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SIMULATION AND OPTIMIZATION OF A STYRENE MONOMER REACTOR USING A NEURAL NETWORK HYBRID MODEL

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Abstract: The modeling and optimization of a dehydrogenation reactor in the industrial styrene monomer plant has been proposed in this study. Because this reactor consumes large amount of expensive high-pressure steam to produce the styrene monomer (usually more than two third of total energy costs), the minimization of the operating cost is highly desirable to maximize the profit. However, it is not easy to develop the accurate mathematical model because of the lack of internal or intermediate measurements of the industrial reactor, and also the lack of experimental results of the catalyst deactivation. To overcome these difficulties, we propose an alternative model of the styrene monomer reactor using a hybrid model in which the mathematical model is combined with neural networks. Major reaction mechanism is described in the mathematical model, and the deactivation effect is modelled in neural network using the real operation data. Using this model, the sensitivity analysis and the optimization of the industrial plant have been performed. The proposed optimal operation enlarges the profit of the plant very much. *Copyright © 2002 IFAC*

Keywords: reactor modeling, parameter optimization, parameter estimation, process simulators.

1. INTRODUCTION

Most of the industrial styrene monomer plants carry out the adiabatic dehydrogenation reaction in the multiple reactors in series. Even though these adiabatic reactors need more expensive equipment cost and operating cost, they have been used in many companies because of their high conversion of ethylbenzene and the small usage of catalyst pellets (Lim, *et al.*, 2001b; Wett, 1981).

The optimization of a styrene monomer(SM) reactor is highly valuable because it can be used in optimizing the current operation, which has high cost due to the large amount of expensive high-pressure steam (Haung, 1983). But, only a few researchers had studied about industrial styrene monomer reactors because they have difficult geometries and unknown reaction mechanisms (Sundaram, *et al.*, 1991). There had been many efforts to improve the productivity of styrene monomer production plants. But most of them had been focused on improvement of the recovery section or the other equipments except the reactor and development of new dehydrogenation catalyst (Cavani and Trifiro, 1995). Reaction models had been formulated (Clough and Ramirez, 1976; Scheel and Crowe, 1969; Hirano, 1986) and characteristics of the styrene monomer plug-flow reactor had been studied (Scheel and Crowe, 1969; Abdalla, *et al.*, 1976). Only Savoretti (Savoretti, *et al.*, 1999) proposed the non-adiabatic radial-flow reactor model. Through these researches, it had been possible to represent the styrene monomer reactor mathematically.

Although the mathematical model of the radial flow reactor gave a reasonable prediction from the industrial point of view (Lim, *et al.*, 2000), the nature of the process presented some difficulties in the prediction of catalyst activity. The lack of internal or intermediate measurements of the reactor represented a limitation for the detailed model validation. In addition, the lack of experiments of catalyst activity represented the uncertainty of parameters in mathematical equations. Thus the mathematical model was not enough to represent changes of catalyst activities.

In this study, alternative models of the styrene monomer reactor have been proposed using a hybrid model in which the mathematical model is combined with neural networks. The mathematical kinetic model for an adiabatic radial-flow styrene monomer reactor has been used as a first principle model. A neural network model has been developed for the

catalyst deactivation model because the exact parameters for the deactivation could not be measured and the deactivation was observed by the plant data only. Using this model, the simulation and optimization have been performed. Some examples have been tested with this simulator and the potential usages of this program have been investigated.

2. SYSTEM DESCRIPTION

2.1 Reactor System: characteristics

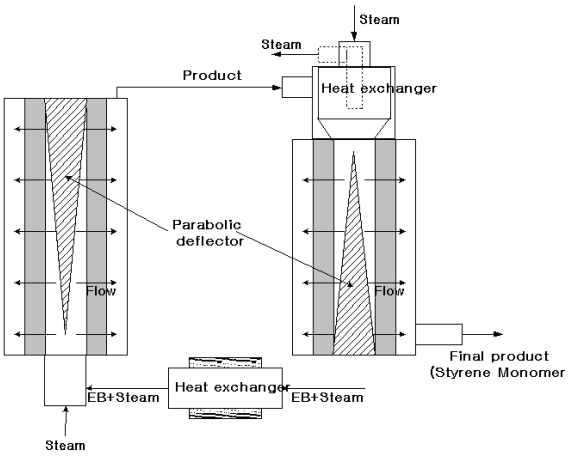

Fig. 1. Reactor configuration of current adiabatic radial flow reactor

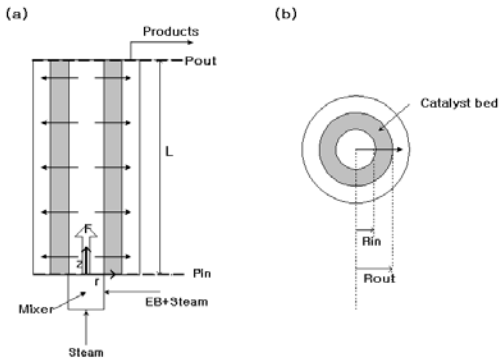

Fig. 2. Simplified reactor model and cross-sectional configuration

This styrene monomer plant has two ethylbenzene dehydrogenation reactors in series as shown in Fig. 1. Between two reactors, there is a heat exchanger. In this heat exchanger, the product stream from the first reactor is reheated up to 630℃.

The conversion of ethylbenzene (EB) and the selectivity of ethylbenzene to styrene are affected by reactor operating pressure, temperature, molar steam-to-hydrocarbon ratio (S/O ratio), and reactor load (=[current ethylbenzene flow rate] / [design ethylbenzene flow rate]).

The desired reaction can be enhanced at the low operating pressure because the main reaction of styrene monomer production increses the number of moles. The hydrocarbon feed to the reactor is the mixture of fresh ethylbenzene and recycled unconverted ethylbenzene. Prior to entering the reactor, this hydrocarbon feed is mixed with superheated steam in the adiabatic radial flow reactor. This steam acts not only as a heating medium but also as a diluent. High S/O ratio and low operating pressure increase the molar conversion of ethylbenzene. Molar conversion of ethylbenzene is also a function of temperature; higher temperature yields higher conversions in nearly a linear function. Therefore, an increment in conversion can be obtained by increasing the reactor temperature. Since the reaction of ethylbenzene to styrene is endothermic, it is carried out in multiple adiabatic radial bed reactors filled with catalysts. As for load, molar conversion of ethylbenzene is an inverse function of this parameter since higher velocity means lower residence time. This in turn reduces molar conversion (Kirk, et al., 1983).

2.2 Reaction Mechanism

In the styrene monomer reactor, three major competing reactions are known (Scheel and Crowe, 1969): reactions (1), (2), and (3). Styrene is produced by dehydrogenation of ethylbenzene. These reactions are endothermic except (3).

$$C_6H_5CH_2CH_3 \leftrightarrow C_6H_5CHCH_2 + H_2 \quad (1)$$

$$C_6H_5CH_2CH_3 \rightarrow C_6H_6 + C_2H_4 \quad (2)$$

$$C_6H_5CH_2CH_3 + H_2 \rightarrow C_6H_5CH_3 + CH_4 \quad (3)$$

In addition to these, there are three side reactions by thermal cracking at higher temperature.

$$H_2O + \frac{1}{2}C_2H_4 \rightarrow CO + 2H_2 \quad (4)$$

$$H_2O + CH_4 \rightarrow CO + 3H_2 \quad (5)$$

$$H_2O + CO \rightarrow CO_2 + H_2 \quad (6)$$

Since the dehydrogenation of ethylbenzene is a reversible endothermic reaction, high styrene yield is favored by high temperature.

3. MODEL DESCRIPTION

3.1 First Principle Model: mathematical model

Reaction model: The kinetic model for the reactions is shown in Table 1.

Governing equations: The adiabatic radial flow reactor is simplified using the following five assumptions.

(1) Quasi-steady state operation
(2) Ideal gas mixture: this assumption is valid because the styrene monomer reactor is operated at high temperature and low-pressure condition.
(3) Uniform distribution: the parabolic deflector inside of the plant makes the uniform flow pattern.
(4) No pressure drop and mass or heat diffusion in the axial direction: only the pressure drop exists inside of catalyst bed in the radial direction.
(5) No reaction except catalyst bed

According to the previous assumptions, the shell mass balance (15), energy balance (16), pressure drop (17) equations in cylindrical coordinates are following.

Table 1 Reaction Model

Reaction amounts in reaction j, f_j	
$f_1 = k_1(P_{EB} - P_{ST}P_{H2}/K_P)$	(7)
$f_2 = k_2 P_{EB}$	(8)
$f_3 = k_3 P_{EB} P_{H2}$	(9)
$f_4 = k_4 P_{H2O} P_{ET}$	(10)
$f_5 = k_5 P_{H2O} P_{ME}$	(11)
$f_6 = k_6 \frac{P}{T^3} P_{H2O} P_{CO}$	(12)
Reaction amounts of component I, f_i,	
$f_i = \sum_{j=1}^{6} \nu_{ij} F_j$	(13)
Reaction rate constant	
$k_i = k_0 \exp\left(\frac{-E_i}{RT}\right)$	(14)

$$\frac{dg_i}{dr} = \frac{1}{F}\left(2\pi r L \sum_{j=1}^{m} f_{ij} MW_i\right), \quad \sum_{i=1}^{NC} g_i = 1 \qquad (15)$$

$$\frac{dT}{dr} = \frac{2\pi r L \sum_{j=1}^{m}\left(-\Delta H_j\right) f_j}{FC_p} \qquad (16)$$

$$\frac{dP}{dr} = \frac{P_{out} - P_{in}}{R_{out} - R_{in}} \qquad (17)$$

3.2 Neural Network Model

The neural network model consists of a set of processing units called neurons, connected to one another. The neural network in this study is a feed-forward network with one hidden layer, seven input variables and one output variable. By adjusting parameters in the coupling, between neurons, the network is capable of learning from a set of numerical data corresponding to the input and desired output. (Nascimento, et al., 1999)
Because of the time-varying characteristics of catalyst activity, the recurrent neural network is used in this hybrid model.

As the input variables are,
(1) temperature (T(*k-1*));
(2) pressure (P(*k-1*)),
(3) partial pressure of steam in feed (P_{STM}(*k-1*));
(4) feed flowrate of EB (F_{EB}(*k-1*));
(5) deactivation factor at time point *k-1* (Φ(*k-1*)).

As the output variable is
(1) deactivation factor at time point *k* (Φ(*k*))

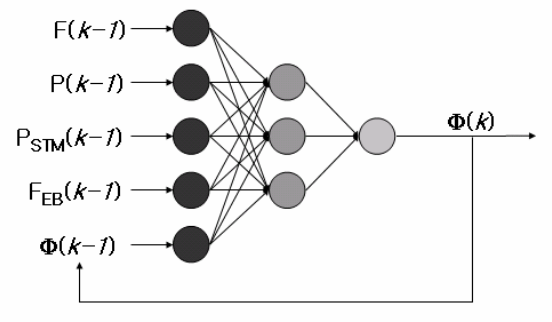

Fig. 3. Neural network model

A set of data obtained in the industrial unit was used to train the neural network and the other set to check the trained neural network. As shown in Fig 4, using the difference of two values, calculated real factor (Φ_0) for target value and predicted factor (Φ) for output value, neural network model can be trained. Because desired output, Φ_0 is not measured in plant, Φ_0 is calculated from governing equation and real plant data.

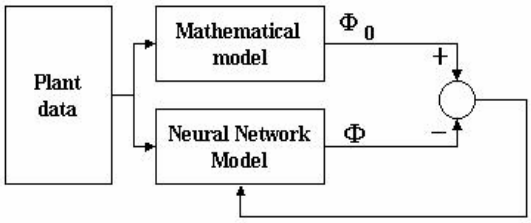

Fig. 4. Schematic diagram of learning method

Figs 5 and 6 show the calculated (Φ_0) versus predicted deactivation factor (Φ) for the learning and test set. The achieved agreement is within the error range, 10%.

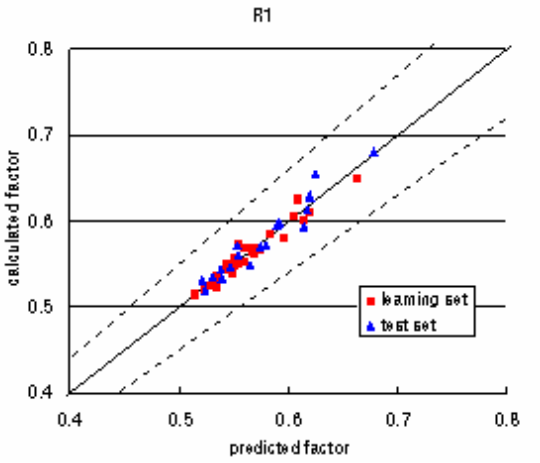

Fig. 5. Calculated vs. predicted factors of reactor 1

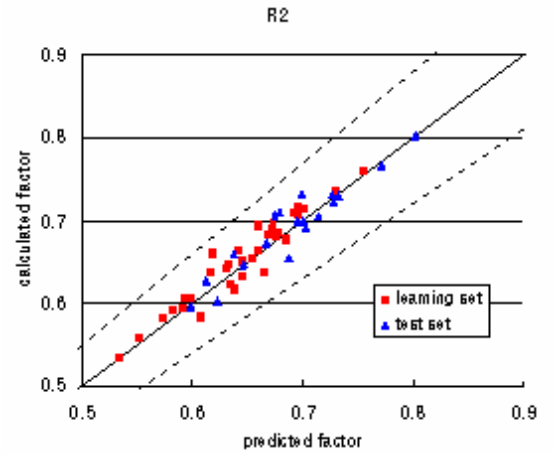

Fig. 6. Calculated vs. predicted factors of reactor 2

3.3 Hybrid Model

After training, the proposed neural network model supplies the catalyst deactivation factor (Φ) at any operating conditions. Mass balance, energy balance and pressure drop equations can be solved using given plant data. From these equations, reactor output data, such as temperature and composition of the output flow of each reactor, can be obtained.
The structure of the hybrid model, in which the mathematical model and the neural network model are coupled, is presented in Fig 7.

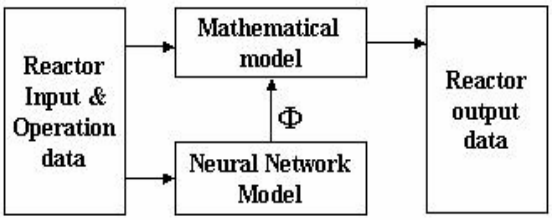

Fig. 7. Schematic diagram of hybrid model

4. Examples

4.1 Hybrid Model Test

The proposed hybrid model is well fitted with the real plant data. Fig. 8 compares the real plant data with simulation results using predicted catalyst deactivation factor (Φ). These figures show the performance of styrene monomer and ethylbenzene that are the main materials of this process. The simulation results show good performance of 0.4% relative error, compare with 1.7% relative error of mathematical model (*Lim* 2001a).

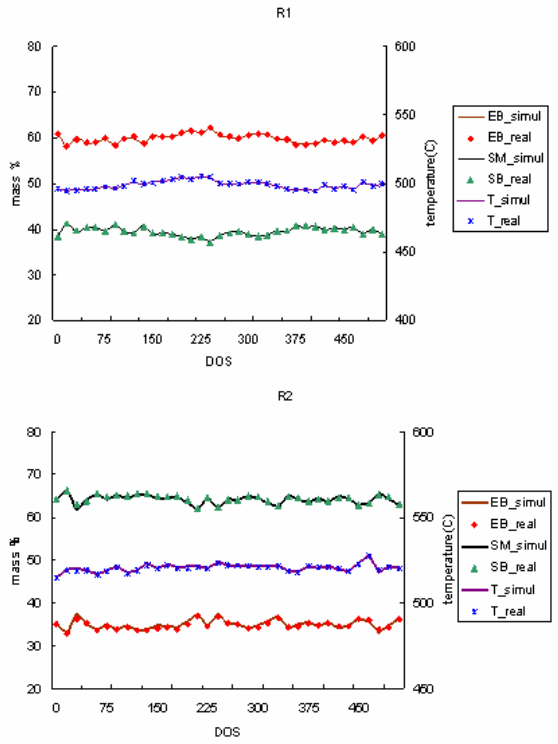

Fig 8. Comparison of simulation data and real plant data of reactors: a) reactor1, b) reactor2

4.2 Sensitivity Analysis for S/O ratio Change

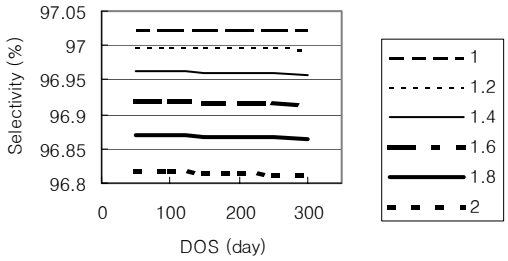

Fig. 9. Selectivity for S/O ratio change

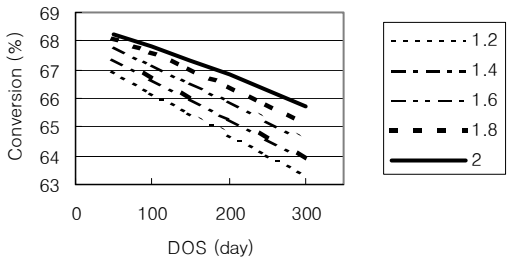

Fig. 10. Conversion for S/O ratio change

To see the simulation of the styrene monomer reactor, one of the operating variables is simulated. The input conditions are based on industrial data. Fig 9 shows the simulation results with changing S/O ratio and DOS (days on stream). According to the figure, the conversion of early DOS is similar. But, increasing DOS, higher S/O ratio condition shows higher conversion of styrene monomer from ethylbenzene and lower catalyst deactivation effect.

4.3 Optimization for Operating Conditions

The operating conditions are optimized using given boundaries. The objective function, given conditions and the results are shown in Table 2. This optimization result shows that higher S/O ratio, load, temperature, and pressure increase the conversion of ethylbenzene to styrene monomer and the production rate of styrene monomer.

Table 2 Calculated and specified values for constraints and objective function

Constraint	Current operation	Calculated	
		Starting	Optimum
S/O ratio	1.5	1.5	2.0
Load (%)	120.0	100.0	150.0
Reactor 1 inlet temperature (℃)	620.4	630.0	620.0
Reactor 2 inlet temperature (℃)	620.2	630.0	620.0
Reactor 2 outlet pressure (mmHg)	-400.0	-421.5	-400.0
Profit	54.0	38.8	79.3

Constraint range		
Operating variable	Min	Max
S/O ratio	1.0	2.0
Load (%)	100.0	150.0
Reactor1 inlet temperature (℃)	620.0	650.0
Reactor2 inlet temperature (℃)	620.0	650.0
Reactor2 outlet pressure (mmHg)	-450.0	-400.0

Objective function:

F = SMSALE – EBVALUE – CATVALUE + TLSALE – STEAMVALUE

SMSALE [$/day]=(SM price [$/ton])*(outlet SM flowrate [ton/day])
EBVALUE [$/day]=(EB price [$/ton])*((total EB inlet flow rate [ton/day])-(recycled EB flowrate [ton/day]))+(EB recycle price [$/ton])*(recycled EB flowrate [ton/day])
CATVALUE=(catalyst price [$/day])*$W_1$
TLSALE=(TL price [$/ton])*(outlet TL flowrate [ton/day])
STEAMVALUE=(steam price [$/day])*(inlet stream flowrate)*W_2

(W_1 & W_2: scalar weight)

5. CONCLUSIONS

In this study, the styrene monomer production process in an adiabatic redial flow reactor has been successfully modeled. This model has been developed via hybrid model in which the mathematical model and neural network models are combined. Also the performances of the developed model have been presented.

The mathematical model can reinforce the insufficient performance owing to lack of data that is not enough to use neural network models. And neural network model can give more accurate catalyst deactivation factor than one that a mathematical model can give. Although neural networks are not difficult to use, process variables should be known, and the quality of data is crucial to obtain reasonable results.

Using this model, determining optimal operating conditions and testing new operating conditions are performed easily. On the situation of changing catalyst, this simulator shows good performance because the catalyst parameters are updated using current process data.

6. ACKNOWLEDGEMENT

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NOMENCLATURE

BZ	Benzene
C_p	Heat capacity of gas flow [kJ/(kg·K)]
EB	Ethylbenzene
ET	Ethane
g_i	Mass fraction of component i [kg/kg]
f_i	Reaction rate of component i [kmol/(m^3·s)]
f_j	Reaction rate of reaction j [kmol/(m^3·s)]
F	Total mass flow rate [kg/s]
$H2$	Hydrogen
$H2O$	Water (steam)
ΔH_j	Enthalpy of reaction j [kJ/kmol]
i	Component
j	Reaction
k_i	Rate constant of component i
k_{i0}	Reference rate constant of component i
	k_{10}, k_{20} [Kmol/(kg·s·kPa)];
	k_{30}, k_{40}, k_{50}[Kmol/(kg·s·kPa^2)];
	k_{60} [Kmol/(kg·s·kPa^3)])
K_p	Equilibrium constant of main reaction [kPa]
m	Number of reaction
ME	Methane
MW_i	Molecular weight of component i [kg/kmol]
NC	Number of component
P	Pressure [kPa]
P_i	Partial pressure of component i
P_{in}	Inlet pressure [kPa]
P_{out}	Outlet pressure [kPa]
r	Radial direction [m]
R	Gas constant [kPa· m^3/(mol·K)]
SM	Styrene monomer
t	Time [day]
T	Temperature [K]
TL	Toluene
Φ	Catalyst deactivation factor

ν_{ij} Reaction stoichiometric coefficient of component i in reaction j

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