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Optimal design of reactive distillation systems: Application to the production of ethyl *tert*-butyl ether (ETBE)

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ABSTRACT

This work addresses the design of reactive distillation columns to produce ETBE, based on a detailed first-principles model that considers equilibrium and kinetic information, rigorous physical property data, and catalyst deactivation. An evolutionary algorithm is used to generate a sequence of feasible designs with improved characteristics in a sequential solution/optimisation strategy, by specifying the design variables (both integer and continuous) that characterise a particular column configuration. Two classes of optimisation algorithms are compared: genetic algorithms and particle swarm optimisation. The objective function considered is the gross annual profit.

The results demonstrate that both algorithms are adequate to solve this design problem. The effect of catalyst deactivation included in the design stage played a determinant role in the optimal column specification. A post-design sensitivity analysis is developed to assess the quality of the solutions obtained, together with the individual effects of each design variable in the optimal configuration identified.

1. Introduction

Reactive distillation (RD) is a process intensification solution that combines reaction and separation in the same vessel (Luyben & Yu, 2008; Stankiewicz, 2003). Even though reactive distillation is not new, in the last two decades the scientific and industrial interest in this technology has risen dramatically. One of the major reasons that contributed to this renewed attention is the high energy prices, which emphasise the need for more energy-efficient processes. An additional cause is also the publication of a number of encouraging case-studies, such as the Eastman Chemical success story, where RD was applied to the production of methyl acetate, reducing by a factor of 5 the energy consumed in the conventional process, and also by a factor of 5 the capital investment of the new RD-based process (Agreda, Partin, & Heise, 1990; Siirola, 1996; Sundmacher & Kienle, 2003).

Reactive distillation has been successfully applied to the production of gasoline oxygenated-additives, such as methyl *tert*-butyl ether (MTBE) and more recently both ethyl *tert*-butyl ether (ETBE) and *tert*-amyl methyl ether (TAME) (Luyben & Yu, 2008). These applications are possible since these systems are equilibrium-controlled, and both reaction and separation can be carried out using mild and similar operating conditions. Since the reactions are exothermic, the reaction heat can be used for liquid vapourisation, thus increasing reboiler energy savings (Sundmacher & Qi, 2003; Tuchlenski et al., 2001).

ETBE is mainly produced from the reaction of isobutene with ethanol, catalysed by acid ion-exchange resins. In the conventional process, the reaction takes place after a pretreatment stage, in two series reactors. Commercial ETBE is then obtained after a separation section, which comprises two distillation columns and a liquid-liquid extraction column (Domingues, Pinheiro, Oliveira, Fernandes, & Vilelas, 2012; Hamid & Ashraf, 2004).

Due to the presence of chemical reactions, the design of RD columns becomes more complex than conventional units. More attention should be paid to variables such as the tray holdup (or the column diameter, in the case of heterogeneous catalysis), since they affect the extent of the reaction (Taylor & Krishna, 2000). In the case of heterogeneous catalysis, the catalyst will be present in some kind of support; structured packings for RD (*e.g.*, the Sulzer KATAPAK-SP) are available on the market. Other design variables, such as the number and location of reactive and non-reactive trays, feed location(s), pressure and reflux ratio are also important in this case.

According to Almeida-Rivera and Grievink (2004), the design methods for RD available in the literature can be divided in three main categories: graphical/topological, optimisation-based and heuristics-based.

The restrictive assumptions of the graphical/topological methods make them suitable for feasibility studies but inefficient for

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Nomenclature

Symbols

A	cross-section area, m^2
a	activity
A'	lateral area, m^2
B	cost coefficient
C	concentration, mol/m^3
c	centroid
C^*	cost, M€/year
D	liquid distillate molar flowrate, mol/s
d	diameter, m
E_a	activation energy, J/mol
F	feed molar flowrate, mol/s
f_{factor}	vapour load factor, $Pa^{0.5}$
H	height, m
h	specific enthalpy, J/mol
I	chemical engineering plant cost index
k	partition coefficients
$k_{0,d}$	pre-exponential factor for deactivation, s^{-1}
$K_{ad,EtOH}$	adsorption constant for EtOH
$K_{ad,TBA}$	adsorption constant for TBA, m^3/mol
$K_{eq,ETBE}$	equilibrium constant for ETBE formation
$K_{eq,TBA}$	equilibrium constant for TBA formation, m^3/mol
$k_{rate,DIB}$	kinetic constant for DIB formation, mol/(kg s)
$k_{rate,ETBE}$	kinetic constant for ETBE formation, mol/(kg s)
$k_{rate,TBA}$	kinetic constant for TBA formation, $m^6/(kg\,mol\,s)$
K_r	sorption constants ratio of EtOH and IB
L	liquid molar flowrate, mol/s
L'	liquid mass flowrate, kg/h
l_t	vessel thickness, m
M	molecular weight, kg/mol
m	mass per stage, kg
N	number
P	absolute pressure, bar
Q	heat exchanged, W
R	ideal gas constant, J/(mol K)
r	reactive stage vector (binary vector with size of N_S and 1 at the position of each reactive stage, otherwise 0)
$\mathcal{R}$	rate of reaction, mol/(kg s)
R^*	revenue, M€/year
T	temperature, K
t	time-on-stream, s
T_s	tray spacing, m
U	heat transfer coefficient, $W/(m^2K)$
V	vapour molar flowrate, mol/s
V'	vapour mass flowrate, kg/h
x	liquid molar composition
y	vapour molar composition
z	normalised design variable
z'	design variable
HETP	height equivalent to a theoretical stage, m
NTSM	number of theoretical stages per meter, m^{-1}
Diversity	population diversity
Obj	objective function, M€/year
Penalty	penalty value, M€/year
Price	price of feedstock or product, €/ton
RR	reflux ratio

Greek letters

α	deactivation decay order
β	conversion factor
ΔH_V	enthalpy of vapourisation, J/kg
ΔP	pressure drop per stage, bar
ΔT	temperature difference, K
γ	cost factor
ν	stoichiometric coefficient
Φ	deactivation factor
ρ	density, kg/m^3

Subscripts, superscripts and abbreviations

bot	bottom
C	components
cat	catalyst
col	column
cond	condenser
d	deactivation
\$/*CHF*	Dollar per Swiss franc
d/y	days per year
desvar	design variables
DIB	diisobutene
€/\$	Euro per dollar
ETBE	ethyl *tert*-butyl ether
EtOH	ethanol
F	feed
h/d	hours per day
i	index for components
IB	isobutene
ind	individuals/particles
j	index for stages
$k\,m$	summation indexes
ton/*kg*	ton per kilogram
L	liquid
max	maximum
min	minimum
MTBE	methyl *tert*-butyl ether
pack	packing
R1	inlet of reactor section
Raf1	raffinate I
reac	reactions
reb	reboiler
Rx	reactive stages
q	index for reactions
S	stages
s/h	seconds per hour
shell	shell
steam	steam
surr	surroundings
TBA	*tert*-butyl alcohol
top	top
tray	tray
V	vapour
bare	bare tubes

a more detailed column design. Heuristics-based methods are more useful for post-design analysis, as the configuration of RD columns varies greatly with the chemical system chosen (Almeida-Rivera & Grievink, 2004). Thus, optimisation techniques, which are mathematical-based and involve both physical constraints (*e.g.*, material balances and thermodynamic relationships), as well as economic information (capital investment and operating costs), appear to be the most flexible and rigorous approach to identify the optimal RD column configurations (Almeida-Rivera & Grievink, 2004). This broad class of methods can be further divided into deterministic and evolutionary (or stochastic) strategies (Ryoo & Sahinidis, 1995). Deterministic-based methods are commonly