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Methodology for design and analysis of reactive distillation involving multielement systems

Amnart Jantharasuk[a], *Rafiqul Gani*[b,*], *Andrzej Górak*[c], *Suttichai Assabumrungrat*[a,**]

[a] *Center of Excellence in Catalysis and Catalytic Engineering, Department of Chemical Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok 10330, Thailand*

[b] *CAPEC, Department of Chemical and Biochemical Engineering, Technical University of Denmark, DK-2800 Lyngby, Denmark*

[c] *Department of Biochemical and Chemical Engineering, TU Dortmund University, Emil-Figge-Str. 70, 44227 Dortmund, Germany*

ABSTRACT

A new methodology for design and analysis of reactive distillation has been developed. In this work, the element-based approach, coupled with a driving force diagram, has been extended and applied to the design of a reactive distillation column involving multielement (multicomponent) systems. The transformation of ordinary systems to element-based ones and the aggregation of non-key elements allow the important design parameters, such as the number of stages, feed stage and minimum reflux ratio, to be determined by using simple diagrams similar to those regularly employed for non-reactive systems consisting of two components. Based on this methodology, an optimal design configuration is identified using the equivalent binary-element-driving force diagram. Two case studies of methyl acetate (MeOAc) synthesis and methyl-*tert*-butyl ether (MTBE) synthesis have been considered to demonstrate the successful applications of the methodology. Moreover, energy requirements for various column configurations corresponding to different feed locations are determined to verify whether the optimal design can be identified by following the proposed methodology.

Keywords: Reactive distillation; Design methodology; Multielement-based approach; Driving force diagram

1. Introduction

Reactive distillation (RD) is a promising multifunctional reactor to improve an ordinary process, as chemical reaction and thermodynamic separation are combined in a single unit. It offers substantial advantages, such as higher reaction rate and selectivity, avoidance of azeotropes and reduced energy consumption as well as solvent usage (Lee et al., 2010; Thotla and Mahajani, 2009; Malone and Doherty, 2000). The impact of this technology to industrial applications was extensively summarised (Sharma and Mahajani, 2003). Although the RD process has received widespread attention in recent years, the development of reliable design tools covering a wide range of applicabilities (Toikka et al., 2009) is a topic of current research. Furthermore, the determination of optimum column configuration has become a great challenge for today's design and optimisation techniques.

For RD column, the experiment-based approach (Mueanmas et al., 2010; De Lima Da Silva et al., 2010; Thompson and He, 2007; He et al., 2005), simulation-based approach (Luo et al., 2009; Sláva et al., 2009) and graphical-based approach (Groemping et al., 2004; Lee and Westerberg, 2000; Lee et al., 2000a,b,c; Ung and Doherty, 1995a,b; Barbosa and Doherty, 1988a) may be considered as three main available design categories. The distinctive advantages provided by the experiment-based design are to visually observe real phenomena as well as explicit behaviours during the operation. Nevertheless, to achieve the optimum column configuration, this method is costly and time-consuming. On the other hand, the works on the simulation-based approach are less expensive, but may require complicated modelling as well as large computational effort. For the graphical-based approach, the design can be visually performed via a simple visual diagram, which is constructed under reasonable

* *Corresponding author.*
** *Corresponding author.* Tel.: +66 2 2186868; fax: +66 2 2186877.
E-mail addresses: rag@kt.dtu.dk (R. Gani), suttichai.a@chula.ac.th (S. Assabumrungrat).
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Nomenclature

G	total Gibbs energy of a system containing
n_i	molar amount of component i
μ_i	component chemical potential i
NP	number of phases
NC	number of components
M	number of elements
NR	number of reactions
b_j	element flowrate of element j
$A_{j,i}$	the number of times the reaction invariant element j presented in molecule i
B	component molar flowrate
L	liquid phase component molar flowrate
V	vapour phase component molar flowrate
x_i	liquid phase molar fraction
y_i	vapour phase molar fraction
W	element fraction
DF	amount of driving force (means to equivalent binary element driving force for this work)
R	element reflux ratio
S	element boilup ratio
α	equivalent binary element relative volatility

Superscripts

β	phase of the concerning system (liquid or vapour)
l	liquid phase
v	vapour phase

Subscripts

R	rectifying section
S	stripping section
F	feed stream
D	distillate stream
B	bottom stream

assumptions. Therefore, many researchers have paid attention to the development of this approach for the RD design. Groemping et al. (2004) proposed a method involving stage composition lines for a single-feed RD column. This method can provide feasible alternatives for both entirely reactive and hybrid column configurations. Lee and Westerberg (2000) presented the design of an RD column for a ternary isomolar decomposition reaction where the identification of the reactive zone is dependent on how the reaction equilibrium curve lies comparative to the bubble point curve. Other techniques (Lee et al., 2000a,b,c) involving the extension of classical design tools, McCabe–Thiele diagram and Ponchon–Savarit method, were proposed to handle the design problems of RD columns. Barbosa and Doherty (1988a,b) developed a technique relying on the concept of a transformed composition variable (Ung and Doherty, 1995a,b) for designing single-feed and double-feed RD columns (Barbosa and Doherty, 1988a,b). The main feature of these techniques is to treat the reactive systems in analogy with the simple system without reactions. The two simple methods, the McCabe–Thiele- and the Ponchon–Savarit-type diagrams, were extended to design RD columns, based on the element mass balance concept (Sánchez Daza et al., 2003). The reactive systems containing binary element or, at least, ternary, in terms of mixture compounds, were considered. Moreover, the driving force approach (Gani and Bek-Pedersen, 2000; Bek-Pedersen and Gani, 2004) was extended to include reactive systems. The use of this concept allows the identification of optimal feed location, where the maximum driving force presents, to obtain an optimum RD column with minimum reboiler duty. While maintaining substantial advantages, the drawbacks of transformed variables in RD design are overcome because the element composition and flow rate are always positive values. Furthermore, the dimension of governing equations is reduced because the number of elements is always lower than or equal to the number of components (Pérez Cisneros et al., 1997). As previously discussed, even though the graphical design for RD column is promising and a number of design concepts have been developed, the number of components (elements) in the system is limited. This definitely hinders the progressive advancement of graphical design methods and tools for RD column, as inert compounds are commonly presented in industrial operations. Consequently, an effort to overcome this limitation is imperative to expand the applicability of the graphical design.

Therefore, the objective of this proposed work is to introduce a new design methodology, related to a graphics-based approach, for the RD columns involving multielement (also implicitly referred to as multicomponent) systems. The methodology starts with the identification of advantages for replacing the conventional system by the RD process. Then, the system is analysed based on pure component properties, mixture properties, reaction analysis and separation analysis. Next, the components presented in the system are represented by elements, forming the multielement system. Using a procedure developed by Hengstebeck (Malone and Doherty, 2000) and selecting a pair of key elements, an equivalent binary (key) element system is created. Then, the reactive driving force diagram for this binary element system is generated and used to design a RD column using the largest possible driving force. Next, a rigorous simulation of a multicomponent (multielement) system is carried out to verify the column design obtained from the multielement-based driving force diagram and to fine-tune the design. Note that the design is expected to represent, at least, a near-optimal solution with respect to energy consumption (reboiler duty). For the design task, the entire system is transformed into equivalent binary element basis. The incorporation of the reactive driving force method allows the investigation of the energy efficient design without any complicated optimisation techniques. Important design parameters, such as the number of stages, feed location and the minimum reflux ratio are determined from a simple diagram, similar to a general McCabe–Thiele method, which is derived from the corresponding reactive driving force diagram. This procedure offers the advantage in that the studied reactive system is to be considered as though no reaction is taking place. The methodology has been demonstrated in a systematic manner with two case studies: methyl acetate (MeOAc) and methyl-*tert*-butyl ether (MTBE) syntheses. In addition, the reboiler duties required for various feed locations have been determined to verify that the optimal column configurations that require the minimum energy consumption are identified through this work. It should be noted that although only the examples of single chemical reaction are presented, this design methodology is also applicable for the systems containing a set of multiple reactions. Nevertheless, the effort may rapidly increase for systems with greater number of elements, as the limiting non-key element compositions have to be determined corresponding to a larger group of pos-