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Ethanol and isobutanol dehydration by heat-integrated distillation

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ABSTRACT

Alternative processes with double-effect distillation (DED) and vapor compression distillation (VCD) were studied for ethanol and isobutanol dehydration from dilute concentrations. The extractants evaluated for ethanol dehydration were glycerol and ethylene glycol. Simulations were performed in Aspen Plus®. The lowest energy consumption for ethanol and isobutanol dehydration were achieved by VCD (2.5 and 3.7 MJ-fuel/kg-product, respectively). The energy consumption for isobutanol and ethanol separations with VCD were 25–30% and 39–40% lower than DED, respectively. Due to the high cost of the compressors, VCD was between 9 and 16% more expensive than DED. Due to the higher ethanol concentration from the fermentation broth, the separation annualized costs and the fuel requirement for ethanol dehydration were 37-44% and 32–46% lower than butanol separation, respectively. However, the energy efficiency, with a maximum theoretical yield from glucose, for isobutanol and ethanol processes was approximately equivalent, 72–73% (DED) and 77% (VCD), due to the higher combustion heat of isobutanol.

1. Introduction

Bioethanol from sugar and grains is the dominant biofuel in the market. Isobutanol, an alternative biofuel, has higher energy content, lower vapor pressure and lower corrosivity than ethanol [1]. Industrially, butanol and ethanol separation are carried out mainly by distillation [2,3]. Distillation is the unit that consumes most energy in the fermentation process [4]. Therefore, this work studied alternatives to conventional distillation with low energy consumption for butanol and ethanol production.

Butanol is mainly produced via petrochemical route [1]. Biotechnological production of butanol is carried out conventionally by acetobutylic fermentation. In this fermentation, acetone and ethanol are obtained as by-products. Low butanol concentration (<2 wt%) causes high toxicity and high product inhibition in biocatalyst [5]. Consequently, acetobutylic fermentation has low productivity, expensive energy consumption, and low yield [1]. Alternatively, recombinant microorganisms have been developed for exclusive production of isobutanol [6–8]. *Escherichia coli*, *Saccharomyces cerevisiae*, and *Bacillus subtilis* are the most well studied microbial strains that can be potentially induce to produce butanol [6]. In these fermentation processes, isobutanol is produced at concentrations lower than 2 wt% [7].

Several integrated reactors with gas stripping, pervaporation, liquid extraction or adsorption have been proposed in the literature to avoid the butanol toxicity [9–15]. In these systems, reactor productivity and the final biobutanol concentration increase. The final separation of this process is carried out by azeotropic distillation. Water-butanol azeotrope behavior is broken by decantation. The energy requirements of an integrated reactor depend on both the distillation and the separation units of hybrid systems. Therefore, an efficient distillation system is necessary to improve the performance of integrated reactors.

Ethanol in contrast to isobutanol is produced mainly through fermentative route. The wine obtained after fermentation contains about 7–12 wt% of ethanol. Due to azeotrope known at 95.3 wt%, an entrainer is necessary in bioethanol purification. Several processes have been proposed for ethanol dehydration such as molecular sieves [16], membranes [17], azeotropic distillation [18], extractive distillation [19–22] or hybrid methods combining these options [17,18,23]. The extractive distillation is one of the most economical ways to produce anhydrous ethanol [24].

Abbreviations: DED, double-effect distillation; LHV, lower heating value; PRE, pre-concentration column; TAC, total annualized cost; TIAC, total investment annualized cost; TED, extractive column; TER, column for extractant regeneration; TOAC, total operational annualized cost; VCD, vapor compression distillation; S-I, isobutanol dehydration process with VCD; S-II, isobutanol dehydration process with DED; S-III, ethanol dehydration process with VCD; S-IV, ethanol dehydration process with DED.

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Process intensification in distillation follows various integration routes such as internal heat integrated distillation columns (HIDiC) [25,26], vapor compression distillation (VCD) [27], Petlyuk or dividing wall columns [28,29] and double-effect distillation (DED) [30–32]. Dividing wall and Petlyuk distillation are used with more than two components. Hence, it is not useful for isobutanol separation. In DED, the fed flow is divided and pumped into the two distillation columns. One column operates at low pressure and the other at atmospheric pressure or higher, so that the condensation heat of the higher pressure column can be used to supply heat to the boiler of the vacuum pressure column. In VCD, the top vapor in the column is compressed, supplying heat to its reboiler.

DED [31] and VCD [33] have been studied to obtain hydrous ethanol (93 wt%). VCD with extractive distillation was studied in a dividing wall scheme [34–36]. In butanol dehydration, multistage VCD was proposed by ABE separation in Cobalt technologies® patent [27]. In this work, heat-integrated distillation with VCD and DED were studied by ethanol and isobutanol dehydration. For ethanol dehydration, extractive distillation was studied. Both integrated process, VCD and DED, involve more capital investment than conventional separation. Therefore, an economic study was performed.

2. Methodology

The distillation columns were simulated using the Aspen Plus V7.3® software with the RadFrac model. The property method for vapor liquid equilibrium was NRTL-HOC. UNIQUAC-LL parameters from Aspen Plus® were used to simulate the decanter in isobutanol separation. Ethanol and isobutanol recovery through distillation process were fixed in 99.8%. The columns were simulated with Murphree efficiencies of 0.7. In the simulation, sieve trays were assumed. Diameter and pressure drop was calculated with Aspen Plus® tool. The spacing plate was fixed at 0.61 m. Compressors were simulated with an isentropic efficiency of 0.75. Heat integration was performed with 10 °C of minimum approach temperature. In all tested cases, vinasses were used for preheating the feed. The purity of solvent was 0.997. The total annualized cost (TAC) was calculated with the Guthrie method [37]:

$$TAC = \frac{TOAC + {}^{T}IAC/_{t_{ri}}}{F_p \times t_a} \quad (1)$$

TOAC, TIAC, PP, F_p, and t_a, stand for the total operation annualized costs, total investment annualized capital, payback period (3 years), product flow, and annual operation time (8150 h), respectively. Costs of steam, cooling water and electricity were assumed as 16.3 $/tonne, 0.0067 $/kg, and 0.086 $/kWh, respectively. Glycerol cost was 3 $/kg. The Marshall & Swift equipment cost index (M&S) has a value of 1569 [38]. Process equipment was designed using stainless steel material. Energy requirement was calculated with an efficiency in steam and electricity production from fuel of 0.9 and 0.3, respectively. The simulation conditions were determined by an energy minimization in Aspen Plus®.

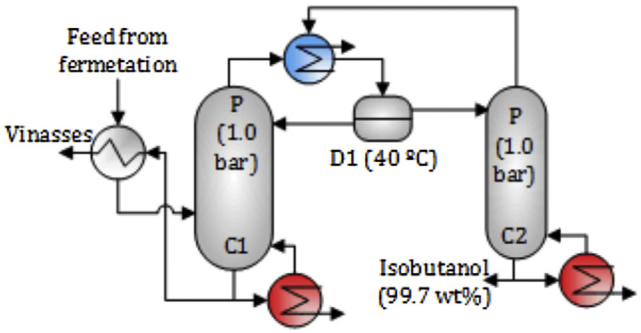

Fig. 1. Isobutanol separation by conventional distillation.

2.1. Description of process for isobutanol dehydration

Isobutanol final purification is conventionally carried out in two steps of distillation (Fig. 1). Although isobutanol has a higher boiling point than water, the presence of a minimum boiling point azeotrope allows obtaining isobutanol at compositions close to the azeotropic point (65.6 wt%) in the pre-concentration column (C1). Water/isobutanol azeotrope was broken in a decanter. The organic phase was fed on top of the depletion column (C2). Isobutanol (99.9 wt%) was obtained in the bottoms of column C2. Aqueous phase was recirculated on top of column C1. The feed flow was pumped to stage 3 of column C1 when the isobutanol concentration in feed flow was lower than 6.5 wt%. Otherwise, the feed flow was directly pumped to the decanter.

System I (S-I) combined VCD with high-pressure distillation (Fig. 2). The column C1, see Fig. 2, was integrated with vapor compression. The column C2 operates at a pressure required for supplying the condensation heat to reboiler of column C1. The condensation heat of column C2 in some cases, depending on the concentration of feed flow, was higher than the boiling requirement of column C1. In these cases, the excess of steam in the top of column C2 was compressed to supply heat to its reboiler. The fed flow of decanter must cooled. Therefore, the exit of decanter was preheated with the fed of decanter. The trays number of columns C1 and C2 were 26 and 10, respectively. The total energy consumption of the process included compressor power and the steam of the reboilers of columns C1 and C2.

Process S-II was a less intensive energetic integration choice than S-I and it had three distillation columns. The feed flow was divided and pumped to columns C1-LP and C1-HP (Fig. 3). Colum

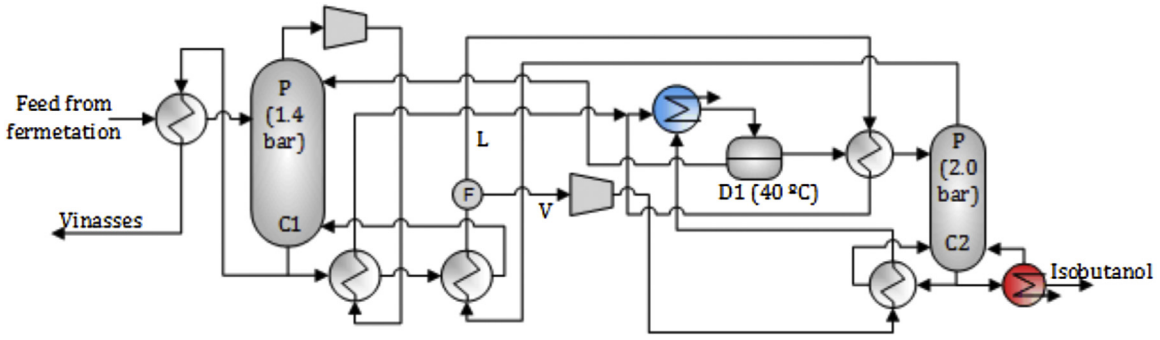

Fig. 2. Isobutanol dehydration by vapor compression distillation (S-I).