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Black-Box Modelling of Ethyl Acetate Reactive Packed Distillation Column

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Abstract

The development of two black-box (ARX and ARMAX) models for ethyl acetate reactive packed distillation process has been carried out in this work. The data used for the model development were generated by performing an experiment in a pilot plant using a unit step change in reflux ratio, a feed ratio of 1 and a reboiler duty of 560 W. The model orders used for the estimation of the model polynomial coefficients were determined by optimizing the Rissanen's Minimum Description Length criterion with the aid of MATLAB 7.12.0. The good agreements between the experimental and each of the ARX and ARMAX simulated top segment temperatures of the column have revealed that the models can be used to represent the process successfully. Furthermore, ARMAX model was discovered to be better in performance because of its higher calculated fit value but the ARX model was faster in getting to the steady state when a step input was applied to both models.

Keywords: *Reactive Packed Distillation Column, Reflux ratio, AutoRegressive with eXogenous Inputs (ARX) Model, AutoRegressive Moving Average with eXogenous Inputs (ARMAX) Model , Rissanen's Minimum Description Length, MATLAB/Simulink.*

1. Introduction

Reactive distillation is a process that combines both separation and chemical reaction in a single unit. It is very attractive whenever conversion is limited by reaction equilibrium (Balasubramhanya and Doyle III 2000) because it combines the benefits of equilibrium reaction with distillation to enhance conversion provided that the product of interest has the largest or the lowest boiling point (Taylor and Krishna 2000). It has a lot of advantages which include reduced investment and operating costs due to increased yield of a reversible reaction by separating the product of interest from the reaction mixture (Pérez-Correa *et al*. 2008), higher conversion, improved selectivity, lower energy consumption, scope for difficult separations and avoidance of azeotropes (Jana and Adari 2009).

However, due to the integration of reaction and separation, reactive distillation exhibits complex behaviors (Khaledi and Young 2005), such as steady state multiplicity, process gain sign changes (bidirectionality) and strong interactions between process variables (Jana and Adari 2009). These complexities have made the modeling of the reactive distillation process extremely difficult especially when the column type is a packed one and the reaction is solid-catalyzed. Thus, the development of a tangible model to represent this process is still a challenge to chemical engineers.

Researches have been carried out on the modeling of reactive packed distillation column using the first principle approach which normally incorporates many assumptions to develop theoretical models for the column. However, the development of rigorous theoretical models may not be practical for a complex process like this where the models require a large number of equations with a significant number of process variables and unknown parameters. An alternative approach is to develop an empirical model directly from

experimental data. This kind of modeling is referred to as black-box modeling.

Therefore, two black-box models, Autoregressive with eXogenous inputs (ARX) and Autoregressive Moving Average with eXogenous inputs (ARMAX), are developed and compared for ethyl acetate reactive packed distillation column in this work using the System Identification Toolbox 7.12.0 of Matlab Simulink (MathWorks 2011).

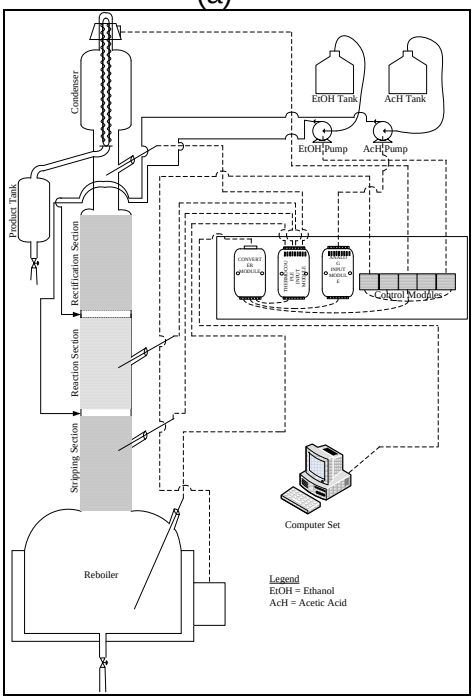
2. Methodology

2.1 Data Generation

The experimental pilot plant in which the experiments were carried out was a reactive packed distillation column (RPDC) set up as shown in Fig. 1a-b. The column had, excluding the condenser and the reboiler, a height of 1.5 m and a diameter of 0.05 m. The column consisted of a cylindrical condenser of diameter and height of 5 and 22.5 cm respectively. The main column section of the plant was divided into three subsections of 0.5 m each. The upper, middle and lower sections were the rectifying, the reaction and the stripping sections respectively. The rectifying and the stripping sections were packed with raschig rings while the reaction section was filled with Amberlyst 15 solid catalyst (the catalyst had a surface area of 5,300 m^2/kg, a total pore volume of 0.4 cc/g and a density of 610 kg/m^3). The reboiler was spherical in shape and had a total volume of 3 Litre. The column was fed with acetic acid at the top (between the rectifying and the reaction sections) while ethanol was fed at the bottom (between the reaction and the stripping sections) with the aid of peristaltic pumps which were operated with the aid of a computer via MATLAB/Simulink program. All the signal inputs (reflux ratio (R), feed ratio (F) and reboiler duty (Q)) to the column and the measured outputs (top segment temperature (T_{top}), reaction segment temperature (T_{rxn}) and bottom segment temperature (T_{bot})) from the column were sent and recorded respectively on-line with the aid of the MATLAB/Simulink computer program and electronic input-output (I/O) modules that were connected to the equipment and the computer system. The esterification reaction taking place in the packed column is given as:

$$CH_3COOH + C_2H_5OH \xleftrightarrow{K_{eq}} CH_3COOC_2H_5 + H_2O. \quad (1)$$

(a)



(b)

Fig. 1. Reactive packed distillation pilot plant: (a) Pictorial view; (b) Sketch view.

The data used for the development of the models were generated from the experiment carried in this column out by applying a unit step change to the reflux ratio and using feed ratio (volumetric flow rate of acetic acid/

volumetric flow rate of ethanol) and a reboiler duty of 1 and 560 W, respectively.

2.2 Modeling

2.2.1 Model Structure: Given the reactive packed distillation process, which has, apart from the disturbance *e*, the reflux ratio (R) and the top segment temperature (T_{top}) as the input and output respectively, that is represented as shown in Fig. 2, its general black-box model structure can be formulated as depicted in Fig. 3.

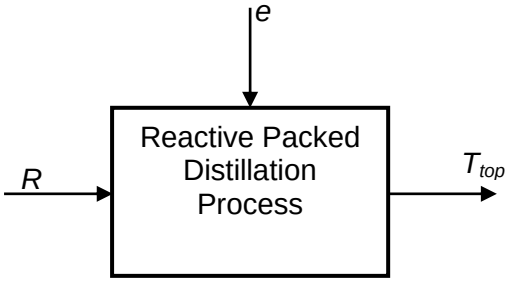


Fig. 2. Reactive packed distillation process.

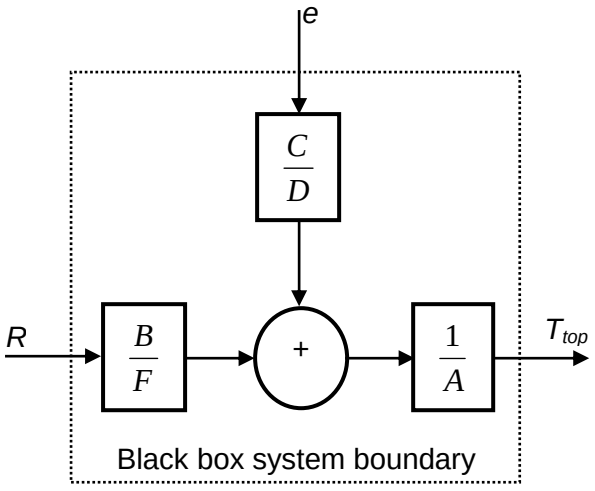


Fig. 3. Black-box model structure of reactive packed distillation process.

Considering the model structure shown in Fig. 2, the general mathematical expression for the black-box model of this process can thus be written as:

$$A(q)T_{top}(t) = \frac{B(q)}{F(q)}R(t-n_k) + \frac{C(q)}{D(q)}e(t), \quad (2)$$

where n_k is the number of delay. The polynomial coefficients contained in the equation above are expressed as:

$$A(q) = 1 + a_1q^{-1} + a_2q^{-2} + \cdots + a_{na}q^{-na}, \quad (3)$$

$$B(q) = b_1 + b_2q^{-1} + b_3q^{-2} + \cdots + b_{nb}q^{-nb+1}, \quad (4)$$

$$C(q) = 1 + c_1q^{-1} + c_2q^{-2} + \cdots + c_{nc}q^{-nc}, \quad (5)$$

$$D(q) = 1 + d_1q^{-1} + d_2q^{-2} + \cdots + d_{nd}q^{-nd}, \quad (6)$$

$$F(q) = 1 + f_1q^{-1} + f_2q^{-2} + \cdots + f_{nf}q^{-nf}. \quad (7)$$

For the ARX model structure of this process,

$$C(q) = D(q) = F(q) = 1, \quad (8)$$

while for the ARMAX model structure,

$$D(q) = F(q) = 1. \quad (9)$$

Based on this, for the reactive packed distillation process, with reflux ratio being the main input and top segment temperature being the output, considered in this work, the structures of the models to be developed are given as:

ARX model:

$$A(q)T_{top}(t) = B(q)R(t-n_k) + e(t); \quad (10)$$

ARMAX model:

$$A(q)T_{top}(t) = B(q)R(t-n_k) + C(q)e(t). \quad (11)$$

2.2.2 Selection of Model Orders: The selection of appropriate model orders (*na*, *nb*, *nc* and *nk* which stand for number of poles, number of zeros plus 1, number of C coefficients and number of delays respectively) is very important when developing any black-box model. During the model development, the optimum values of these model orders are necessary to be determined in order to avoid under-fitting or over-fitting of the developed model equation. Many criteria (such as AIC, BIC, and MDL) are available in the literature for the optimum selections of these model orders. In this work, the MDL (Rissanen's Minimum Description Length) criterion

(shown in Eq. (12) below) was used because it allows the shortest possible description of the observed data (Ljung 1999).

$$MDL = V\left(1+\frac{d\log(N)}{N}\right). \quad (12)$$

In Eq. (12),

V is the loss function;

d is the total number of parameters in the structure; and

N is the number of data points used for the estimation.

2.2.3 Parameter Estimation: The estimations of the model parameters were carried out in MATLAB Environment by minimizing, using the Levenberg-Marquardt algorithm as the search method, the absolute of the errors between the experimentally measured dynamic responses ($T_e(t)$) and the theoretically simulated outputs ($T_s(t)$) of the developed model equations, as shown in Eq. (13). That is,

$$\min|e(t)| = \min|(T_e(t) - T_s(t))|. \quad (13)$$

The subscripts *e* and *s* in Eq. (13) stand for experimental and simulated, respectively.

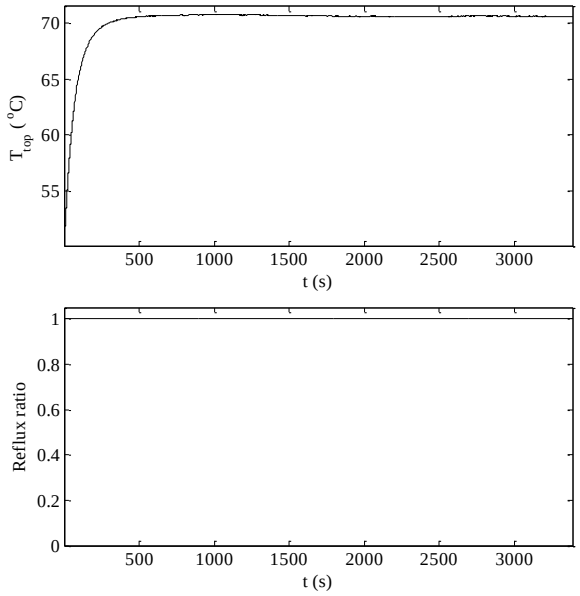


Fig. 4. Experimental input-output data: (Up) Output; (Down) Step input.

3. Results and Discussion

The data acquired from the experiment carried out in the pilot plant are as shown in Fig. 4. It can be observed from Fig. 4 that a change in the reflux ratio from total reflux to a unit step resulted in a change in the temperature of the top segment of the column.

Using the data acquired from the experiment, the model orders were estimated by optimizing the MDL criterion. The optimum model orders obtained from the optimization of the criterion were $n_a = 5,\ n_b = 4$ and $n_k = 4.$

Thereafter, the model orders were used to develop the black-box models (ARX and ARMAX) for the reactive packed distillation process in MATLAB 7.12.0 environment. That is, the polynomial coefficients of the models were estimated using the obtained model orders. The polynomial coefficients estimated are as outlined below.

For ARX model:

$$A(q) = 1 - 0.2575q^{-1} - 0.411q^{-2} + \cdots + 0.003887q^{-3} - 0.01404q^{-4} - 0.2856\,q^{-5} \quad (14)$$

$$B(q) = 0.8383q^{-4} + 0.8383\,q^{-5} + \cdots + 0.8383q^{-6} + 0.01368q^{-7} \quad (15)$$

For ARMAX model:

$$A(q) = 1 - 0.5269q^{-1} - 1.493q^{-2} + \cdots + 0.3158q^{-3} + 1.004q^{-4} + \cdots - 0.3003q^{-5} \quad (16)$$

$$B(q) = 0.1934\,q^{-4} + 0.1934q^{-5} + \cdots + 0.1934\,q^{-6} - 0.5684q^{-7} \quad (17)$$

$$C(q) = 1 - 0.04597q^{-1} - 1.458q^{-2} + \cdots - 0.3092q^{-3} + 0.8265q^{-4} \quad (18)$$

The developed models were then simulated and their simulated results were compared to that of the experimental top segment temperatures of the column. Shown in Fig. 5 is the comparison between the

experimental and the simulated top segment temperatures of the column for the ARX model. As can be seen from Fig. 5, there is a good agreement between the experimental and the simulated top segment temperatures.

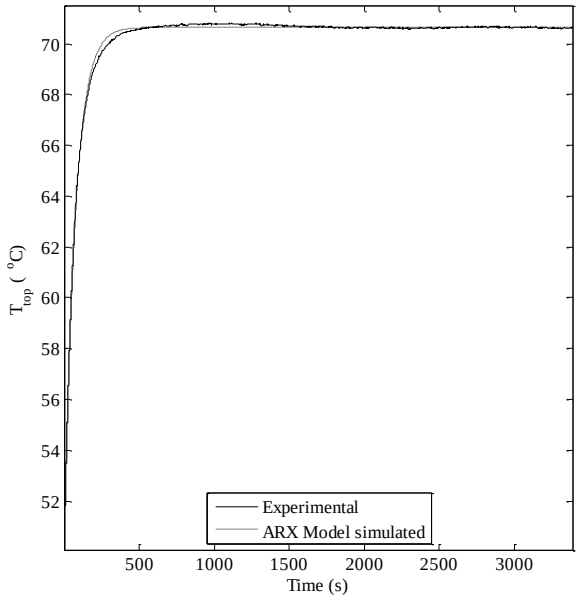


Fig. 5. Comparison between experimental and ARX model simulated top segment temperatures.

Also, the simulated values of the top segment temperatures obtained from ARMAX model were compared to that of the experimental ones as shown in Fig. 6. As in the case of the ARX model simulation, similar good agreements were observed between the experimental and the simulated values.

Moreover, the two sets of the simulated temperatures were compared with each other by calculating their fit values (using the expression given in Eq. (19)) and the mean squared errors (MSE). The calculated fit values and the mean squared errors are as shown in Table 1 below.

$$fit = \frac{1 - norm(T_s - T_e)}{norm(T_e - mean(T_e))} \cdot 100 \qquad (19)$$

It has been revealed from Table 1 that, even though the difference was not too much, the performance of the ARMAX model with the fit value of 96.7827% was better than of the ARX model that had a fit value of 95.4047%. Since the fit value refers to the percentage of the data that the data could account for, then it means that the ARMAX model developed could account for 96.7827% of the experimental data while the developed ARX could account for 95.4047% of the data.

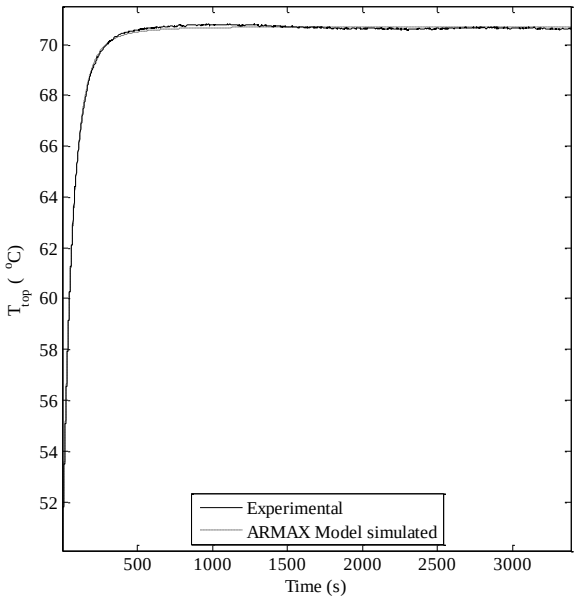


Fig. 6. Comparison between experimental and ARMAX model simulated top segment temperatures.

Table 1. Fit values and MSE of the models.

Model	Fit value (%)	MSE
ARX	95.4047	0.009530
ARMAX	96.7827	0.004671

Another criterion used to compare the performances of the developed models was the mean squared error. The mean squared error of each of the models was calculated also with the aid of MATLAB and the values obtained can be seen in Table 1 above. From the table, it was observed that the mean squared error of the developed ARX model which was calculated to be 0.009530 was higher than that of the ARMAX model which was calculated to be 0.004671. This is another indication of the better performance of the developed ARMAX model for the ethyl acetate reactive packed distillation process over the developed ARX model for the process because the smaller the mean squared error of a model, the better the model.

Now, considering the relationship between the fit values and the mean squared errors of the models, as can be observed from Table 1, the model with the higher fit value had the lower mean squared error. In other words, especially for this process, the fit value has been found to be inversely proportional to the mean squared error.

In order to have ideas about the dynamics of the developed models, the two models were simulated with the aid of Simulink 7.7 version of MATLAB 7.12.0 by applying a step input to each of them and their dynamic responses were recorded. The obtained dynamic responses for the ARX and ARMAX models are as shown in Figs. 7 and 8, respectively.

From Fig. 7, it was observed that the response of the developed ARX model to the applied step input resembled that of a first order system or of a higher order system with overdamped behavior. However, the transfer function analysis of the model revealed that the system was not first order. Therefore, it is very clear that the developed ARX model is a higher order model with overdamped response.

Similar dynamic response was also observed in the case of the developed ARMAX model as shown in Fig. 8. The existence of the higher order dynamic for the reactive packed distillation column was attributed to the complex nature of the process.

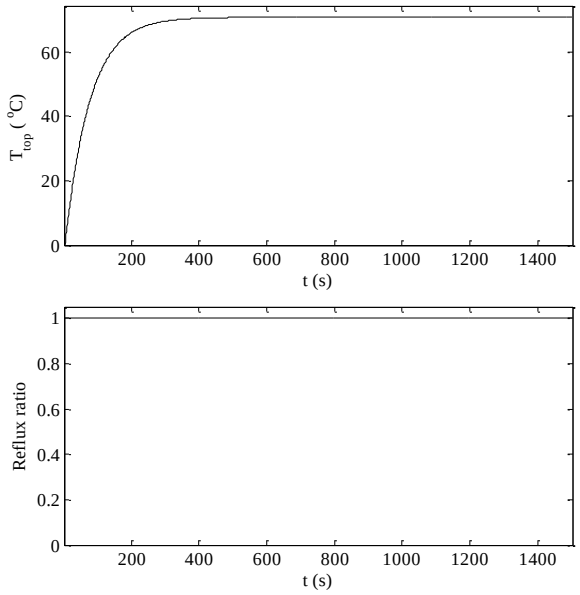


Fig. 7. Dynamic responses of ARX model to a step input.

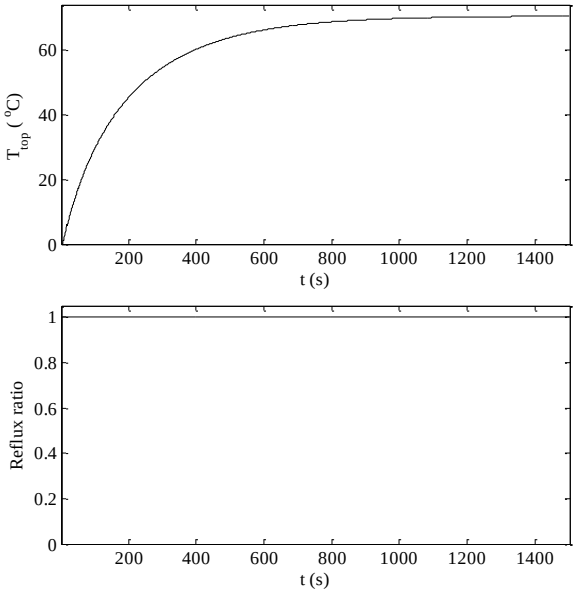


Fig. 8. Dynamic responses of ARMAX model to a step input.

The comparison between the dynamic responses of the two models is also shown in Fig. 9. It can be seen from Fig. 9 that the two models were stable because they were able to get to the steady state successfully.

However, despite the higher fit and lower mean squared error values of the developed ARMAX model than that of the developed ARX model, the response of the ARX model was found to be faster in getting to the steady state than that of the ARMAX model.

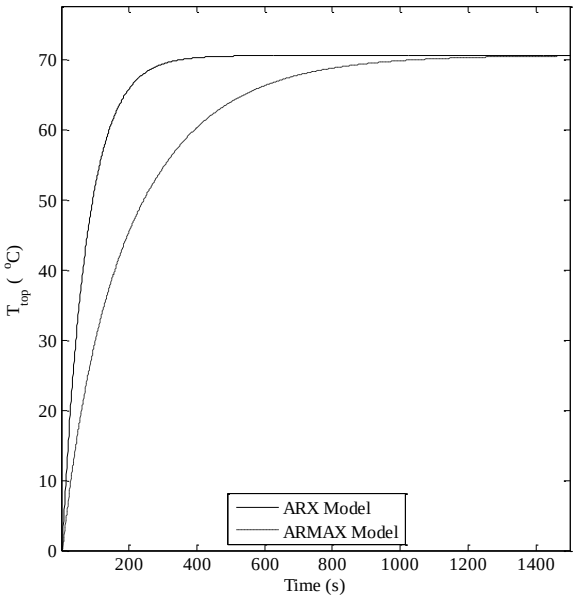


Fig. 9. Comparison between the dynamic responses of ARX and ARMAX models to a step input.

4. Conclusion

The good comparisons between the experimental and the simulated values of the top segment temperatures of the developed ARX and ARMAX models for the reactive packed distillation process has shown that the models can be used to represent the behavior of the process successfully. However, due to its higher fit value and lower mean squared error, ARMAX model was discovered to be better than ARX one for this process.

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