The Effect Of Implementing Thermally Coupled Distillation Sequences On Snowball Effects For Reaction-Separation-Recycle Systems

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Abstract

It is known that the steady and dynamic characteristics of processes with recycles can be significantly different from those without recycle streams. Particular attention has been given to the analysis of the reactor-separator-recycle system (RSR); because of the existence of snowball effects, a small load change can translate into a large change in the recycle flow rate, which propagates throughout the whole system. Previous works have analyzed the influence of separators on snowball effects using flash units and conventional distillation columns. This work explores the existence of snowball effects in steady state conditions under the implementation of thermally coupled distillation sequences in RSR systems. In particular, the implementation of coupled systems with side columns (sequences with side rectifiers or side strippers) is studied. The effects of considering thermally coupled systems on snowball effects are compared to those generated by conventional distillation systems.

Keywords Thermally coupled distillation, snowball effect, reactor separator recycle

1. Introduction

In most chemical and petrochemical industries, distillation is used as the primary separation and purification technique. Because of their higher energy efficiency, thermally coupled distillation systems have provided an attractive alternative to the conventional distillation sequences. Most of the research work in this area has been focused on design methods (Glinos and Malone 1985; Hernández and Jiménez, 1996), optimization procedures (Grossmann et al., 2005) and control issues (Serra et al, 2001, Segovia et al, 2007). Although such studies have shown that there is an incentive for the implementation of thermally coupled arrangements, they have been carried out with the separation system in isolation, that is without taking into account the interactions with the whole chemical process. Nevertheless, it is important to consider that separation units in chemical plants are generally connected with other process units, in most cases by recycle streams.

Luyben (1994) analyzed the dynamic performance of Rector-Separation-Recycle (RSR) systems. He focused on the high sensitivity that the recycle stream has to the feed flowrate, or “snowball effect”, pointing that its dynamic effect depends on the control strategy. Luyben suggests that dynamic snowballing can be avoided if one flowrate in the recycle loop is controlled (Luyben’s rule). Larsson et al. (2003) analyzed the RSR
control problem subject to active constrains to optimize the economic performance of
the process. They took into account two scenarios: when the objective function is to
minimize the energy consumption, and when the objective function is to maximize
production rate. Their results showed that, depending on economic and operational
goals, Luyben’s rule is not always accurate.
Snowballing effect is influenced by the nonlinearity in the RSR models. Nonlinearity
can be introduced by the complex kinetics in the reaction step or by the separator model
solution. Zeyer et al. (2003) studied the influence of separator control structure into the
whole process dynamics, using a simple flash unit. They showed that, even for this
simple model, systems exhibit complex patterns such as infeasible solutions or multiple
steady states when the recycle is closed.
Most RSR studies have considered conventional distillation units. This work explores
the existence of snowball effects in steady state conditions under the implementation of
thermally coupled distillation sequences in RSR systems. We analyze the steady state
performance of these systems when one recycle stream is closed and identify sources of
instability when two recycles are closed. The results are compared with the use of
conventional sequences.

2. Case study

2.1 Process studied

The base case considered in this work is a simplified plant to produce ethylbenzene
from ethylene and benzene. All kinetic and design parameters for this base case were
taken from Luyben (2002). The reaction system can be described as:

\[ \text{benzene}(B) + \text{ethylene}(E) \rightarrow \text{ethylbenzene}(EB) \]

\[ \text{ethylbenzene}(EB) + \text{ethylene}(E) \rightarrow \text{diethylbenzene}(DEB) \]  \hspace{1cm} (1)

\[ \text{diethylbenzene}(DEB) + \text{benzene}(B) \rightarrow 2\text{Ethylbenzene} \]

Fresh benzene stream and a recycle benzene stream are fed to the first reactor
(isothermal and isobaric). The effluent from the first reactor and a recycle DEB stream
are fed to the second reactor, which operates adiabatically. The second reactor effluent
is fed to a first distillation column, where the recycled benzene goes overhead, and a
mixture of DEB and EB (bottoms product) are fed to the second column, where EB is
recovered as a distillate product; the bottoms stream, with a high composition of DEB,
is recycled back to the second reactor.

2.2 Distillation Sequences

We considered four distillation sequences: a conventional direct sequence (DS), a
conventional indirect sequence (IS), a scheme with a side rectifier (TCSR), and a
scheme with a side stripper (TCSS). The conventional sequences were designed by
shortcut methods (Seader and Henley, 2003). For thermally coupled distillation
sequences, the design parameters for minimum energy consumption were obtained
following the methodology proposed by Hernández and Jiménez (1996).
The thermally coupled distillation sequences provide lower energy consumption than
the conventional sequences. After they were designed, the sequences were
implemented into the whole process. The resulting process flowsheets are shown in
Figure 1. One of the main concerns is that in the TCSR and TCSS arrangements both
recycle streams come from the main column; the distillate goes back to reactor 1, and
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the bottoms product to reactor 2. All design parameters for each distillation sequence are given in Tables 1 and 2.

![Flowsheet A](image1.png)  ![Flowsheet B](image2.png)

**Table 1. Design Parameters. Conventional Distillation Sequences**

<table>
<thead>
<tr>
<th></th>
<th>Direct Sequence</th>
<th>Indirect Sequence</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stages/ Feed stage of column 1</td>
<td>26/14</td>
<td>29/10</td>
</tr>
<tr>
<td>Stages/ Feed stage of column 2</td>
<td>32/17</td>
<td>20/6</td>
</tr>
<tr>
<td>Pressure column 1/column 2 (bar)</td>
<td>4.5/2</td>
<td>4.5/2</td>
</tr>
<tr>
<td>Reflux ratio 1/ Reflux ratio 2</td>
<td>2.66/0.8</td>
<td>0.68/0.7</td>
</tr>
<tr>
<td>Feed stream flowrate (kmol/hr)</td>
<td>1596</td>
<td>1596</td>
</tr>
<tr>
<td>Distillate flowrate 1 kmol/hr)</td>
<td>900</td>
<td>1531</td>
</tr>
<tr>
<td>Bottoms flowrate 2 (kmol/hr)</td>
<td>696</td>
<td>65</td>
</tr>
<tr>
<td>Distillate flowrate 1 kmol/hr)</td>
<td>631</td>
<td>900</td>
</tr>
<tr>
<td>Bottoms flowrate 2 (kmol/hr)</td>
<td>65</td>
<td>631</td>
</tr>
<tr>
<td>Qreb (Heat duty, kcal/hr)</td>
<td><strong>21,172,538</strong></td>
<td><strong>28,789,569</strong></td>
</tr>
</tbody>
</table>

Figure 1. Process Flowsheets.
Table 2. Design Parameters. Thermally Coupled Distillation Sequences

<table>
<thead>
<tr>
<th></th>
<th>TCSR</th>
<th>TCSS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stages of main column</td>
<td>41</td>
<td>32</td>
</tr>
<tr>
<td>Feed/ interconnecting stage</td>
<td>14/29</td>
<td>23/6</td>
</tr>
<tr>
<td>Stages of side column</td>
<td>15</td>
<td>14</td>
</tr>
<tr>
<td>Interconnection flowrate, kmol/hr</td>
<td>950</td>
<td>1200</td>
</tr>
<tr>
<td>Pressure (bar)</td>
<td>1.7</td>
<td>1.7</td>
</tr>
<tr>
<td>Reflux ratio</td>
<td>0.69</td>
<td>2.13</td>
</tr>
<tr>
<td>Feed stream flowrate (kmol/hr)</td>
<td>1534</td>
<td>1596</td>
</tr>
<tr>
<td>Distillate flowrate (REC1, kmol/hr)</td>
<td>900</td>
<td>900</td>
</tr>
<tr>
<td>Side Product flowrate (kmol/hr)</td>
<td>630</td>
<td>630</td>
</tr>
<tr>
<td>Bottoms Flowrate (REC2, kmol/hr)</td>
<td>65</td>
<td>65</td>
</tr>
<tr>
<td>$Q_{reb}$ (Heat duty, kcal/hr)</td>
<td>16,706,638</td>
<td>17,966,567</td>
</tr>
</tbody>
</table>

2.3 Snowball effect

Common load disturbances are changes in flow or feed composition, because these variables affect directly the reactor conversion. In this work we explore the existence of snowballing varying fresh feed ratios and temperature in reactor 1. The changes analyzed are listed in Table 3. To observe variations on recycles flows, we decided to specify the boil up ratio in the distillation columns instead of product flowrates. The response variables were the recyle flowrates and reboilers duties.

Table 3. Variables analyzed

<table>
<thead>
<tr>
<th>VARIABLE</th>
<th>RANGE</th>
</tr>
</thead>
<tbody>
<tr>
<td>FFE</td>
<td>Fresh Feed of Ethylene (kmol/h)</td>
</tr>
<tr>
<td>FFB</td>
<td>Fresh Feed of Benzene (kmol/h)</td>
</tr>
<tr>
<td>Reactor Temperature of R1 (°C)</td>
<td>100-300</td>
</tr>
</tbody>
</table>

3. Results

For the process with conventional distillation arrangements, both recycles closed without any complication. However, because of the complexity of the process flowsheet with thermally coupled distillation systems, we carried out a first analysis when only one recycle was closed. All results were obtained through rigorous simulations using the commercial simulator Aspen Plus One ®. Results are summarized in Figure 2. Because it is fed in excess to the first reactor, benzene is the main component in recycle 1. Changes in fresh feed of both reactants were considered for a better evaluation of the process response when the amount of unreacted benzene was considered. In Figure 2a we can observe that the conventional sequences show more stable patterns than thermally coupled distillation sequences against changes in fresh feed of benzene. The column with a side rectifier is particularly sensitive to such changes, and presents a clear snowball effect. However, when we analyzed the system response to changes in fresh feed of ethylene (Figure 2b), the results show that the TCSR sequence is the one with less variability, followed by the conventional sequences; the most sensitive arrangement to this change was the TCSS. When changes in the fresh feed of ethylene (FFE) were considered, the results showed that in all cases the system was less sensitive to decreasing FFE than to increasing the amount of benzene in the fresh feed.
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Figure 2c shows the variation in recycle stream when the reactor temperature was considered. It is fairly clear that TCSR presents a snowball effect, while the indirect sequence is the most stable in the entire range.

The topologies considered thermal couplings because of the expected energy savings, which is why Figures 2 d), e) and f) focus on the variations in reboiler duties due to changes in fresh feed and reactor temperature.

When the benzene stream was increased, the largest changes in the reboiler duty were observed for the thermally coupled distillation sequences, with a three fold increase (snowball effect, Figure 2d). The effect of variations in the ethylene feed stream are shown in Figure 2e. The DS, IS and TCSS do not show major changes in reboiler duties.

Figure 2. Recycle 1 and steady state reboilers’ duties as functions of different loads
duties, keeping their energy requirements close to the original values. Overall, the less sensitive sequence was the TCSR. These results are consistent with those under changes in recycle 1, the distillate product.

Finally, for IS and TCSS an interesting observation is pointed out. According with Zeyer et al. (2003) multiplicity can be expected in the separator solution caused by energy balances. In this work the multiplicity was observed only for indirect sequences (IS and TCSS) when the variable was FFE.

4. **Concluding Remarks**

We have analyzed the influence of separators in a RSR problem when thermally coupled distillation systems are implemented. The results for the case study showed that in most cases Conventional Sequences were less sensitive to load changes than Thermally Coupled Distillation arrangements, but when changes in fresh feed of ethylene were considered, the TCSR reduced snowball effects and energy demand; also, multiplicity was observed for the indirect sequences (IS and TCSS). Results for thermally coupled distillation sequences with both recycles closed are not included because of space limitations. This work was carried out under steady state conditions, but it provides the basis for further studies on dynamic analysis and control strategies.

**Acknowledgements**

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**References**


