Journal of Process Control 21 (2011) 1193–1207

Contents lists available at ScienceDirect

Journal of Process Control

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

Design and control of reactive-distillation process for the production of diethyl carbonate via two consecutive trans-esterification reactions

Hon-Yu Wei[a], Anisatur Rokhmah[b], Renanto Handogo[b], I-Lung Chien[c,*]

[a] Department of Chemical Engineering, National Taiwan University of Science and Technology, Taipei 106, Taiwan
[b] Department of Chemical Engineering, Sepuluh November Institute of Technology (ITS), Jawa Timur, Indonesia
[c] Department of Chemical Engineering, National Taiwan University, Taipei 106, Taiwan

ARTICLE INFO

Article history:
Received 22 February 2011
Received in revised form 8 May 2011
Accepted 16 June 2011
Available online 23 July 2011

Keywords:
Diethyl carbonate
Trans-esterification reaction
Reactive-distillation
Optimal design
Control strategy

ABSTRACT

A reactive-distillation process to produce diethyl carbonate (DEC) and methanol (MeOH) from dimethyl carbonate (DMC) and ethanol (EtOH) via two consecutive trans-esterification reactions is proposed in this study. In the design study, an unusual feed location design of the reactive-distillation column is needed to suppress the formation of the intermediate (ethyl–methyl carbonate, EMC) while achieving near complete conversion of the limiting reactant. The heavier DMC feed should be located at the bottom of the reaction section, while the lighter EtOH feed should be located at the top part of the reaction section. The comparison of the TAC of this proposed design to that of a traditional process will be given which shows great benefit to utilize the reactive-distillation technology. Finally, a simple control strategy is proposed with only one temperature loop in each column. The proposed control strategy works effectively to hold the product compositions despite feed flow rate and feed composition changes.

1. Introduction

Diethyl carbonate (DEC) is an important solvent and an intermediate for preparing norfloxacin, a broad-spectrum antibiotic, previously made by using phosgene [1]. It is also a reactant used in the manufacture of fertilizers, pesticides, dyes and polymers [2]. In addition, DEC can also be used as the electrolyte of lithium ion battery [3]. The reaction system consists of two consecutive reversible reactions with the intermediate product of ethyl–methyl carbonate (EMC). In this paper, reactive-distillation technology will be explored for the production of this important chemical.

In a book presenting the status and future directions of reactive-distillation (cf. Sundmacher and Kienle [4]), a survey of chemical reaction systems that performed successfully in reactive-distillation columns is given. In Table 1.1 and 1.2 of the Sundmacher and Kienle's book, over one hundred industrially or potentially important reactions for reactive-distillation applications are given. An updated literature survey from Luyben and Yu [5] shows a total of 236 reaction systems in the Appendix of this recent book. This illustrates the importance of this technology in industrial applications.

Because these reactive-distillation processes combine reaction and separation in a single vessel, the operation and control of these processes become more difficult than for conventional columns. In Luyben and Yu [5], control of various reactive-distillation processes are discussed including ideal systems of $A + B \Leftrightarrow C + D$, $A + B \Leftrightarrow C$ and $A \Leftrightarrow B + C$. Several real systems have also been discussed in the book including reactive-distillations for acetic acid esterification, TAME, MTBE and ETBE reactive-distillation systems.

Most reactive-distillation systems studied in the literature focused on the above $A + B \Leftrightarrow C + D$, $A + B \Leftrightarrow C$ and $A \Leftrightarrow B + C$ reaction type. The reaction type of obtaining the final products via two consecutive trans-esterification reactions has rarely been studied. Previous studies on this system only focused on the design aspect for one product stream obtaining the DEC product alone. For example, Luo and Xiao [1] designed a reactive-distillation column with DEC as bottom product. However, the top product may consist of MeOH, EtOH and other components. Mueller and Kenig [6] designed a dividing wall reactive-distillation column with DEC as bottom product and EtOH as a sidedraw product. However, the top product is an azeotropic mixture of MeOH and DMC. The same research group also designed a flowsheet including a pre-reactor and a dividing wall reactive-distillation column. The bottom and the top products are pure DEC and MeOH, respectively. However, the sidedraw product consisted of EtOH, DMC and EMC needing further purification. Furthermore, the dynamic and control aspect of this reactive-distillation process has not been studied before.

In this paper, an overall design flowsheet of this process including recycle stream will be developed with products of DEC and MeOH at their high-purity specifications. This designed flowsheet will be optimized in terms of total annual cost (TAC). The economics of this reactive-distillation process will also be compared with a

* Corresponding author. Tel.: +886 3 3366 3063; fax: +886 2 2362 3040.
E-mail addresses: ilungchien@ntu.edu.tw, ilungchien@hotmail.com (I.-L. Chien).

0959-1524/$ – see front matter © 2011 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jprocont.2011.06.006