



Contents lists available at ScienceDirect

Chemical Engineering Research and Design

journal homepage: www.elsevier.com/locate/cherd

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Conceptual design for the recovery of 1,3-Butadiene and methyl ethyl ketone via a 2,3-Butanediol-dehydration process

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ARTICLE INFO

Article history:

Received 22 February 2017

Received in revised form 12 May 2017

Accepted 19 May 2017

Available online 28 May 2017

Keywords:

2,3-Butanediol

1,3-Butadiene

Methyl ethyl ketone

Dehydration

Azeotropic-distillation column

Process design

ABSTRACT

This paper proposes a process scheme for the recovery of 1,3-Butadiene (BD) and methyl ethyl ketone (MEK) from the dehydration products of 2,3-Butanediol (BDO). BDO is produced via the bio-fermentation of the industrial-gas wastes from the steel industry and various biomasses. The suggested process is environmentally beneficial because it can extend the feedstock rather than petroleum, and it replaces the use of the industrial-plant gas wastes. The proposed process is composed of a catalytic-dehydration reactor, a quencher, a BD-purification unit, a decanter, a water-removal column, and an MEK-purification unit. Based on the proposed scheme, the BD- and MEK-recovery rates are approximately 94% and 98%, respectively. Most of the units in the process are well developed, except for the azeotropic distillation column (ADC) that is used in the MEK-purification unit. A pilot test is also performed, and the results are compared with those of a simulation for an analysis of the viability of an ADC. Through the use of the experimental ADC, a mixture of MEK and 2-methylpropanal (2-MPA) is obtained via the removal of water. The results of the pilot test show that a water trace, MEK 84.5%, and 2-MPA 15.5% are separated to the bottom stream, which is in sound agreement with the simulation data. Furthermore, the simulated temperature profile is consistent with the experimental data with an error range of only $\pm 2\%$.

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1. Introduction

1,3-Butadiene (BD) and methyl ethyl ketone (MEK) are important industrial chemical products. BD is mainly used as a synthetic rubber for the production of automobile tires. Also, the demand for acrylonitrile butadiene styrene (ABS), which is a high-performance plastic that is produced from BD, is expected to increase in the automobile industry. The global BD market is expected to grow to USD 33.01 billion and 14.179 million tons by 2020 ("Global 1,3 Butadiene (BD) Market By Application (SBR, Butadiene Rubber, SB Latex, ABS, HMDA, NBR) Expected To Reach USD 33.01 Billion By 2020: Grand View Research, Inc," 2015; White, 2007). The application of MEK is mostly regarding organic solvents that are

used for coating, painting, and peroxide synthesis, as well as in the pharmaceutical industry. The MEK market is estimated to increase to USD 3.26 billion and to over 1.754 million tons by 2020 (Liu et al., 2006; "Methyl Ethyl Ketone (MEK) Market Analysis By Application (Paints & Coatings, Printing Inks, Adhesives) And Segment Forecasts To 2024," 2016; Nikitina et al., 2016).

In terms of the production of BD and MEK, several environmental, economical, and technical issues remain. The raw materials of the two compounds are derived from petroleum, and their production can be harmful to soils, surface ground water, and ecosystems (Kharaka and Dorsey, 2005; Spooner, 2012). Also, petroleum is not a sustainable resource, as its price fluctuates depending on the market (Haveren et al.,

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<http://dx.doi.org/10.1016/j.cherd.2017.05.019>

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Nomenclature

ABS	Acrylonitrile butadiene styrene
ACM	Activity coefficient model
ADC	Azeotropic distillation column
ATP	Adenosine triphosphate
BD	1,3-Butadiene
BDO	2,3-Butanediol
BTM	Bottom
CH	Cyclohexane
DC	Distillation column
EOS	Equation of state
FC	Flow controller
FID	Flame ionization detector
GC	Gas chromatograph
HETP	Height equivalent to theoretical plate
LLE	Liquid liquid equilibrium
LT	Level transmitter
MEK	Methyl ethyl ketone
NADH	Reduced nicotinamide adenine dinucleotide
NRTL	Non-random two-liquid model
O/H	Overhead
PBR	Pebble-bed reactor
PT	Pressure transmitter
SBA	Secondary butyl alcohol
SV	Solenoid valve
WHSV	Weight hourly space velocity, [1/h]
VLE	Vapor liquid equilibrium
VLE	Vapor liquid liquid equilibrium
2-MPA	2-Methylpropanal
3B2OL	3-Buten-2-ol

2008). In the steam-cracking process for BD, the operation of the cracking furnace requires high temperatures and enormous energy levels are therefore consumed (Ren et al., 2006). For the MEK-production process, the olefin hydration requires an H_2SO_4 solution that is higher than 75 wt%, and this can cause serious health problems; moreover, an exposure to the excess 2-butanol levels that are from industrial processes can cause fatigue, dizziness, and eye irritations (Hahn et al., 2013).

BDO (2,3-Butanediol) is a potential intermediate compound for the production of hydrocarbons. BDO can be produced via a fermentation of the industrial waste from the steel industry and biomasses such as molasses, cereal mashes, starch, wheat, sulfite-waste liquor, and corn starch, and this is achieved with the use of a number of micro-organisms such as *Aerobacter aerogenes*, *Aeromonas hydrophila*, *Aerobacillus polymyxa*, *Bacillus polymyxa*, and *Klebsiella* (Afschar et al., 1991; Garg and Jain, 1995; Ji et al., 2008; Köpke et al., 2011; Ma et al., 2009; Petrov and Petrova, 2009; Rodwell and Lafayette, 1963; Syu, 2001; Ui et al., 2004). However, the BDO production that is achieved through bio-fermentation is in an initial stage of development at the industrial scale (Celińska and Grajek, 2009). Also, this method is limited in the laboratory setting wherein the dehydration of BDO to MEK and BD has been studied with only the use of suitable catalysts such as bentonite clay, metal and earth oxides, zeolites, a perfluorinated resin with sulfonic-acid groups, heteropoly acids, calcium phosphates, and Cs/SiO_2 (Bourns and Nicholls, 1947; Bucsi et al., 1994; Duan et al., 2015, 2014; Han et al., 2013; Kannan and Pillai, 1969; Kim et al., 2014, 2015; Lee et al., 2000; Molnár et al., 1988; Nikitina and Ivanova, 2016; Tsukamoto et al., 2016; Winfield, 1950a,b; Zhang et al., 2012). In addition, a recovery method does not exist for the products that are obtained through the dehydration of BDO.

In this paper, a conceptual industrial-scale design is suggested for the recovery of highly pure BD and MEK that is achieved through the dehydration of the BDO that is produced from bio-fermentation. The process consists of a catalytic reactor, a quencher, a BD-purification

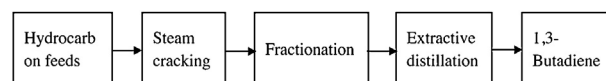


Fig. 1 – Conventional process for BD production.

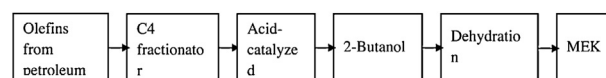


Fig. 2 – Conventional process for MEK production.

unit, a decanter, a water-removal unit, a wastewater-treatment unit, and an MEK-purification unit. The other equipment except for ADC in the proposed process are commercially used or are simple distillation column for separating components having high relative volatility, compressor, heat exchangers, and pumps. A pilot test for which cyclohexane (CH) was employed as an entrainer was conducted to validate the performance of the ADC, and both the experiment and simulation results are well matched below a $\pm 2\%$ deviation.

2. Theoretical background

2.1. Conventional process

2.1.1. BD process

Steam cracking is normally used to produce BD, as shown in Fig. 1. BD is one of the by-products of the ethylene production that is from steam cracking. The corresponding feedstocks (C_2 – C_4 , naphtha, and oil) are cracked at around 850°C . Hydrogen, ethylene, propylene, butadiene, and olefin are produced from the steam-cracking furnace, and they go through a quencher, a compressor, and a dryer. During this process, the hydrocarbons with more than four carbons ($\text{C}-5$ and higher components) are removed, and the BD is purified via an extractive distillation. Typically, through one or two extractive distillations, the C_4 raffinate and the vinyl acetylene are eliminated. Thereafter, the main stream produces the BD through a fractionator to remove the methyl acetylene (White, 2007).

2.1.2. MEK process

A commercial process is illustrated in Fig. 2 for the MEK production that is achieved through the hydration of butylene, secondary butyl alcohol (SBA) production, and SBA dehydration. Butylene is produced from commercial-scale petroleum cracking. SBA can be obtained from an acid-catalysis-based hydration of 1-butene and 2-butene. The hydration process is conducted at approximately 250°C , and it requires a 75% H_2SO_4 solution; specifically, the 1-butene/2-butene are reacted in a reactor and transported to a distillation column that separates the unreactants from the 2-butanol. In addition, the SBA is transported to an intermittent distillation column. The produced SBA undergoes dehydration, and the MEK is produced practically (Song, 2016).

2.2. BDO and the BDO-dehydration process

2.2.1. BDO (2,3-Butanediol)

BDO is a platform chemical and fuel that can be used for the production of fine chemicals and antifreeze and in the food industries. BDO is produced via the fermentation of biomass-based xylose and glucose, as shown in Fig. 3. Pentose and hexose are the intermediate compounds of BDO; that is, through glycolysis and reduction, they can be converted into BDO. Glycolysis is a biochemical route which converts glucose or xylose into pyruvate. Through the process, ATP

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