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Case study: Using the Dynamic Flowsheet DIVA® Simulator for Modeling Methyl Acetate Reactive Distillation Process

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Abstract

In this paper, the author presents a point of view for the modeling of the methyl acetate reactive distillation process using the dynamic flowsheet simulator DIVA® (Dynamische Simulation verfahrenstechnischer Anlagen). The focus is on the description of the modeling concept, the main aspect being its "module-oriented" structure. Furthermore, the process model is presented and some simulations are being illustrated in the end of this work.

Key words: *reactive distillation, mathematical model, dynamic simulation.*

Introduction

Reactive distillation (RD) combines a chemical reactor and a distillation column into a single operating unit; therefore this process, compared to classical distillation processes, has a high complexity, compensated by infrastructure simplification and costs reduction.

Because of the advanced research made in this field, RD has been successfully applied in the last years and it covers a wide area of commercial processes [5].

Due to the strong interaction between reaction and separation, RD processes can sometimes show an intricate nonlinear dynamic behavior including steady state multiplicities, bistability and self sustained nonlinear oscillation [7, 14].

A good review regarding the modeling of reactive distillation is detailed in Taylor and Krishna's paper [16].

RD Dynamic Model and Column Details

During the research stage made by the author at Max Planck Institute for Dynamics of Complex Technical from Magdeburg[1], the production of methyl acetate process was investigated [1].

The studied methyl acetate RD column has 27 trays in all, it is a lab scale column and consists of three sections: from 1 to 6 trays is the stripping section, 7 to 17 represents reactive section and from 18 to 27 is the rectifying section (as illustrated in fig. 1). The molar flow rates of

[1] http://www.mpi-magdeburg.mpg.de

MeAc and AcH are $8.4033*10^{-4}$ kmol/min, respectively $8.1667*10^{-4}$ kmol/min, on the stages 7 and 21. The reflux ratio is set at 1.68. The column operates at a pressure of 1 atm. The reaction is catalyzed by sulfuric acid, added to the liquid phase on the tray 11 [3].

Feed rates, reboiler rate, reaction rate and measured temperatures are inputs and outputs of the model for controller synthesis and implementation purposes.

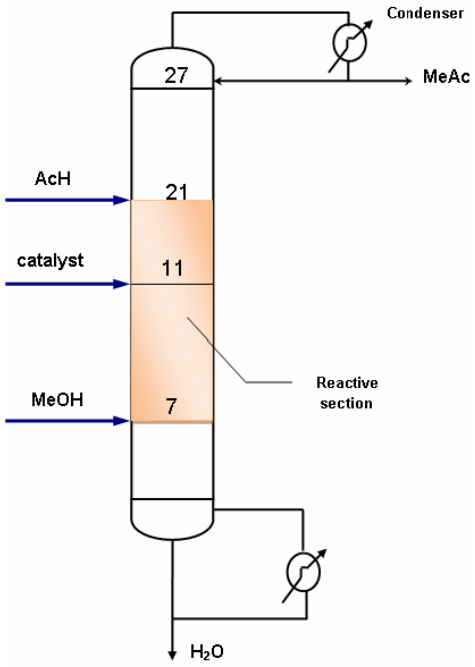


Fig. 1. Reactive distillation column

The reaction equilibrium is due to Agreda et al. [1] and vapour-liquid equilibrium is calculated according to Doherty & Barbosa [2].

For this particular process a simple model, good for further studies is presented. The model is described in [15], here will be emphasized only the basic assumptions for obtaining the good and robust model:

- the column has NSTAGE theoretical stages, including the condenser, decanter and the reboiler;
- there is constant holdup on trays;
- constant pressure is assumed;
- negligible heat effects;
- deviations from thermodynamic equilibrium are modeled with the tray efficiency **η**;
- the vapor and liquid phases on each tray are in equilibrium;
- the reaction takes place only in the liquid phase, where there is a perfect mixture between reactants and catalyst.

The simulations were carried out in DIVA simulator (Dynamische Simulation Verfahrenstechnischer Anlagen), developed at the Stuttgart University. This simulation environment runs under Linux operating system and integrates some sets of high-level routines

to solve/integrate complex algebraic and differential equation systems [6, 10]. For a detailed simulator presentation, the reader may consult the references [4, 6, 8, 9, 10, 11, 12, 13].

DIVA Simulator

The structure of DIVA is presented in [10]. The main components of the DIVA environment "work" together for obtaining a complex process model. The new processes models are written in symbolic form, which is converted by the preprocessor and then included into the model library. During runtime, the unit operation models are aggregated to a plant model, which forms a different algebraic (DAE) system of differential index one. [10].

The plant model formulated in DIVA in linearly implicit form is described in equation (1):

$$\begin{aligned} &B(x,u,p,m,t)\dot{x} = f(x,u,p,m,t); \\ &x(t_0) = x_0; \\ &y = \bar{x}, \end{aligned} \tag{1}$$

where, B is the system matrix, f is free values vector, x represents the state vector, u is the input vector, y is the input vector, p represents the parameter vector, y is the output vector, m is the "disturbances" parameter vector (state variables that are at the same time output variables), t represents the time variable, x_0 is the initial state vector, t_0 represents the initial time.

For the studied methyl acetate process, the model implemented in DIVA consists in seven parts: Parameters, Model Inputs, Model States, Model Outputs, Help Variables, Process Values (grouping of data output), Model Equations (condenser, column trays inside and reboiler).

The parameters are of three types: structure, integer and real parameters. Figure 2 illustrates the way of defining all three parameter types, whereas Table 1 shows some of the most significant parameters, as well as their values.

```
STRUCTURE PARAMETERS

(define - structure - parameters
   (: name "NC"
    : meaning "number of components"
    : default -value 27
)

INTEGER PARAMETERS

(define - integer - parameters
   (: name "IKATZU"
    : meaning "position of catalyst input positions 1(1)
                NSTAGE are allowed)"
    : default -value 11
)

REAL PARAMETERS

(: name "FLZU"
 : meaning "liquid feed rates [kmol/min] FLZU(NLZU+1) is a
            dummy and not further needed"
 :indices ((:lower-boundary 1 :upper-boundary "NLZU+1"))
 : default – value
    (array-list :lower-boundary 1 :upper-boundary "NLZU+1")
                : values (8.1667e-4  8.4033e-04  0.0)
)
```

Fig. 2. Structure, integer and real parameters definition

Table 1. Examples of some structure, integer and real parameters for the methyl acetate process

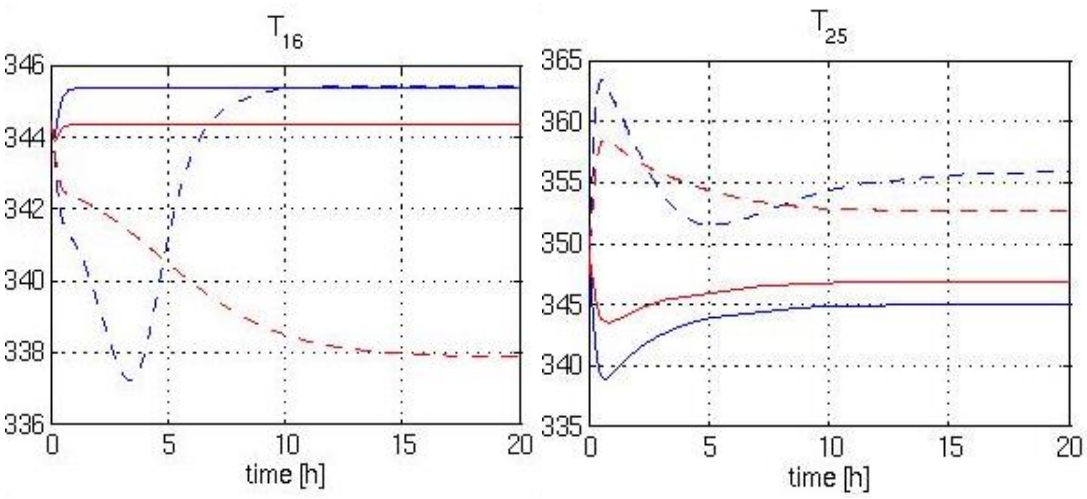
Type	Name	Meaning	Default value
Structure parameters	NC	number of components	4: 1 – acetic acid 2 – methanol 3 – methyl acetate 4 – water
	NSTAGE	number of stages	27
	NLZU	number of liquid feeds	2
	NGZU	number of vapor feeds	0
Integer parameters	IKATZU	position of catalyst input	11
	ILZU	positions of liquid feeds	(7 21 0)
	NUE	stoichiometric coefficients	(-1 -1 1 1)
Real parameters	FLZU	liquid feed rates [kmol/min]	(8.1667e-4 8.4033e-04 0.0)
	N	holdup of tray [kmol]	0.1
	P	pressure [Pa]	1.013e+05

The A-, B- and C-parameters for Antonine saturation pressure equation are illustrated in the table above:

Table 2. Parameters for Antoine equation

Component	A-parameters	B-parameters	C-parameters
AcH	9.51271	1533.313	-50.851
MeOH	10.20586	1582.271	-33.434
MeAc	9.19013	1157.630	-53.434
H_2O	10.19620	1730.630	-39.734

As an example of the simulation results, the steady-state variations of a reactive tray temperature, T_{16} and a stripping tray temperature T_{25}, both measured in K, with respect to acetic acid feed flow rate F_{AcH} are plotted in figure 3. The input is changed by ±5% and ±10% around the base case.



a) b)

Fig. 3. Step responses of a) T_{16} and b) T_{25} in F_{AcH} step changes

Figure 3 shows the process's nonlinearity, an inverse response being illustrated in fig. 3 a), at -10% step change in disturbance F_{AcH}, which requires a suitable control strategy, which will be investigated in a future paper. For a ±5% step disturbance in F_{AcH} (from figure 3b)) the responses are symmetrical.

Conclusions

RD combines both reaction and distillation in a single unit, offering significant economic benefits when compared to conventional process designs (reaction followed by separation). Its application area became wider in the last years due to researches that had been made in this field.

This paper briefly presented some basic ideas for the model implementation in DIVA simulator for the specific process of methyl acetate, as well as the main parameters of this particular model. As an example, results of the dynamic simulation with the simulation environment DIVA were shown.

Nomenclature

AcH – acetic acid

MeOH – methanol

MeAc – methyl acetate

F_{AcH} – acetic acid feed flow rate

F_{MeOH} – methanol feed flow rate

Q_r – heating rate

T_{16} – reactive temperature

T_{25} – stripping temperature

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O abordare privind utilizarea mediului integrat DIVA® pentru modelarea procesului de obținere a metil acetatului prin distilare reactivă

Rezumat

În cadrul acestui articol este prezentat pe scurt procesul de obținere a metil acetatului prin distilare reactivă. conceptul de distilare reactivă, precum şi modul de implementare a modelului aferent procesului în mediul de simulare DIVA®.

De asemenea, sunt prezentate valorile pentru anumiți parametri ai modelului, precum şi câteva exemple de grafice obținute în urma simulării procesului.