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Design and control of transesterification reactive distillation with thermal coupling

San-Jang Wang^{a,*}, David S.H. Wong^b, Shuh-Woei Yu^c

^a Department of Chemical and Material Engineering, Ta Hwa Institute of Technology, Chiunglin, Hsinchu 307, Taiwan, ROC

^b Department of Chemical Engineering, National Tsing Hua University, Hsinchu 300, Taiwan, ROC

^c Institute of Environmental Engineering, National Central University, Chung-Li, Taoyuan 320, Taiwan, ROC

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ABSTRACT

In the study, the design and control strategies of a reactive distillation process with partially thermal coupling for the production of methanol and *n*-butyl acetate by transesterification reaction of methyl acetate and *n*-butanol are investigated. Since methanol and methyl acetate formed an azeotrope, the products of a reactive distillation column include *n*-butyl acetate and the mixture of methanol and methyl acetate, which must be separated by an additional column. Partially thermal coupling can be used to eliminate the condenser of the second column. Not only energy reduction but also better operability and controllability can be obtained for the thermally coupled reactive distillation process. Proper selection and pairing of controlled and manipulated variables chosen for three control objectives were determined by using steady-state analysis. A simple control scheme with three temperature control loops is sufficient to maintain product purities and stoichiometric balance between the reactant feeds.

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1. Introduction

Process intensification is a design philosophy that deliberately integrates different processes into a single operation to reduce energy consumption and capital investment, as well as achieve environmental and safety benefit. Reactive distillation and thermally coupled distillation are successful examples of process intensification.

Reactive distillation has become one of the extensively researched process intensification methods due to its ability to reduce energy consumption and number of process units. Research on reactive distillation has been comprehensively reviewed by [Doherty and Buzad \(1992\)](#) and [Taylor and Krishna \(2000\)](#). One of the processes that can make effective use of reactive distillation is the production of methanol and *n*-butyl acetate by the transesterification reaction of methyl acetate and *n*-butanol. *n*-Butyl acetate is an important solvent in the chemical industry. It is used as a solvent for acrylic polymers, vinyl resins, and leather dressings in cosmetic formulations and other applications. It is also used as a reaction medium for adhesives. Methyl acetate is a by-product of the poly-(vinyl)-alcohol production and has to be converted to methanol which is a feedstock of the poly-(vinyl)-alcohol production. Like

esterification, transesterification is a typical equilibrium-limited reaction. Therefore, reactive distillation is used to improve conversion. [Jiménez and Costa-López \(2002\)](#) examined a four-column process configuration. First a feed of containing a low methyl acetate composition was first concentrated near to the methyl acetate/methanol azeotrope. A reactive distillation was added to convert methyl acetate by reacting with *n*-butanol into methanol and *n*-butyl acetate. *o*-Xylene was also added to this column so that extractive and reactive distillations were carried out. This was followed by two columns which recover the solvent *o*-xylene and further purify *n*-butyl acetate. [Luyben, Pszalgowski, Schaefer, and Siddons \(2004\)](#) proposed a three column design which converts a feed stock containing 60 mol% of methyl acetate and 40 mol% of methanol into methanol and *n*-butyl acetate. Extractive solvent was not used. In a reactive distillation column, a mixture of methanol and methyl acetate was reacted with a mixture of *n*-butanol and *n*-butyl acetate. The top product containing methanol was recycled to a pre-fractionation column to mix with the fresh feed so that the overall feed composition falls between methanol and methyl acetate/methanol azeotrope. The bottom product containing *n*-butyl acetate and *n*-butanol was fed to a purification column to produce high purity *n*-butyl acetate and a mixture containing *n*-butanol and *n*-butyl acetate which was recycled to the reactive distillation column. [Lin, Chen, Huang, and Yu \(2006\)](#) and [Chen \(2006\)](#) proposed a new flowsheet, which includes only one conventional distillation column followed by RDC. In the flowsheet,

* Corresponding author. Tel.: +886 3 5927700x2853; fax: +886 3 5927310.

E-mail address: cwjsj@et4.thit.edu.tw (S.-J. Wang).

Nomenclature

C_i	molar concentration of i component (mol/L)
K_c	proportional gain
K_{cu}	ultimate gain
m_1	reboiler duty of reactive distillation column
m_2	reboiler duty of side stripper column
m_3	n -butanol feed flow
P_u	ultimate period
r	reaction rate (mol/L min g_{cat})
R	gas constant (J/mol K)
T	temperature (K)
T_i	integral time

fresh methyl acetate/methanol feed and a recycled stream rich in methanol were fed to the first column, which produces methanol at the bottom and methyl acetate/methanol mixture close to azeotropic composition at the top. The distillate of the first column and fresh n -butanol were fed to the RDC. Butyl acetate product was withdrawn from the column base, while the methanol-rich distillate was recycled back to the first column. The two-column design was capable of producing high-purity butyl acetate and methanol with a total annual cost only 2/3 of the three-column design. In this work, a conventional reactive distillation process similar to the one proposed by [Lin et al. \(2006\)](#) and [Chen \(2006\)](#) was used as the starting point of thermal coupling study. A methyl acetate/methanol mixture, rich in methyl acetate, is fed to a reactive distillation column to react with n -butanol to produce a bottom product of n -butyl acetate. The top product is fed to a separation column to produce a distillate approximating the methyl acetate/methanol azeotrope, which is then recycled into the reactive distillation column. Methanol is recovered at the bottom of the separation column.

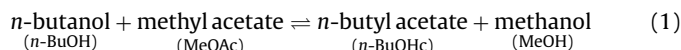
The separation of multi-component mixtures by continuous distillation has traditionally been accomplished by arranging columns in series. Several alternative configurations exist, most notably the direct and indirect sequences. Thermal coupling between two columns in a sequence has proven to be very successful in providing energy savings around 30% with respect to the conventional distillation trains ([Annakou & Mizsey, 1996](#); [Emtir, Rev, & Fonyo, 2001](#); [Glinos & Malone, 1988](#); [Triantafyllou & Smith, 1992](#); [Wolff & Skogestad, 1995](#)). However, their application was limited, mainly because of complexity in design and control of these structures ([Dunnebie & Pantelides, 1999](#)). In thermally coupled configurations at least one end of a distillation column has both liquid and vapor exchange with the other column. This eliminates either a reboiler or a condenser from the distillation column end. In three well known thermally coupled column configurations (side rectifier, side stripper, and fully coupled) for ternary separations, fully coupled configuration has been described to have higher thermodynamic efficiency for most separations ([Rudd, 1992](#)). However, [Agrawal and Fidkowski \(1998\)](#) showed that the fully coupled configuration is favorable only in certain operating region. Side rectifier and side stripper configurations tend to provide the most thermodynamically efficient designs more often than the fully coupled configuration over a wide range of relative volatilities and feed compositions.

In this study we will show that the reactive distillation process with thermal coupling for the production of high-purity methanol and n -butyl acetate by the transesterification reaction of methyl acetate and n -butanol cannot only save energy consumption but also have better operability and controllability than the conventionally complete reactive distillation process. The control strategy

of the thermally coupled reactive distillation process will also be investigated.

2. Design of reactive distillation process without thermal coupling

The transesterification reaction of methyl acetate and n -butanol is proceeded as follows:



n -Butyl acetate and methanol are the high and low boiling products, respectively. n -Butanol and methyl acetate are the high and low boiling reactants, respectively. The yield for the transesterification reaction is strongly limited by the equilibrium conversion. This reversible reaction needs to be catalyzed by strong acids. Kinetic data for a sulfonic ion-exchange resin, Amberlyst 15, were reported by [Jiménez, Garvin, and Costa-López \(2002\)](#) and a pseudohomogeneous model was also given:

$$r = 2.018 \times 10^8 \exp(71,960/RT) C_{n-BuOH} C_{MeOAc} - 2.839 \times 10^8 \exp(72,670/RT) C_{n-BuOAc} C_{MeOH} \quad (2)$$

where r is the reaction rate and C_i is the concentration of the i component.

The simulation of the transesterification distillation was carried out using the rigorous distillation model provided by ChemCad software. The liquid phase activities were calculated by using modified UNIFAC ([Gmehling, Lohmann, Jakob, Li, & Joh, 1998](#)). The predicted azeotropic temperatures and compositions agree well with experimental results provided by [Gmehling, Menke, Krafczyk, and Fischer \(2004\)](#).

[Fig. 1](#) is a schematic illustration of the conventionally complete reactive distillation process, including a reactive distillation column and a separation column. There are three zones in the reactive distillation column with 27 (including a total condenser), 16, and 7 (including a partial reboiler) equilibrium stages in the rectification, reaction, and stripping zones, respectively. Rectification zone and stripping zone operate as a non-reactive distillation column, purifying top and bottom products. n -Butyl acetate and

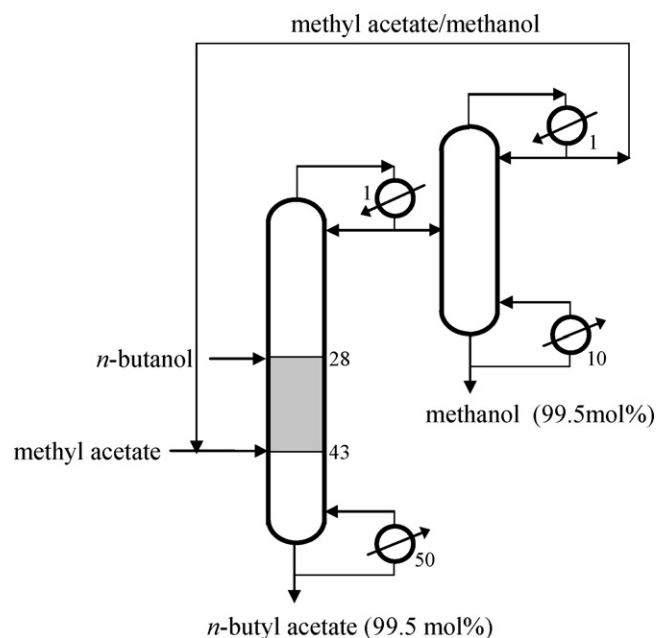


Fig. 1. Complete conventionally configuration of transesterification distillation.

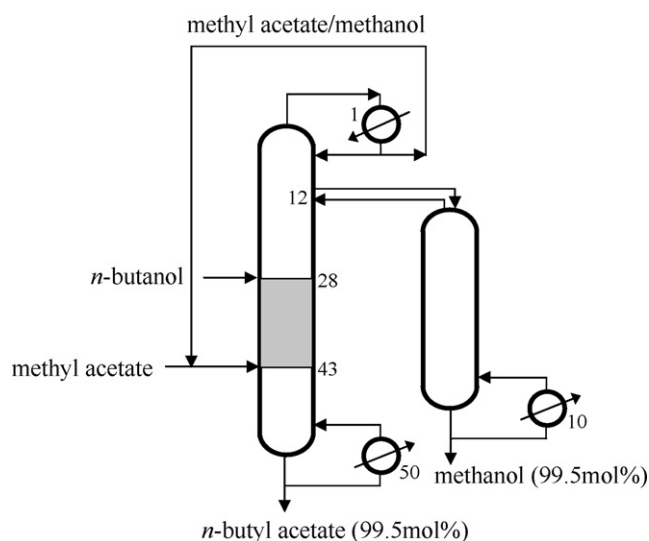


Fig. 2. Thermally coupled transesterification distillation.

methanol are formed in the reaction zone and withdrawn from the column bottom and top, respectively. Two reactants, *n*-butanol with saturated vapor at 1 atm and methyl acetate at 25 °C and 1 atm, are fed to the column.

n-Butanol and methyl acetate reactants with 100 kmol/h each are fed to the stages 28 and 43, respectively, numbering from the column top. The column is operated at 1 atm. The pressure drop in the column is assumed to be 0.2 atm. In the study, some design parameters about column geometry are determined by the criterion that the flood percent on each stage, calculated by using the sizing function provided by ChemCad software, is guaranteed not to exceed 75% at the nominal operating conditions. The column is sized as a tray-column with diameter 2.7 m. A liquid holdup of about 200 L was obtained. We assume that a reaction volume of 50 L and catalyst of 500 g are available on every tray in the reactive zone. *n*-Butyl acetate with high-purity 99.5 mol% is withdrawn from the column bottom. The distillate product is a mixture of methanol and un-reacted methyl acetate and then fed to a separation column with 10 stages. High methanol purity, 99.5 mol%, is obtained from the column bottom. The distillate of the column, approximate the minimum boiling azeotrope of methanol and methyl acetate, is recycled back to the fresh methyl acetate feed location.

Jiménez and Costa-López (2002) proposed a four-column process. The description of the four-column process can also be found in the paper of Luyben et al. (2004). They stated that *o*-xylene used to break the methyl acetate/methanol azeotrope perhaps affects the separation of *n*-butanol and *n*-butyl acetate as well; and they showed that the use of *o*-xylene is not necessary. In the work of Lin et al. (2006) and Chen (2006), a RDC started with a near azeotropic feed and produced a distillate rich in methanol. The RDC was used to cross from the methanol lean side of the azeotrope to the methanol rich side of the azeotrope. Breaking of azeotrope is not necessary. In our work, a similar design was used except that a methyl acetate/methanol feed rich in methyl acetate was directly fed to a RDC to produce a methyl acetate/methanol mixture rich in methanol. The crossing of methyl acetate/methanol azeotrope was also done by the RDC.

3. Design of reactive distillation process with thermal coupling

Fig. 2 shows the flowsheet of a reactive distillation process with thermal coupling. The same stage numbers of a reactive distillation

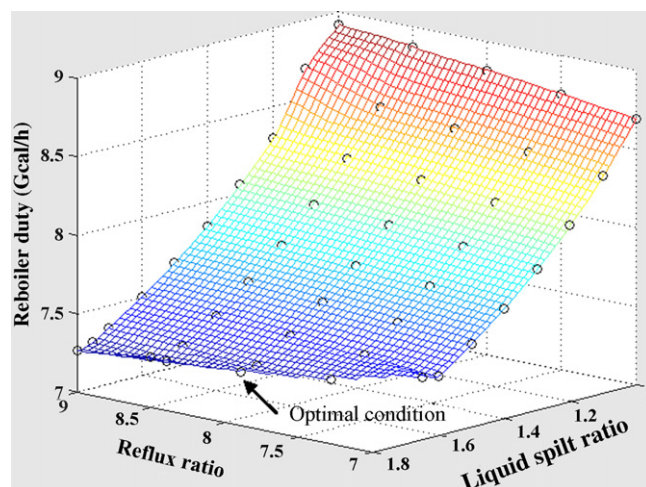


Fig. 3. Relationship between reboiler duty and reflux ratio and liquid split ratio at high product purities (99.5 mol%).

column and a side stripper column as those of the reactive distillation column and separation column in Fig. 1, respectively, are used in the study to compare the energy consumption of reactive distillation processes without and with thermal coupling. The same reaction volume and catalyst on every tray in the reactive zone are also used. Liquid and vapor are interchanged between these two units and then a condenser is reduced. The maximum boiling component, *n*-butyl acetate is withdrawn from the column bottom of the reactive distillation. The approximate homogeneous azeotrope of methanol and methyl acetate is distilled at the column top and recycled back to the fresh methyl acetate feed location. A side draw is designed to purge the methanol product to the side stripper column. Its location is decided by selecting the tray with the maximum methanol composition. High purity methanol with 99.5 mol% is obtained from the bottom of side stripper column.

There are four design degrees of freedom: reflux ratio and boilup flow of the reactive distillation column, boilup flow of the side stripper column, and liquid split ratio with only two product specifications for the reactive distillation process with thermal coupling. Liquid split ratio is specified as the ratio of the liquid flow directed to the side stripper column to the flow directed to the reactive distillation column. In this study, reflux and liquid split ratios are used as the remaining degrees of freedom to minimize the reboiler duties of the reactive distillation process with thermal coupling while the other two are used to satisfy the purity specifications. Fig. 3 shows the relationship between reboiler duty and two ratios when two product specifications are set at high purities (99.5 mol%). The optimal condition with the minimum energy consumption (7.24 Gcal/h) is obtained with reflux and liquid split ratios equal to 8.0 and 1.75, respectively. A region with low energy consumption is observed for high liquid split ratio. At a given liquid split ratio, the operation becomes infeasible when the reflux ratio becomes too low. Similarly, a maximum liquid split ratio can be found for a given reflux ratio.

There are also four design degrees of freedom: two reflux ratios and two boilup flows for the reactive distillation process without thermal coupling. In this study, two reflux ratios are used as the remaining degrees of freedom to minimize the reboiler duties of the reactive distillation process without thermal coupling while the other two are used to satisfy the purity specifications.

Fig. 4 shows the optimal reboiler duty comparison of the reactive distillation processes without and with thermal coupling for different reactant feed flows. The reactive distillation process with

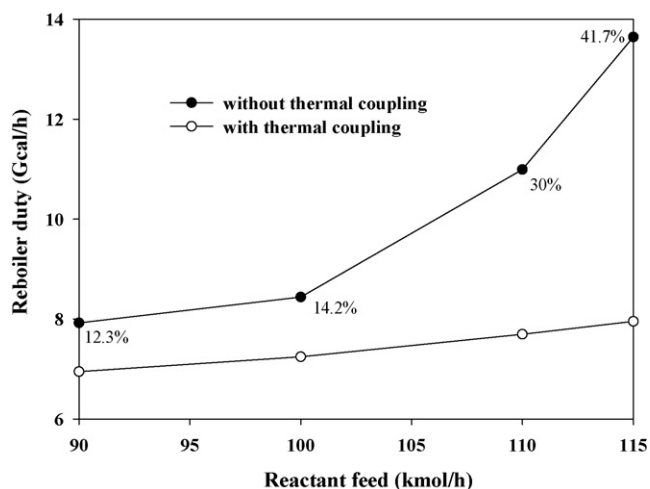


Fig. 4. Relationship between reactant feed and optimal reboiler duty for reactive distillation processes without and with thermal coupling.

thermal coupling can save 14.2% energy consumption compared with the one without thermal coupling at the nominal reactant feed, 100 kmol/h. The energy savings decrease slightly with decreases in reactant feed. The reboiler duty of the process without thermal coupling increases sharply with the reactant feed for the same amount of catalyst in the reactive distillation column. However, the energy consumption increases proportionally with the reactant feed for the process with thermal coupling. Its energy saving percent can reach 41.7% when reactant feed is equal to 115 kmol/h. There is no solution to obtain high-purity (99.5 mol%) products at two column bottoms when the reactant feed exceeds 115 kmol/h for the process without thermal coupling. Fig. 5 shows the reactant composition profiles of reactive distillation columns for the optimized processes without and with thermal coupling when reactant feed flows are 100 kmol/h and 115 kmol/h. Deviation from stoichiometric condition (*n*-butanol:methyl acetate = 1:1) is higher in the reaction zone for the process without thermal coupling. In addition, the deviation increases with the reactant feed rate. There is only little variation of the mole fractions of two reactants in the reaction zone of the process with thermal coupling when reactant feed changes. These results shown in Figs. 4 and 5 reveal that the reactive distillation process with thermal coupling not only saves energy consumption but also has better operability than the one without thermal coupling.

4. Temperature control of reactive distillation process with thermal coupling

For a reactive distillation column in the kinetic regime, the desired steady-state temperature or composition profile changes when feed rate changes and product purities are kept at their designed values (Wang, Wong, & Lee, 2003a). Hence there are three keys to controlling such a column: (1) to maintain the correct stoichiometric balance between the feeds, (2) to maintain the product quality, and (3) to account for possible changes in control objective when throughput rate changes.

The stoichiometric balance between the reactant feeds must be maintained when operating a reactive distillation column. Feed-ratio control is the simplest way to maintain the balance. However, when there is a measurement bias in feed flow rate, the feed-ratio control scheme will not be able to keep the desired feed ratio. To overcome this problem, Al-Arfaj and Luyben (2000) suggested that the reactant composition of some column stage be controlled by

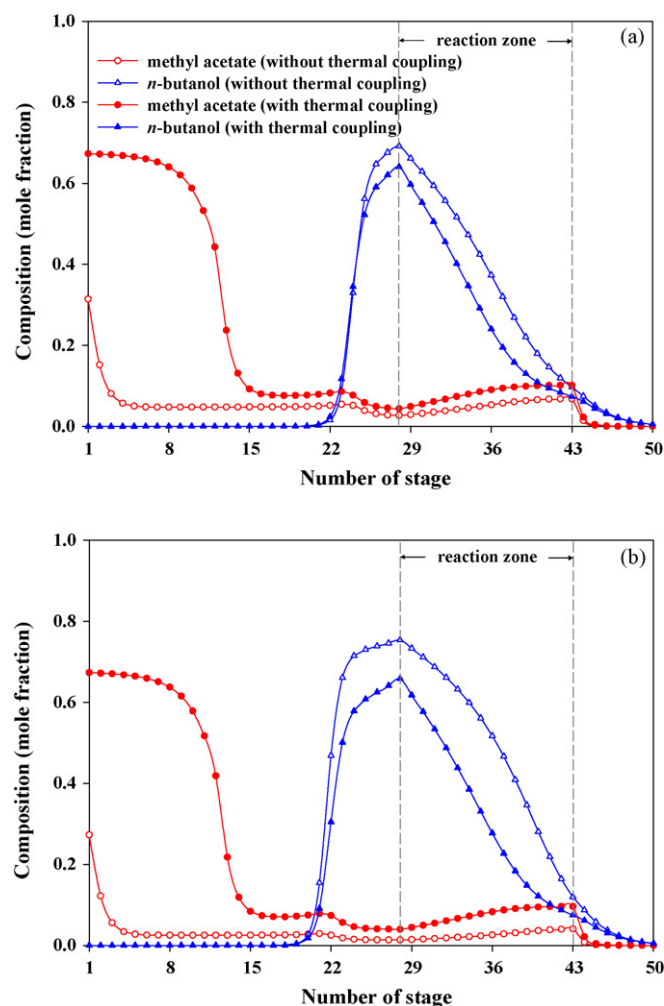


Fig. 5. Reactant composition profiles of reactive distillation columns for the optimized processes without and with thermal coupling when reactant feed flows are (a) 100 kmol/h and (b) 115 kmol/h.

the reactant feed flow. In industrial applications, temperature control is usually used instead of composition control. The reason is that most product analyzers, such as gas chromatographs, suffer from large measurement delays and high investment and maintenance cost. The internal composition loop can be replaced by an internal temperature loop. Hence the internal stage temperature selected should have near linear relationship and sufficient sensitivity with respect to the feed flow rate. Methyl acetate and *n*-butanol feed flows are two candidates for the manipulated variable of the internal temperature control. Reactive distillation processes often exhibit input multiplicities, which increases the difficulties of operation and control. Input multiplicity occurs when two or more sets of input variables produce the same output variable. With reference to control structures, input variables are those that can be manipulated by control valves or other actuating devices. Output variables are those that are either controlled or measured to describe the conditions of the process. Figs. 6 and 7 show the effect of methyl acetate and *n*-butanol feeds on different stage temperatures of the reactive distillation and side stripper columns, respectively. Input multiplicity appears just around the nominal operating point between methyl acetate feed and temperatures of stages 35–50 of reactive distillation column. However, there is also input multiplicity between *n*-butanol feed and temperatures of stages 3–10 of the side stripper column.

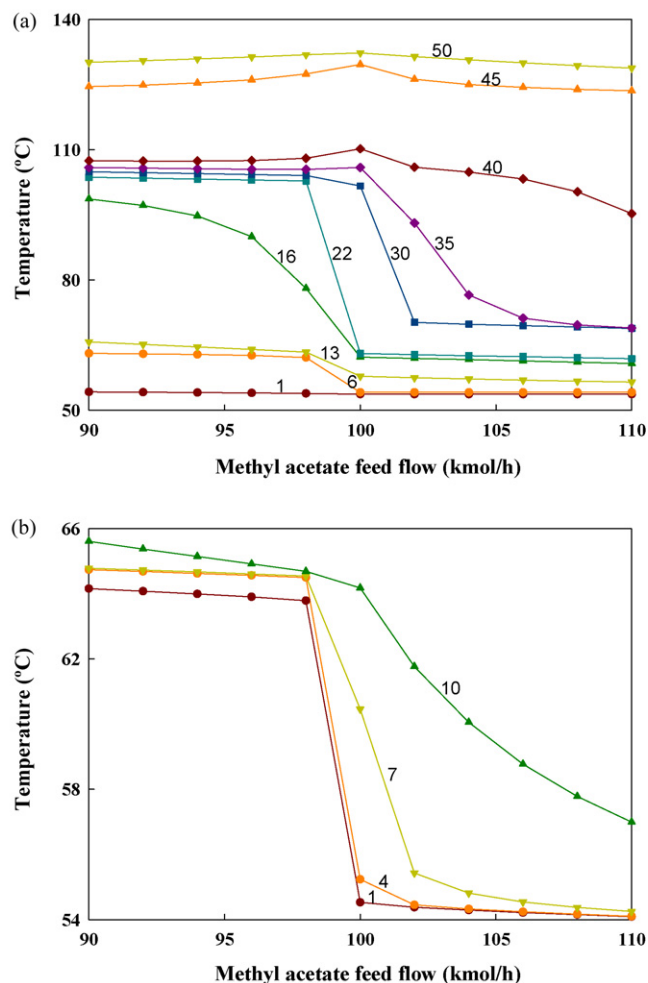


Fig. 6. Sensitivity analysis between methyl acetate feed flow and stage temperatures of (a) reactive distillation column and (b) side stripper column.

To maintain high-purity *n*-butyl acetate and methanol at the bottoms of reactive distillation column and side stripper column, respectively, two temperature loops can be used, in which two stage temperatures are maintained by manipulating the reboiler duties of the two columns. The input–output relation between stage temperature and reboiler duty should have a near linear relationship without multiplicity and sufficient sensitivity around the nominal operating condition. Figs. 8 and 9 show the relationship between stage temperatures of reactive distillation and side stripper columns and reboiler duties of these two columns, respectively. Very low sensitivity and near-constant value of temperature can be found between stages 1 and 12 of reactive distillation column. Temperatures of stages 15–37 of reactive distillation column show very strong nonlinearity around the nominal operating condition. Input multiplicity occurs between these two reboiler duties and the temperatures of stages 3–10 of side stripper column. Wang, Wong, and Lee (2003b) showed that interaction multiplicity is one of the most important factors to be considered in selecting the manipulated variable. The stage temperatures with input multiplicity or interaction multiplicity with manipulated variables should not be used as controlled variables to avoid the problem of control stability. From the results of Figs. 6–9, temperatures of stages 35–50 of the reactive distillation column and those of stages 3–10 of the side stripper column cannot be used as controlled variable candidates due to input multiplicity or interaction multiplicity if methyl acetate feed flow is used as the manipulated variable. However, only temperatures of

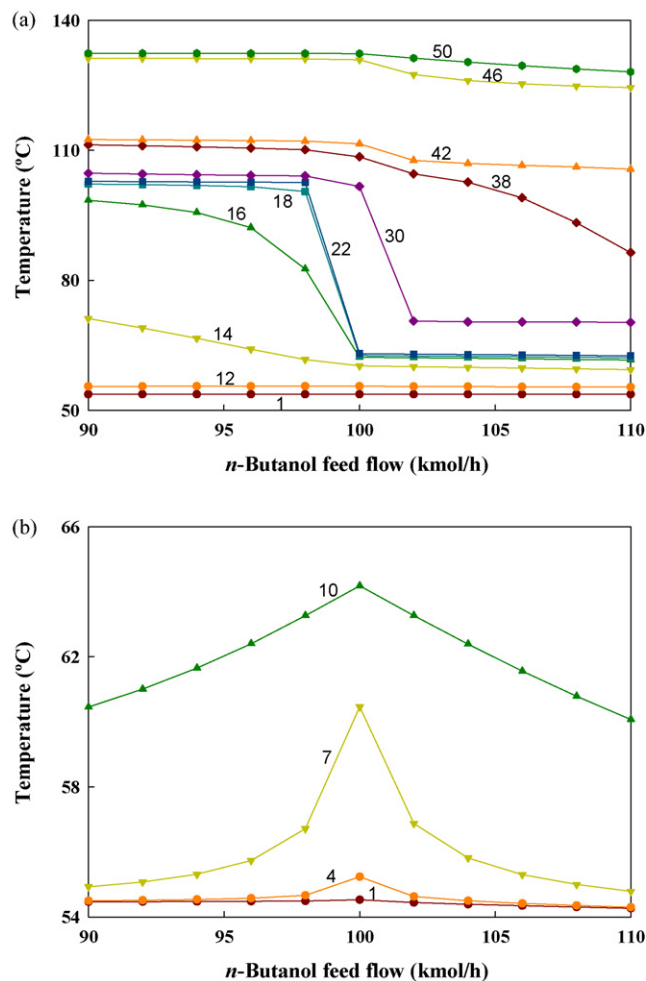


Fig. 7. Sensitivity analysis between *n*-butanol feed flow and stage temperatures of (a) reactive distillation column and (b) side stripper column.

stages 3–10 of the side stripper column occur input multiplicity or interaction multiplicity and therefore more stage temperature can be used as the controlled variable candidates when *n*-butanol feed flow is used as the manipulated variable. Therefore, *n*-butanol feed flow is chosen in the study as the manipulated variable to maintain reactant inventory in the process.

Fig. 10 shows the desired steady-state temperature profiles when both methyl acetate and *n*-butanol feed flows are simultaneously increased or decreased by the same amounts, 10%, and with bottom methanol and *n*-butyl acetate compositions kept as their respectively designed values. Different desired temperature profiles are found for various throughput rates due to residence time change on the reactive stages as the throughput rate changes for the reactive distillation column in the kinetic regime. However, there is almost no change for the desired steady-state temperature profile of the side stripper column.

For a reactive distillation column in the kinetic regime, the desired steady-state temperature profile changes as feed rate changes (Wang et al., 2003a). Hence the set point of the temperature control loop must be adjusted as long-term disturbances occur. This will cause a lot of difficulty in column operation. However, in this transesterification case, the temperatures except those between stages 23 and 31 of the reactive distillation column are almost invariant or have little variations with respect to throughput rate changes and can thus be used as the controlled variable candidates of temperature loops.

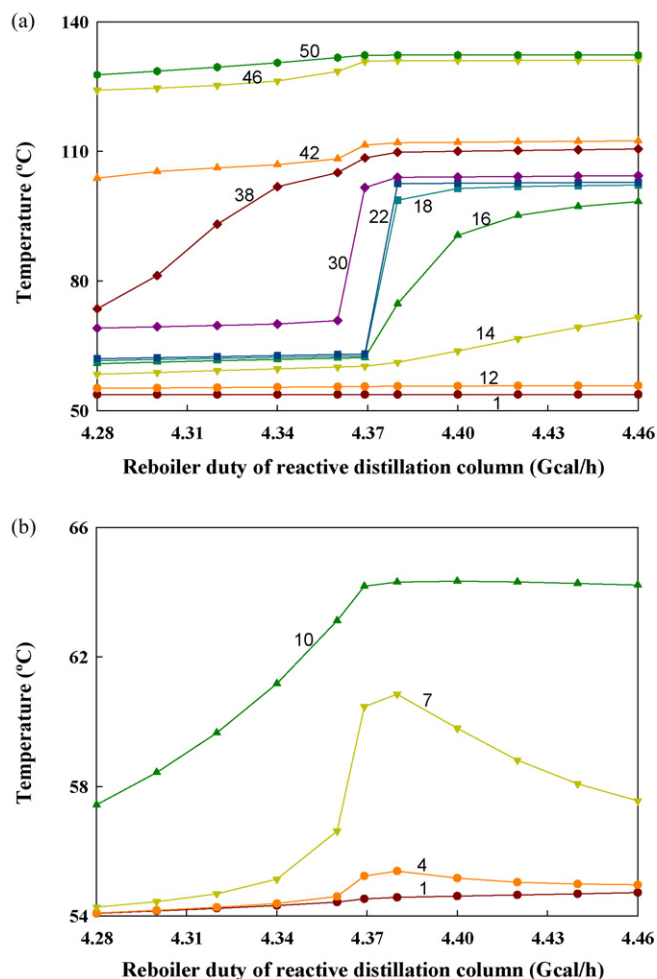


Fig. 8. Sensitivity analysis between reboiler duty of reactive distillation column and stage temperatures of (a) reactive distillation column and (b) side stripper column.

To select the most appropriate location used for control purpose, singular value decomposition method (Moore, 1992) was used in the study. The controlled temperatures are selected at stages 13 ($T_{13(RD)}$), 38 ($T_{38(RD)}$), and 48 ($T_{48(RD)}$) of the reactive distillation column from the stage temperatures having no input or interaction multiplicities with manipulated variables and being almost invariant or having little variations with respect to throughput rate changes.

Table 1 shows the relative gain array (RGA) analysis of these controlled stage temperatures and manipulated variables. m_1 , m_2 , and m_3 represent the reboiler duty of reactive distillation column, reboiler duty of side stripper column, and *n*-butanol feed flow, respectively. The control loop pairing is that the temperatures at stages 13, 38, and 48 in the reactive distillation column are controlled by manipulating the reboiler duty of the side stripper column, reboiler duty of the reactive distillation column, and *n*-butanol feed flow, respectively. To compare the controllability

Table 1
RGA analysis between controlled stage temperatures and manipulated variables for the process with thermal coupling

	m_1	m_2	m_3
$T_{13(RD)}$	−0.12	1.94	−0.82
$T_{38(RD)}$	8.24	−6.04	−1.20
$T_{48(RD)}$	−7.12	5.10	3.02

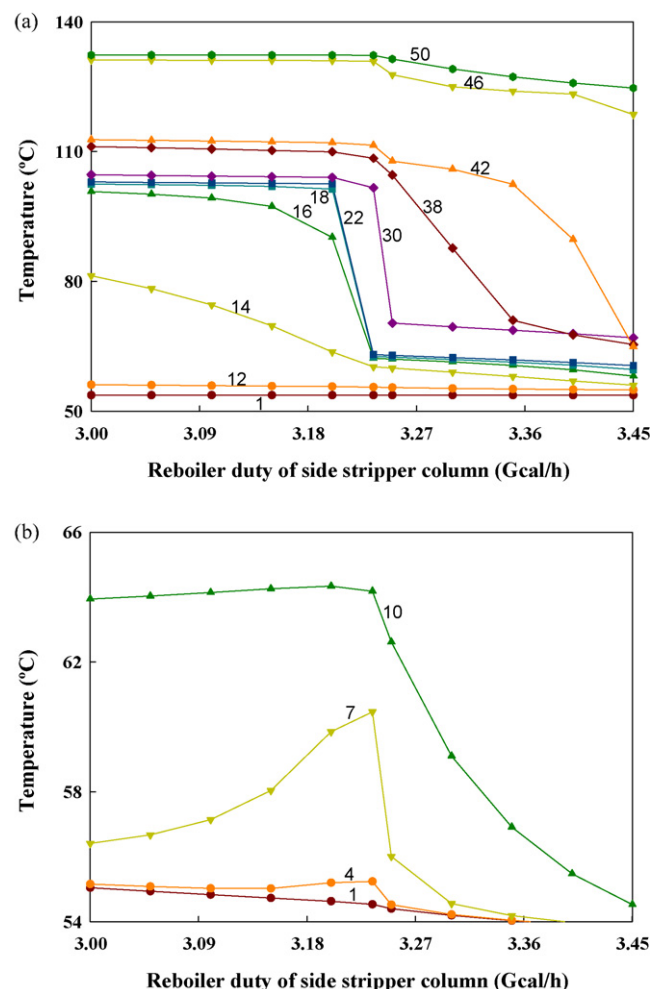


Fig. 9. Sensitivity analysis between reboiler duty of side stripper column and stage temperatures of (a) reactive distillation column and (b) side stripper column.

between the processes with and without thermal coupling, the same steady-state analysis stated above is also used to select the controlled stage temperatures and manipulated variables of the process without thermal coupling. Table 2 shows the RGA analysis of chosen controlled stage temperatures and manipulated vari-

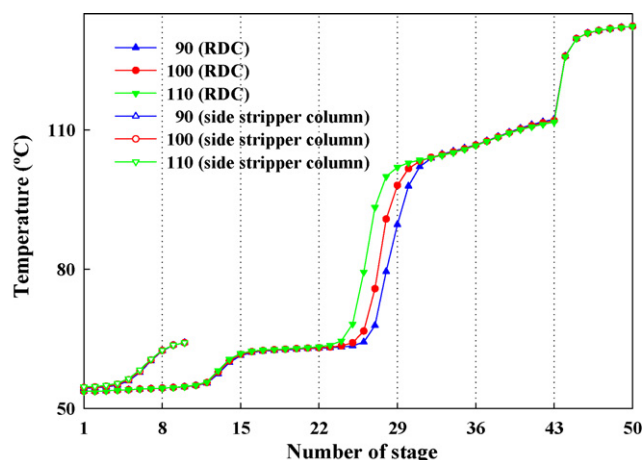


Fig. 10. Effect of throughput rate on the desired steady-state temperature profile.

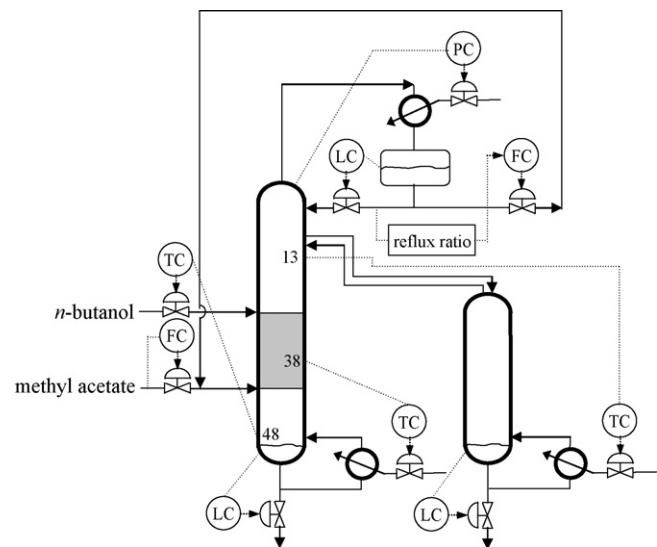


Fig. 11. Control scheme for the reactive distillation process with thermal coupling.

Table 2
RGA analysis between controlled stage temperatures and manipulated variables for the process without thermal coupling

	m_1	m_2	m_3
$T_{2(RD)}$	7.29	−6.10	−0.19
$T_{40(RD)}$	−77.52	87.48	−8.96
$T_{46(RD)}$	71.23	−80.38	10.15

ables. It indicates that much better controllability can be obtained for the process with thermal coupling.

In the following discussions on control performance, the controllers were tuned using a sequential design approach (Shen & Yu, 1994) after the pairing was determined. For each controller, relay-feedback test (Luyben, 1987) is performed to obtain ultimate gain and ultimate frequency. The following equations are used to calculate the tuning parameters of PI controllers:

$$K_C = \frac{K_{CU}}{3} \tag{3}$$

$$T_I = \frac{P_U}{0.5} \tag{4}$$

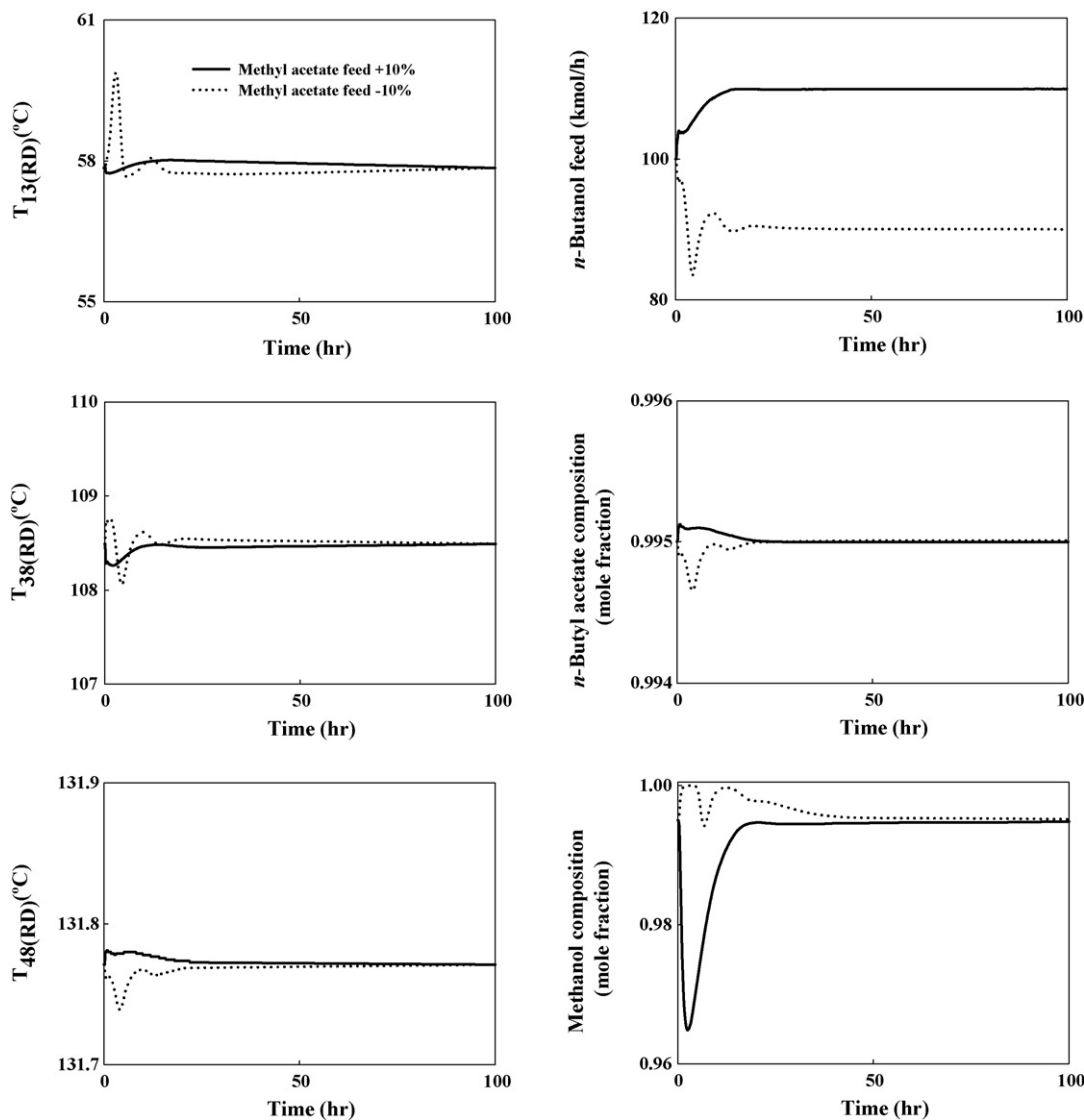


Fig. 12. Dynamic responses of controlled stage temperatures, n-butanol feed flow, and product purities under temperature control for ±10% changes in methyl acetate feed flow.

where K_C and T_I represent proportional gain and integral time, respectively, and K_{CU} and P_U are ultimate gain and ultimate period, respectively. Fig. 11 shows the control scheme for the reactive distillation process with thermal coupling. Three temperature control loops are designed by the above steady-state analysis. In the reactive distillation column, column pressure is controlled by manipulating coolant flow rate. Reflux-drum level and base level are maintained by changing reflux flow rate and bottom flow rate, respectively. Reflux ratio is controlled by manipulating distillate flow rate. Liquid split ratio is maintained by a liquid flow distributor. The methyl acetate feed is flow controlled. In the side stripper column, the base level is maintained by changing bottom flow rate. In the distillation control system, pressure, level, and flow control belong to inventory control maintaining the basic operation of column. Thus in the following discussion, emphasis is placed on the response of temperature control strategy used to maintain product quality and stoichiometric balance.

Fig. 12 shows the dynamic responses of controlled stage temperatures, *n*-butanol feed flow, and the corresponding product purities under temperature control for $\pm 10\%$ methyl acetate feed flow disturbances. The controlled stage temperatures are settled at their corresponding set points. Stoichiometric balance is maintained and two product compositions can return to their desired operating values under temperature control.

5. Conclusions

In the study, a reactive distillation process with thermal coupling is presented to produce *n*-butyl acetate and methanol by the transesterification reaction of methyl acetate and *n*-butanol. High product purities can be obtained at the bottoms of a reactive distillation column and a side stripper column. Not only energy consumption can be reduced but also better operability and controllability can be obtained for the thermally coupled process with respect to a conventionally complete process.

n-Butanol feed is used to maintain the reactant inventory in the reactive distillation column for its more flexible ability in selecting controlled stage temperatures. Controlled stage temperatures having no input multiplicity and interaction multiplicity with manipulated variables are selected by steady-state analysis. Dynamic simulation results show that the temperature control strategy without changing the set points of temperature loops can maintain reactant inventory in the reactive distillation column and bottom product purities at their desired operating values.

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References

- Agrawal, R., & Fidkowski, Z. T. (1998). Are thermally coupled distillation columns always thermodynamically more efficient for ternary distillations? *Industrial & Engineering Chemistry Research*, 37, 3444.
- Al-Arfaj, M. A., & Luyben, W. L. (2000). Comparison of alternative control structures for an ideal two-product reactive distillation column. *Industrial & Engineering Chemistry Research*, 39, 3298.
- Annakou, O., & Mizsey, P. (1996). Rigorous comparative study of energy-integrated distillation schemes. *Industrial & Engineering Chemistry Research*, 35, 1877.
- Chen, J. H. (2006). *Design and control of reactive distillation processes for the production of butyl acetate via transesterification*. Master Thesis. National Taiwan University.
- Doherty, M. F., & Buzad, G. (1992). Reactive distillation by design. *Chemical Engineering Research & Design*, 70, 448.
- Dunnebie, G., & Pantelides, C. C. (1999). Optimal design of thermally coupled distillation columns. *Industrial & Engineering Chemistry Research*, 38, 162.
- Emtir, M., Rev, E., & Fonyo, Z. (2001). Rigorous simulation of energy integrated and thermally coupled distillation schemes for ternary mixture. *Applied Thermal Engineering*, 21, 1299.
- Glinos, K. A., & Malone, M. F. (1988). Optimality regions for complex column alternatives in distillation systems. *Chemical Engineering Research & Design*, 66, 229.
- Gmehling, J., Lohmann, J., Jakob, A., Li, J., & Joh, R. (1998). A modified UNIFAC (Dortmund) model. 3. Revision and extension. *Industrial & Engineering Chemistry Research*, 37, 4876.
- Gmehling, J., Menke, J., Krafczyk, J., & Fischer, K. (2004). *Azeotropic data*. Weinheim: Wiley-VCH.
- Jiménez, L., & Costa-López, J. (2002). The production of butyl acetate and methanol via reactive and extractive distillation. II. Process modeling, dynamic simulation, and control strategy. *Industrial & Engineering Chemistry Research*, 41, 6735.
- Jiménez, L., Garvin, A., & Costa-López, J. (2002). The production of butyl acetate and methanol via reactive and extractive distillation. I. Chemical equilibrium, kinetics, and mass-transfer issues. *Industrial & Engineering Chemistry Research*, 41, 6663.
- Lin, Y. D., Chen, J. H., Huang, H. P., & Yu, C. C. (2006). Process alternative for methyl acetate conversion using reactive distillation: Transesterification versus hydrolysis. In *Proceedings of the AIChE Annual Meeting*.
- Luyben, W. L. (1987). Derivation of transfer functions for highly nonlinear distillation columns. *Industrial & Engineering Chemistry Research*, 26, 2490.
- Luyben, W. L., Pszalgowski, K. M., Schaefer, M. R., & Siddons, C. (2004). Design and control of conventional and reactive distillation processes for the production of butyl acetate. *Industrial & Engineering Chemistry Research*, 43, 8014.
- Moore, C. F. (1992). Selection of controlled and manipulated variables in practical distillation control. In W. L. Luyben (Ed.), *Practical distillation control* (pp. 140–177). New York: Van Nostrand Reinhold.
- Rudd, H. (1992). Thermal coupling for energy efficiency. Supplement to *The Chemical Engineer* (August 27), 525, S14.
- Shen, S. H., & Yu, C. C. (1994). Use of relay-feedback test for automatic tuning of multivariable systems. *AIChE Journal*, 40, 627.
- Taylor, R., & Krishna, R. (2000). Modelling reactive distillation. *Chemical Engineering Science*, 55, 5183.
- Triantafyllou, C., & Smith, R. (1992). The design and operation of fully thermally coupled distillation columns. *Transactions IChemE*, 70, 118.
- Wang, S. J., Wong, D. S. H., & Lee, E. K. (2003a). Control of a reactive distillation column in the kinetic regime for the synthesis of *n*-butyl acetate. *Industrial & Engineering Chemistry Research*, 42, 5182.
- Wang, S. J., Wong, D. S. H., & Lee, E. K. (2003b). Effect of interaction multiplicity on control system design for a MTBE reactive distillation column. *Journal of Process Control*, 13, 503.
- Wolff, E. A., & Skogestad, S. (1995). Operation of integrated three-product (Petlyuk) distillation columns. *Industrial & Engineering Chemistry Research*, 34, 2094.