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Dynamics and control of benzene hydrogenation via reactive distillation

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ABSTRACT

This work develops a dynamic, first principles-based model of a reactive distillation column used for benzene hydrogenation of a reformate stream and investigates different control structures for this process. The model is used initially to develop and evaluate a feedback control strategy which provides good regulatory performance for small disturbances, however, it tends to be sluggish for significant disturbances in the feed composition. In order to address this point, adding a feedforward controller to the feedback structure has also been investigated. However, the feedforward controller can only be implemented if composition measurements of the feed are taken. As online composition measurements are expensive in practice, several different scenarios have been investigated where samples of the feed are taken and subsequently analyzed in a lab, as represented by measurement time delays. Simulation results show that adding feedforward control to the feedback scheme can be very beneficial for this process, however, this is only the case if the composition disturbance measurements do not involve a significant time delay.

1. Introduction

Automotive emissions are a significant contributor to poor air quality [1]. As such, specifications for automobile fuels obtained from petroleum have received increasing levels of attention from the Environmental Protection Agency (EPA). Benzene is one of the compounds that is regulated as it is a carcinogen and the EPA requires all refiners to limit the amount of benzene in gasoline to 0.62 vol% [2]. While benzene in the gasoline pool results from a variety of sources, the main contributor is the reformer unit resulting in significant amounts of benzene present in reformate streams. As the reformate stream is used to boost octane rating, there are economic objectives that have to be taken into account while complying with environmental regulations.

One option to remove benzene is to hydrogenate in the presence of a catalyst. However, a problem arises as the catalyst used for the reaction is not exclusively selective for benzene, and toluene, which is present in the reformate stream in considerable quantities, will also be hydrogenated. Toluene hydrogenation is undesirable as toluene has a high octane rating (RON) and should be retained in the final product.

$$\text{Benzene}\,(100\,\text{RON}) + 2\text{H}_2 \rightarrow \text{cyclohexane}\,(83\,\text{RON}) \tag{1}$$

$$\text{Toluene}\,(120\,\text{RON}) + 3\text{H}_2 \rightarrow \text{methylcyclohexane}\,(75\,\text{RON}) \tag{2}$$

In order to avoid problems related to the selectivity of the catalyst, the reformate stream is split into light and heavy components in the conventional process (Fig. 1a). As benzene is a reasonably light component of this mixture, it is mostly concentrated in the distillate, and accordingly, is hydrogenated before being sent to the gasoline pool. The downside of this process is that a high capital investment is needed. Reactive distillation (Fig. 1b) offers an alternative route for solving this problem. By combining reaction with separation it is possible to selectively react one component in a specified region of the column while suppressing unwanted reactions of other components. Furthermore, additional savings can be achieved as the heat of reaction can directly be used for separation of the mixture.

While reactive distillation (RD) can have significant advantages over traditional designs, there are also challenges that need to be considered. The simultaneous presence of reaction and separation phenomena can result in complex dynamic behavior. Combining reaction and separation into a single vessel results in fewer manipulated variables, thus increasing interactions between control loops [3]. RD columns have been observed to be very sensitive to changes in feed concentration. This is a crucial aspect

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Notation

a	geometric surface area of packing per unit volume ($m^2\,m^{-3}$)
A	cross sectional area of column
C_A	concentration ($mol\,m^{-3}$)
d_p	packing particle diameter (m)
d_s	column diameter (m)
D	distillate flow rate ($mol\,s^{-1}$)
E_a	reaction activation energy ($J\,mol^{-1}$)
F	feed flow rate ($mol\,s^{-1}$)
h_i	total liquid holdup based on empty column ($m^3\,m^{-3}$)
H_{l_j}	molar enthalpy of liquid stream on stage j ($J\,mol^{-1}$)
H_{v_j}	molar enthalpy of vapor stream on stage j ($J\,mol^{-1}$)
HETP	height equivalent to a theoretical stage (m)
k	reaction rate constant ($mol\,s^{-1}\,kg^{-1}$)
K	wall factor
K_A	reaction adsorption coefficient ($m^3\,mol^{-1}$)
K_H	reaction adsorption coefficient ($m^3\,mol^{-1}$)
L	liquid flow rate ($mol\,s^{-1}$)
M	mass holdup (kg)
M_{l_j}	liquid molar holdup on stage j (mol)
M_{v_j}	vapor molar holdup on stage j (mol)
N	number of stages
P	pressure (Pa)
$\Delta P_{0,j}$	dry column pressure drop across stage j (Pa)
ΔP_j	irrigated column pressure drop across stage j (Pa)
Q	external heat energy input (J)
R	reflux ratio
R_{gas}	gas constant ($J\,mol^{-1}\,K^{-1}$)
Re_v	vapor Reynolds number
rxn	reaction rate ($mol\,s^{-1}\,kg^{-1}$)
s	Laplace variable
T_0	reaction reference temperature (K)
T_j	temperature on stage j (K)
V	vapor flow rate ($mol\,s^{-1}$)
u	specific liquid load ($m\,s^{-1}$)
x	liquid mole fraction
y	vapor mole fraction
y^*	equilibrium vapor mole fraction
z	feed mole fraction
Greek letters	
ε	packing void fraction
$\phi_{l_{j,i}}$	liquid fugacity coefficient of component i on stage j
$\phi_{v_{j,i}}$	vapor fugacity coefficient of component i on stage j
ρ_{cat}	catalyst density ($kg\,m^{-3}$)
η	Murphree efficiency
λ	relative gain
Λ	relative gain array
τ	transfer function time constant (s)
τ_c	controller design parameter (s)
θ	transfer function time delay (s)
ψ	resistance coefficient
Subscripts	
i	component index
j	stage index

for benzene hydrogenation as the concentration of some of the main components in the feed can vary by 50% or more due to disturbances upstream from the column [4]. The importance of addressing these disturbances is increased by the fact that changes in the feed happen on a daily basis and a column operating under a feedback control can take several hours to return to an acceptable steady states. This paper investigates these points by developing a detailed dynamic model, studying the dynamic behavior in simulations, and developing a control scheme. Furthermore, the possibility of implementing a feedforward control scheme, in addition to a feedback one, is investigated where it is taken into account that feed composition measurements may involve time delays if the measurements are taken as samples analyzed in a lab.

The outline of this paper is as follows. A literature review is presented in the following subsection and Section 2 presents preliminary information. A detailed description of the model and control structure is presented in Section 3. Section 4 discusses column responses to a series of commonly occurring disturbances. Conclusions are given in Section 5.

1.1. *Literature review*

Reactive distillation has received a lot of attention as part of process intensification efforts in the last couple of decades. Employing reactive distillation can result in energy savings as the heat of reaction is directly used for separation of the mixture. Harmsen [3] has reviewed commercial applications of reactive distillation. Reactive distillation systems have been shown to reduce variable cost, capital expenditure and energy requirements by 20% or more for some processes [3]. Also, since the heat of reaction is used for evaporation in a column, increased reaction rates can results in increased evaporation rates without significant changes of the temperature. Thus, reactive distillation columns have been found to be less susceptible to runway behavior than conventional reactors [3]. Reactive distillation models have been surveyed extensively by Taylor and Krishna [5] and several articles describing dynamic models, and control structures [6–9] are available. A variety of different applications of reactive distillation in refineries have been reported, such as processes involving ethers (MTBE, ETBE, and TAME [10]). Sneesby et al. [11–13] have developed dynamic models for ETBE and MTBE, and also made general recommendations for control system design. Different control strategies for MTBE reactive distillation columns were highlighted by Bartlett and Wahnschafft [14]. A number of authors have also explored the dynamics and control for reactive distillation of TAME [15–18]. However, despite these extensive efforts on reactive distillation in general, no papers on benzene hydrogenation via reactive distillation can be found in the open literature. This situation is especially peculiar as benzene hydrogenation is an important step in a refinery and several RD columns used for benzene hydrogenation are in operation in refineries throughout the world.

2. Preliminaries

This section reviews preliminary information needed for the remainder of the paper. Section 2.1 reviews existing modeling approaches for reactive distillation columns, some of which will be used in this work. Existing control strategies for reactive distillation columns are discussed in Section 2.2.1 and Section 2.2.3 reviews the principles of feedforward control which will also be used.

2.1. *Packed column modeling*

Reactive distillation can be viewed as an extension of conventional packed columns, where some of the packing includes a catalyst to facilitate a reaction taking place. A number of papers have discussed modeling of conventional packed columns. The key methods used for packed columns are equilibrium (EQ) stage modeling and non-equilibrium stage modeling (NEQ). In EQ stage