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Heat-integrated triple-column pressure-swing distillation process with multi-recycle streams for the separation of ternary azeotropic mixture of acetonitrile/methanol/benzene

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Abstract: The steady-state economics and dynamic controllability of heat-integrated triple-column pressureswing distillation (TCPSD) process are explored by taking the separation of ternary mixture of acetonitrile, methanol and benzene as demonstrating example. The performance evaluation indicators of second-law efficiency and CO₂ emissions are employed to rank different arrangements except only reference of total annual cost (TAC). Compared to the conventional process, the economically optimum flowsheets are the partially heat-integrated processes (Cases 1 and 2) since it can reduce about 20.00% in TAC, 35.19% in energy-saving and enhancement of 48.77% in thermodynamic efficiency. Dynamic control of the economic efficient processes are also investigated. A series of control structures are developed and assessed by the throughput and feed composition disturbances, which are divided to two categories including control schemes with or without split ratio of the distillate stream of second column. The *snowballing* effect is efficiently attenuated by control schemes (CS4 and CS5) with the ratio strategy to adjust the recycle stream flowrate for partial Case 1. These control strategies are also applied to partial Case 2, the stable regulatory control is achieved, yet *oscillation* phenomena for the response of condenser heat removal of second column exists for control strategies without split ratio scheme, while it is efficiently handled by split ratio scheme coupling with pressure compensated temperature-composition cascade control strategy. **Keywords:** Pressure-swing distillation; Heat integration; Snowball effect; Plantwide control; Ternary mixture

1. Introduction

The separation of compounds with azeotrope or close-boiling mixture is a rigorous challenging, inevitable and intriguing project for the chemical and petrochemical industries. The existence of this phenomena makes the separation inefficiency or infeasible by conventional simple distillation. Some enhanced strategies had been employed, for example, pervaporation, adsorption, pressure-swing distillation (PSD), extractive distillation (ED) and homogeneous or heterogeneous azeotropic distillation, as well as hybrid methods combining these options ^[1-6]. Pervaporation is an energy efficient method for separating close-boiling mixture or azeotrope with the assistance of membrane. However, the restrictions can be reached in terms of large scale separations. Adsorption operation became recently more popular since it had more energy efficiency than distillation, while membrane regeneration was such difficult that it was not suitable for industrial production ^[2, 7]. Distillation methods, such as PSD and ED, present relatively high energy costs, however, they are still the options for large scale production ^[3, 8].

PSD is one of viable strategies, the subject of this article, for separating azeotropes when their azeotropic compositions are pressure-sensitive ^[9]. The one of properties of this approach is that it can avoid the potential problem of product contamination by inserting extra component, which might be the inherent characteristics of ED process. For PSD, it is feasible for separating either minimum- or maximum-boiling azeotropes. Currently, the exploration of PSD was mainly focused on the binary system. Two columns were operated at different pressures. High-purity product streams were taken out from one-end of columns, and the other end was the recycle stream whose composition was near their azeotropes. There are many literatures focusing on investigating the separation *Corresponding author: Aiwu Zeng: E-mail: awzeng@tju.edu.cn.

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