their results show an improved selectivity of the product (diethyl carbonate) compared to a conventional stirred tank reactor. Holtbruegge et al. [7] reported a study involving both experiments and simulations for simultaneous production of dimethyl carbonate and propylene glycol in a pilot-scale RD column. In these experiments, selected parameters were varied to test the feasibility of simultaneously producing the two products. In the simulation, NEQ model was employed to analyze trends of reactant conversion and product purities. Results in Holtbruegge et al. [7] suggest the possibility of achieving high propylene carbonate conversion while recovering an azeotropic mixture of dimethyl carbonate and methanol in the distillate. Later, Holtbruegge et al. [8] suggested a modified process scheme, where a membrane is added to assist the RD column, and their results show its potential for decreasing energy required.

It is clear that the topic of RD is not new, and there have been many studies on RD. Nevertheless, many of them are focused on grass-roots design. In case of process retrofitting, more constraints from the process and original design (since existing equipment should be reused as much as possible) need to be considered. RD related process retrofitting involves two aspects: one is introducing RD to replace the conventional reactor and downstream distillation column, and another is to modify the existing RD process, especially those with complex systems, so as to improve its efficiency. Although both aspects are essential, the latter is novel because, to the best of our knowledge, there is no previous study to retrofit an existing RD process. Hence, the present paper analyzes the isopropanol (IPA) process based on RD using excess propylene and propylene-propane separation column (splitter) to identify its limitations, and then suggests two retrofit designs. One retrofit design involves a new RD column using excess water (instead of excess propylene); this is the second novelty in this study. Finally, the two retrofit schemes are compared to find the most promising one.

IPA, also known as isopropyl alcohol and 2-propanol, is a secondary alcohol with the chemical formula of C_3H_7OH . It is

widely used as a solvent in many industries and finds applications in food processing, coatings to reduce flammability, as thinner and additive in paints; it is also used as a disinfectant such as alcohol wipes in household products and medical applications [4]. IPA is manufactured by two major commercial processes: indirect and direct hydration of propylene, of which the latter is common due to less corrosion in unit operations. Direct hydration of propylene is an exothermic, reversible reaction carried out with an acid catalyst, which could be cation-exchange resins such as molybdophosphoric acid, titanium and zinc oxides [11]. The main reaction is:



The main side product, diisopropyl ether (DIPE) forms by the etherification reaction:



Conventional IPA process consists of 7 main unit operations, namely, trickled bed reactor, propylene-propane splitter, lights column, pre-concentrator, ether column, extractive distillation column and regenerator for solvent recovery [11]. A novel process was studied by Xu et al. [28], which only requires two unit operations, namely, the RD column and the propylene-propane splitter; rather than 7 units in the conventional process. However, simulation of splitter is not included in the study of Xu et al. [28]. Recently, Niu and Rangaiah [16] investigated the simulation and cost estimation of the complete RD-based IPA process, and used it to retrofit the conventional IPA process. By this retrofitting, around 12% savings is achieved in unit production cost. Even then, cost estimation showed that operating cost, especially steam consumption, is quite high. Therefore, using the RD-based IPA process as the base case, this study explores the retrofit designs to reduce operating cost.

The rest of this paper is organized as follows. Section 2 presents the IPA process based on RD using excess propylene and propylene-propane splitter, and its cost estimation; it also includes the analysis of this process, as the base case, to identify the contributing units (i.e., unit operations requiring the most utilities) and other key limitations. Based on this analysis, two retrofit solutions are proposed, simulated and evaluated in Section 3. Conclusions of this study are presented in Section 4.

2. Base case of RD-based IPA process

In this section, simulation and analysis of the RD-based IPA process (consisting of RD column and propylene-propane splitter) are carried out to identify the key limitations of this process.

2.1. Simulation of the original process

Simulation of the RD-based IPA process is conducted using Aspen Plus 8.6 and UNIFAC model as the fluid package. Process flowsheet with key stream data is shown in Fig. 1. As can be seen, fresh propylene (at -47.6°C and 101.325 kPa, [1] is pressurized to 870 kPa, heated to 13.5°C in a heat exchanger (with assumed pressure drop of 20 kPa) and then mixed with recycled propylene in a molar ratio of 2.29:1. This mixed reactant is further pressurized to 2013.5 kPa and fed to 5th stage of the RD column. Meanwhile, fresh water is pressurized to 2013.5 kPa and fed to the third stage of the RD column. The RD column has 28 theoretical stages, and the reaction zone is from 3rd to 5th stage, with acidic proton-exchange resin as the catalyst [11]. The product stream (99.99 mol% IPA) leaves the RD column from the bottom, and then sent to E_2 for heat integration (severing as one heat source for the reboiler of propylene-propane splitter). The outlet temperature of the product stream from E_2 is set at 35.4°C to ensure sufficient driving force

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