



## Control of plantwide reactive distillation processes: Hydrolysis, transesterification and two-stage esterification

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### ABSTRACT

General design principles for controlling plantwide reactive distillation processes are developed in this work. Three cases of plantwide RD processes with large recycle flow rates are used to illustrate the application of the design principals. The cases are hydrolysis of methyl acetate, transesterification of methyl acetate and esterification of adipic acid. The design principles produce workable control structures for the three systems. Temperature and temperature–composition control structures can deal with the two major disturbances: changes in the feed flow rates and reactant composition. Results show that the control structures offer good robustness and acceptable dynamic performances.

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## 1. Introduction

Reactive distillation (RD) combines reaction and separation in a single unit which provides substantial economic benefit for some chemical processes. The coupling of both functions in a single unit makes the system highly nonlinear. Therefore, the operation and control of reactive distillation columns must be addressed at the design stage for successful implementation of the technology. A robust control strategy that ensures safe, stable and economic operation in the presence of disturbances is essential.

Many articles have appeared in the literature on the control of RD columns in last decade. Most RD design is based on “neat” operation, with no excess reactant fed to the RD column, because this configuration usually has greater economic potential. However even in this case, the control issue is more complex than for a traditional distillation column. A main issue is that reactants must be fed in the “exact” amount to satisfy the stoichiometry down to the last molecule. Luyben and co-workers propose eight control structures for “neat” reactive distillation with a reversible reaction with two reactants and two products: (CS1–CS6 in Al-Arfaj and Luyben (2000); CS7 in Al-Arfaj and Luyben (2002); CS7–CS8 in

Kaymak and Luyben (2004, 2005)). In general, basic concepts for a RDC with a second order reversible reaction (*i.e.*,  $A + B \leftrightarrow C + D$ ) are investigated. Other authors have also studied the control of RD columns. Sneesby *et al.* (1999) study the two-point control of an ethylene glycol column. Linear and nonlinear control of semi-batch reactive distillation for ethyl acetate production has been explored by Engell and Fernholz (2003). The quantification of nonlinearity measurements and the relationships between nonlinearity and control have been investigated by Hung *et al.* (2006b).

“Neat operation” is not suitable for all RD system designs. Sometimes excess reactant is needed to achieve high conversion of another species and avoid byproducts, for example, Fuchigami (1990) and Subawalla and Fair (1999). Thus control systems sometimes must be developed for an entire plant consisting of several interconnected unit operations including RD. Studies of the plantwide control of processes with reactive distillation columns and separation columns with recycle is less common compared to the control of a reactive distillation column alone or plantwide reactor/separator systems (Luyben *et al.*, 1998; Wu and Yu, 1996). Al-Arfaj and Luyben (2004) develop an effective control structure for the TAME process (one RD with two columns) using temperature control. Tang *et al.* (2005) presented control schemes that are workable for the ethyl acetate RD flowsheet but the two reactant feeds are still close to neat in his work. Recently, Hung *et al.* (2008) discussed the steady-state design of a reactive distillation flowsheet

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**Nomenclature**

AA	adipic acid
$B_i$	bottom flow rates in $i$ th column
BuAc	butyl acetate
$D_i$	top flow rates in $i$ th column
DMA	dimethyl adipate
EA	esterification of adipic acid
$F$	feed flow rates
FR	feed ratio in reactive distillation column
HAc	acetic acid
HM	hydrolysis of methyl acetate
$K_c$	controller gain
$K_{eq}$	equilibrium constant
$K_u$	ultimate gain
$m_{cat}$	catalyst weight
MeAc	methyl acetate
MeOH	methanol
MMA	monomethyl adipate
$N_r$	number of rectifying trays
$N_{rxn}$	number of reaction trays
$N_s$	number of stripping trays
$NF_{Azeotrope}$	azeotrope feed tray location
$NF_{BuOH}$	butanol feed tray location
$NF_{Recycle}$	recycle stream feed tray location
$N_T$	total number of trays
$P_u$	ultimate period
$QR_i$	heat duty in $i$ th column
RDC	reactive distillation column
RF	recycle flow rate
$RR_i$	reflux ratio in $i$ th column
$T_{i,j}$	temperature of component $j$ in $i$ th column
TAC	total annual cost
TM	transesterification of methyl acetate
$X_{Bi,j}$	bottom composition of component $j$ in $i$ th column
$X_{Di,j}$	top composition of component $j$ in $i$ th column
$\tau_1$	controller reset time

with large excess recycle stream for esterification of adipic acid, but the authors did not discuss the control aspects. Lin *et al.* (2008) gave the optimal reactive distillation plant for hydrolysis of methyl acetate and gave a workable temperature control configuration.

In this work, we investigate a new RD flowsheet for transesterification of methyl acetate first. Then we develop control structures for the three RD flowsheets: the new transesterification flowsheet, the esterification of adipic acid by Hung *et al.* (2008) and the hydrolysis of methyl acetate by Lin *et al.* (2008). The objective of this study is to examine and compare the control of three plantwide RD systems and also try to provide generic and effective control structure design procedures for such processes.

The transesterification and hydrolysis processes are especially useful for the recovery of methyl acetate from a poly-vinyl alcohol (PVA) plant. The PVA process produces a byproduct stream which is a mixture of methyl acetate and methanol near the azeotropic composition. Methyl acetate has a low economic value, and therefore it is desirable to upgrade the methyl acetate to a more valuable product while at the same time separating the methanol. The transesterification process converts methyl acetate into butyl acetate, and the hydrolysis process converts methyl acetate into acetic acid.

In Section 2, the characteristics of the three systems are introduced and the process flowsheets are shown. Also, the detailed steady-state design results for the TM flowsheet are shown in this section. The principals of control structure design developed in this work are given in Section 3. Because on-line temperature measurement is easy, temperature control structures will be applied to the three systems first. Later we discuss modified control structures for the case where composition measurement is possible. Modified control structures are also discussed if high purity products are required. Conclusions are given in the last section. In this work, dynamic simulation is carried out using AspenPlus<sup>®</sup> and AspenDynamics<sup>®</sup>.

**2. System characteristics**

Reaction kinetics and thermodynamic properties are important issues for the design of a RD process. If the system characteristics are not known, feasible and economic design is much more difficult.

**Table 1**  
Kinetic equations for three esterification systems.<sup>a</sup>

System	Kinetic model (catalyst)	Equilibrium constant ( $T = 323$ K)
HM	Adsorption-based model (Amberlyst 15) $R = m_{cat} \times \frac{k_f a'_{MeAc} a'_{H_2O} - k_r a'_{HAc} a'_{MeOH}}{(a'_{MeAc} + a'_{H_2O} + a'_{HAc} + a'_{MeOH})^2}, \quad a'_i = \frac{K_i a_i}{M_i} \quad k_f = 6.127 \times 10^5 \exp(-63730/RT)$ $K_{MeAc} = 4.15, \quad K_{H_2O} = 5.24, \quad K_{HAc} = 3.15, \quad K_{MeOH} = 5.64$ $k_r = 8.498 \times 10^6 \exp(-60470/RT)$	$K_{eq} = 0.01$
TM	Pseudo-homogeneous model (Amberlyst 15) $r = k_f \left( C_{MeAc} C_{BuOH} - \frac{1}{K_{eq}} C_{MeOH} C_{BuAc} \right)$ $k_f = 2.7 \times 10^6 \exp(-71960/RT)$ $k_r = 3.64 \times 10^6 \exp(-72670/RT)$	$K_{eq} = 0.97$
EA	Quasi-homogeneous model (Amberlyst 35) $\frac{-dC_{AA}}{dt} = k_1 C_{AA} C_{MeOH} - k_{-1} C_{MMA} C_{H_2O}$ $\frac{-dC_{MeOH}}{dt} = k_1 C_{AA} C_{MeOH} - k_{-1} C_{MMA} C_{H_2O} + k_2 C_{MMA} C_{MeOH} - k_{-2} C_{DMA} C_{H_2O}$ $\frac{dC_{MMA}}{dt} = k_1 C_{AA} C_{MeOH} - k_{-1} C_{MMA} C_{H_2O} - k_2 C_{MMA} C_{MeOH} + k_{-2} C_{DMA} C_{H_2O} \quad k_1 = 5.857 \times 10^6 \exp(-4097.8/T)$ $\frac{dC_{H_2O}}{dt} = k_1 C_{AA} C_{MeOH} - k_{-1} C_{MMA} C_{H_2O} + k_2 C_{MMA} C_{MeOH} - k_{-2} C_{DMA} C_{H_2O}$ $\frac{dC_{DMA}}{dt} = k_2 C_{MMA} C_{MeOH} - k_{-2} C_{DMA} C_{H_2O}$ $k_2 = 2.024 \times 10^6 \exp(-4201.1/T)$ $K_{1eq} = (k_1/k_{-1})$ $K_{2eq} = (k_2/k_{-2})$	$K_{1eq} = 0.997^b, \quad K_{2eq} = 2.56^b$

<sup>a</sup>  $R = 8.314$  [kJ/kmol/K],  $T$  [K],  $r$  [kmol/s],  $m_{cat}$  [kg<sub>cat</sub>],  $C_i$  [kmol/m<sup>3</sup>],  $M_i$  [kg/kmol]. (i) Pöpkén *et al.* (2000), (ii) Jimenez and Costa-Lopez (2002) and (iii) Chan *et al.* (2010).

<sup>b</sup> Equilibrium constants are temperature independent.

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