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ARX and ARMAX Modelling of olefin metathesis reactive distillation process

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ABSTRACT

This work has been carried out to develop AutoRegressive with eXogenous Inputs (ARX) and AutoRegressive Moving Average with eXogenous Inputs (ARMAX) models for olefin metathesis process accomplished using a reactive distillation column. To achieve the work, Aspen HYSYS model of the process was first developed and simulated to obtain the data used to formulate the transfer function model of the process. The transfer function model was tested for stability, which was ascertained from the attainment of steady state by the output of the process, through simulation and, later, run using random number to generate the data required for the development of ARX and ARMAX models of the process that were also simulated and their performances compared. The results obtained revealed that the developed ARX and ARMAX models had moderate orders. Also, good agreements were found to exist between the measured mole fraction of bottom cis-2-hexene and the simulated ones, implying that the developed models were good representatives of the process. Moreover, the lower mean of squared error value and the higher fit value of the developed ARMAX model revealed that it was able to represent the process better than the developed ARX model. It is, therefore, recommended that ARMAX model should preferably be used to represent the olefin metathesis reactive distillation process for further studies.

Keywords: Olefin metathesis, reactive distillation, Aspen HYSYS, ARX, ARMAX, MATLAB/Simulink.

INTRODUCTION

Olefin metathesis is a process of converting an olefin into lower and higher molecular weight olefins. It is a class of reactions that are ideally suited for reactive distillation applications because many of the reactions are in liquid phase at ambient to moderate conditions, and the reactants and the products are similar chemicals; so, they exhibit very little deviation from Raoult's law. Moreover, the relative order of olefin boiling points can be determined based on their molecular weights. Thus, for metathesis reactions, the boiling points of the products straddle those of the reactants. In such a case, reactive azeotropes do not form, and this allows easy removal of the products, thereby minimizing side reactions or additional metathesis of products and overcoming reaction equilibrium limitations. A commercial process for conducting 1-butene metathesis within a distillation column was patented by Dow Chemical Co. [1]. They claimed that conducting reaction and separation simultaneously was able to reduce by-product formation and overcome equilibrium limitations and, thus, resulted in increased selectivity and higher yield over conventional series processing ([2],[3]). This process of conducting reaction and separation simultaneously is, actually, referred to as "reactive distillation".

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 ([4-9]). It is an excellent alternative to conventional flow sheets with different reaction and separation sections ([10],[11]) as it combines the benefits of equilibrium reaction with distillation to enhance conversion ([12], [13]). Combining reaction and distillation has several advantages such as shift of chemical equilibrium and an increase of reaction conversion by simultaneous

reaction and separation of products, suppression of side reactions and utilization of heat of reaction for mass transfer operation ([14],[15]). The utilization of heat of reaction for mass transfer operation, which resulted into low external energy consumption of the process normally gives rise to reduced investment and operating costs ([16]), which is much desired in any process industry.

However, due to the integration of reaction and separation, reactive distillation exhibits complex behaviors ([17], [12],[18]) such as steady state multiplicity, process gain sign changes (bidirectionality) and strong interactions between process variables ([19],[12]). These complexities have made the modeling of the reactive distillation process extremely difficult. Thus, the development of a tangible model to represent this process is still a challenge to process engineers ([12]).

Consequently, researches have been carried out on representing a reactive distillation processes using models such as ARX and ARMAX. For instance, Giwa and Karacan (2012a)[18]developed two black-box models (AutoRegressive with eXogenous Inputs (ARX) and AutoRegressive Moving Average with eXogenous Inputs (ARMAX) models) using experimental data to represent a reactive distillation process used for ethyl acetate production. They discovered in the work that ARMAX model was able to perform better than ARX model because its calculated fit value was higher. Also, Giwa and Karacan (2012e)[6] developed two nonlinear black-box (tree partition and sigmoid network NARX) models for the reactive distillation process producing ethyl acetate from the esterification reaction between acetic acid and ethanol, and they discovered from the studies carried out that sigmoid network NARX model was better than tree partition NARX model for the reactive distillation process considered in the work.

It has, thus, been discovered that very few researches have been carried out on representing reactive distillation processes using models such as ARX and ARMAX. Besides, no one was found concerning olefin metathesis. Therefore, this research study was aimed at covering this gap by developing ARX and ARMAX models for olefin metathesis process accomplished in a reactive distillation column.

MATERIALS AND METHODS

2.1 Transfer Function Model Development

The Parametric Utility incorporated Aspen HYSYS ([20])model of the olefin metathesis reactive distillation process from which the data used for the development of the transfer function model of the process is shown in Figure 1. The feed of the process that was fed at room temperature (25 ^{o}C) and pressure (1 atm) at the rate of 25 mL/min into the eleventh (11th) stage of the reactive distillation column having 21 stages was pure *cis*-2-pentene. The pressure of the condenser employed, which was a total type, was 1 atm while that of the reboiler was 1.1 atm.

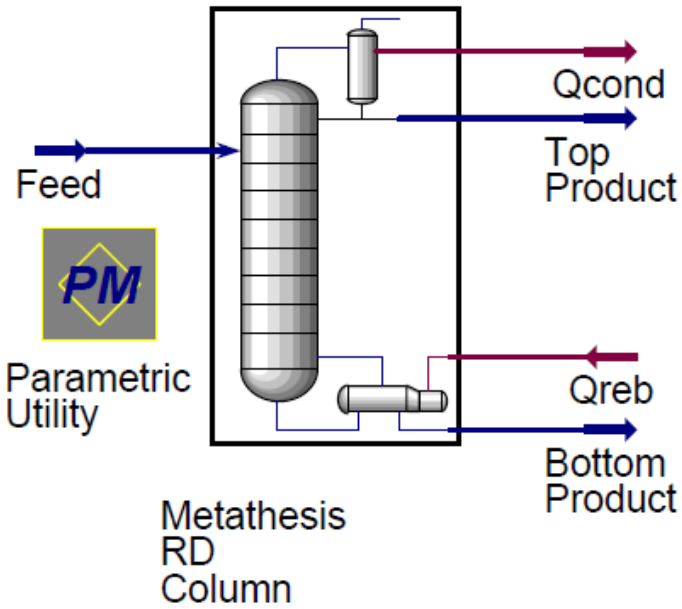

Figure 1. Developed Aspen HYSYS model of the olefin metathesis reactive distillation process for data generation

The metathesis reaction (Equation 1) taking place between the seventh and the fifteenth stage of the column was an equilibrium type occurring in liquid phase, and its equilibrium constant was calculated using the Gibbs free energy

relationship that is given in Equation (2). Owing to the reactive distillation process occurring in the column, the solver used to simulate the developed Aspen HYSYS model was Sparse Continuation Solver while the Fluid Package employed was Peng-Robinson-Stryjek-Vera (PRSV)equation of state.

$$2C_5H_{10} \xleftrightarrow{K_{eq}} C_4H_8 + C_6H_{12} \tag{1}$$

$$ln(K_{eq}) = \frac{\Delta G_{reaction}}{R_g T} \tag{2}$$

The initial simulation of the developed Aspen HYSYS model was carried out using a reboiler duty and a reflux ratio of 1 kW and 2, respectively. Thereafter, the required data were generated by varying the input parameters (reboiler duty and reflux ratio) within the ranges given in Table 1. The output of the process was taken to be the mole fraction of *cis*-2-hexene obtained from the bottom section of the reactive distillation column.

Table 1. Limits of the input variables used

Parameter	Low limit	High limit
Reboiler duty (kW)	0.5	1.5
Reflux ratio	1	3

After the data were obtained, a transfer function model of the process having the form given in Equation (3) was formulated using the*mfile* codes written in MATLAB ([21]) with the main command of the code given as *pem*.

In order to verify whether theformulated transfer functionof the process would be stable or not, it was simulated using its Simulink model developed and shown in Figure 2.

$$x_{cis-Hexene}(s) = \frac{K_p e^{(-T_{dp}s)}}{\tau_p s + 1} Q(s) + \frac{K_d e^{(-T_{dd}s)}}{\tau_d s + 1} R(s) \tag{3}$$

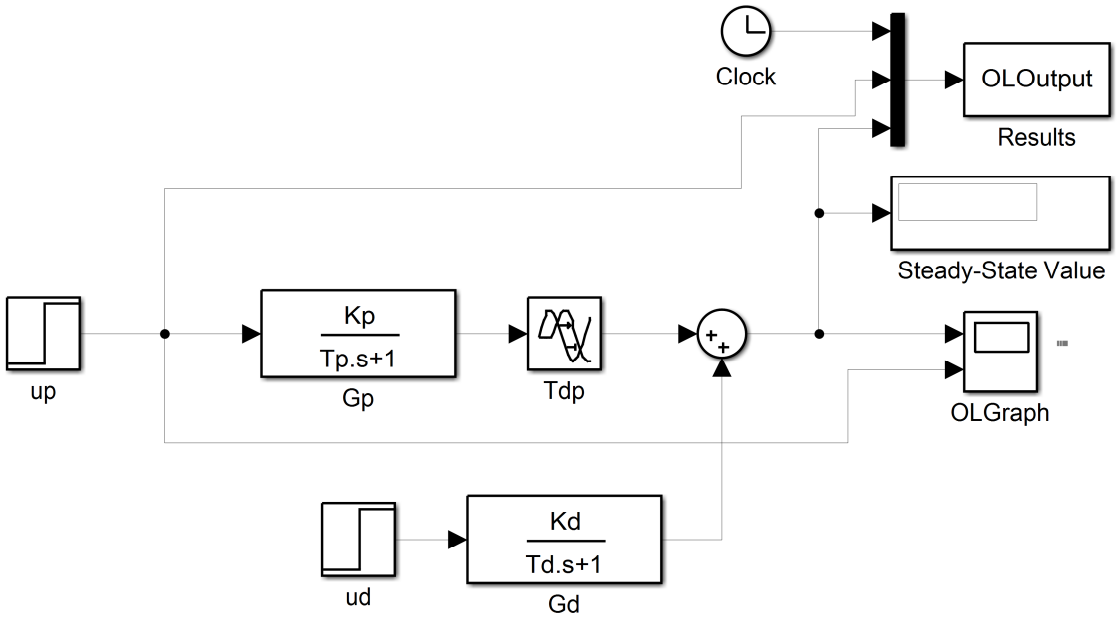

Figure 2. Simulink model of the olefin metathesis reactive distillation process for simulation

After ascertaining that the transfer function model of the process was a stable one, random number input was passed into it to generate the data required for the development of the ARX and ARMAX models of the olefin metathesis reactive distillation process.

2.2 ARX and ARMAX Modelling Data Acquisition

The data used for the development of the ARX and ARMAX models of the olefin metathesis reactive distillation process were acquired by simulating the transfer function model developed for the process with the aid of Simulink

using random number. Shown in Figure 3 is the developed Simulink model of the process with the incorporated random number block.

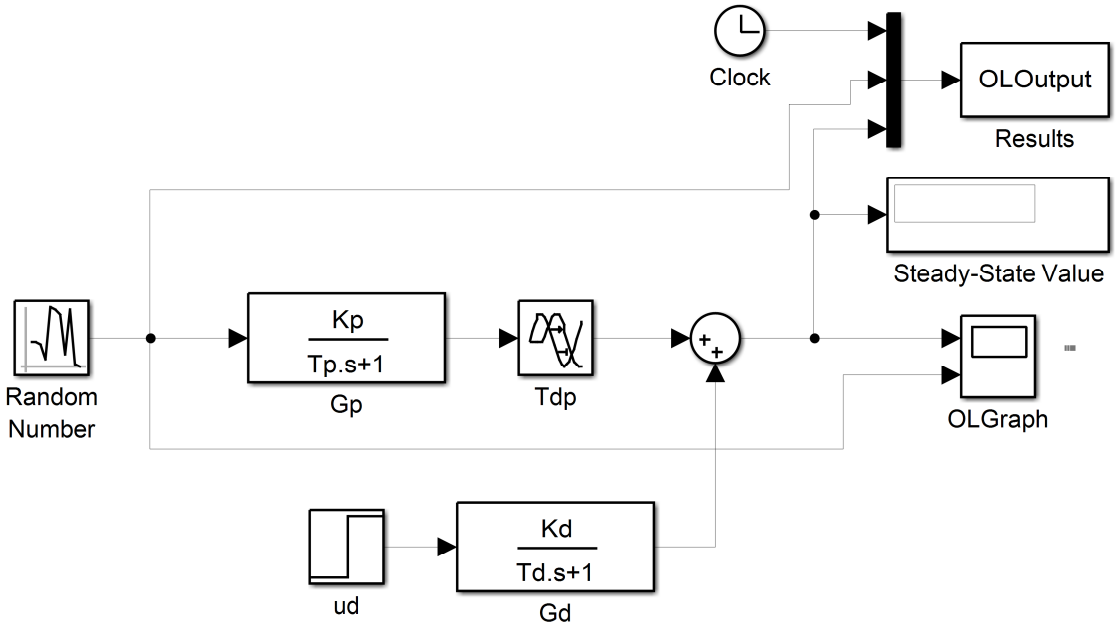

Figure 3. Simulink model of the process for generation of ARX and ARMAX model formulation data

Two different data sets were generated. One was used for model development and the other was used for validation of the developed models. The random number parameters used for the data generation were as given in Table 2.

Table 2. Random number parameters used

Parameter	Random number for	
	Model formulation data	Model validation data
Mean	0.1	0.2
Variance	1.2	1.5
Seed	2	1

2.3 Model Development

2.3.1 Selection of Model Orders

The selection of appropriate model orders (*na*, *nb*, *nc* and *nk*) of the models developed was accomplished using the MDL (Rissanen's Minimum Description Length) criterion (Equation 4) because it was found in the literature that it allows the shortest possible description of the observed data [22].

$$MDL = V\left(1+\frac{d\,log(N)}{N}\right) \tag{4}$$

where
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.

2.3.2ARX Modelling

Denoting the input variable, $Q(t)$, as $u(t)$, and the output variable, $x_{cis-Hexene}(t)$, as $y(t)$, for the ARX modelling of the olefin metathesis reactive distillation process considered in this work, its structure, given in form of a difference equation, is given as:

$$y(t)+a_1y(t-1)+\cdots+a_{na}y(t-n_a)=b_1u(t-n_k)+\cdots+b_{nb}u(t-n_k-n_b+1)+e(t) \tag{5}$$

where

$y(t)$ is the output at time t, n_a, an order of the model, is the number of poles, n_b, also an order of the model, is the number of zeroes plus 1, n_k is the number of input samples that occur before the it (input) affected the output, also called the dead time (delay) in the system, $y(t-1)\cdots y(t-na)$ are the previous outputs on which the current output was depending, $u(t-nk)\cdots b_{nb}u(t-nk-nb+1)$ are the previous and delayed inputs on which the current output was depending and $e(t)$ is the white-noise disturbance value.

The ARX model difference equation can also be written as:

$$A(z)y(t) = B(z)u(t-n_k)+e(t) \quad (6)$$

where z is the delay operator and

$$A(z) = 1 + a_1 z^{-1} + a_2 z^{-2} + \cdots + a_{n_a} z^{-n_a} \quad (7)$$

$$B(z) = b_1 + b_2 z^{-1} + b_3 z^{-2} + \cdots + b_{n_b} z^{-n_b+1} \quad (8)$$

2.3.3ARMAX Modelling

Using the same notation that the input variable, $Q(t)$,is represented as $u(t)$, and the output variable, $x_{cis-Hexene}(t)$, is represented as $y(t)$, the structure of the ARMAX model of the process is given as:

$$y(t)+a_1 y(t-1)+\cdots+a_{n_a} y(t-n_a) = b_1 u(t-n_k)+\cdots+b_{n_b} u(t-n_k-n_b+1)+c_1 e(t-1)+\cdots+c_{nc} e(t-n_c)+e(t) \quad (9)$$

where n_c is the number of c coefficients. The ARMAX difference equation can as well be written as:

$$A(z)y(t) = B(z)u(t-n_k)+C(z)e(t) \quad (10)$$

where

$$A(z) = 1 + a_1 z^{-1} + a_2 q^{-2} + \cdots + a_{n_a} z^{-n_a} \quad (11)$$

$$B(z) = b_1 + b_2 z^{-1} + b_3 z^{-2} + \cdots + b_{n_b} z^{-n_b+1} \quad (12)$$

$$C(z) = 1 + c_1 z^{-1} + c_2 z^{-2} + \cdots + c_{n_c} z^{-n_c} \quad (13)$$

2.3.4 Parameter Estimation

After selecting the model orders and defining the structure of the models, the estimations of the parameters contained in the model were carried out in with the aid of MATLAB using the Levenberg-Marquardt algorithm as the search method to minimize the absolute of the errors between the measured responses $(y_m(t))$ and the simulated ones $(y_s(t))$ of the developed models (see Equation 14).

$$min|e(t)| = min|[y_m(t)-y_s(t)]| \quad (14)$$

RESULTS AND DISCUSSION

The input data generated that were used to obtain the output data (bottom section *cis*-2-hexene mole fraction) from the olefin metathesis carried out in a reactive distillation column are given in Figures 4 and 5. Shown in Figure 4 are the data for the reboiler duty while the data for the reflux ratio are given in Figure 5. It was observed from the data given in the figures (Figures 4 and 5) that they were within the limits used for thesimulation, as given in Table 1. This was found to be an indication that the developed Aspen HYSYS model of the process used for the generation of these data was working very well.

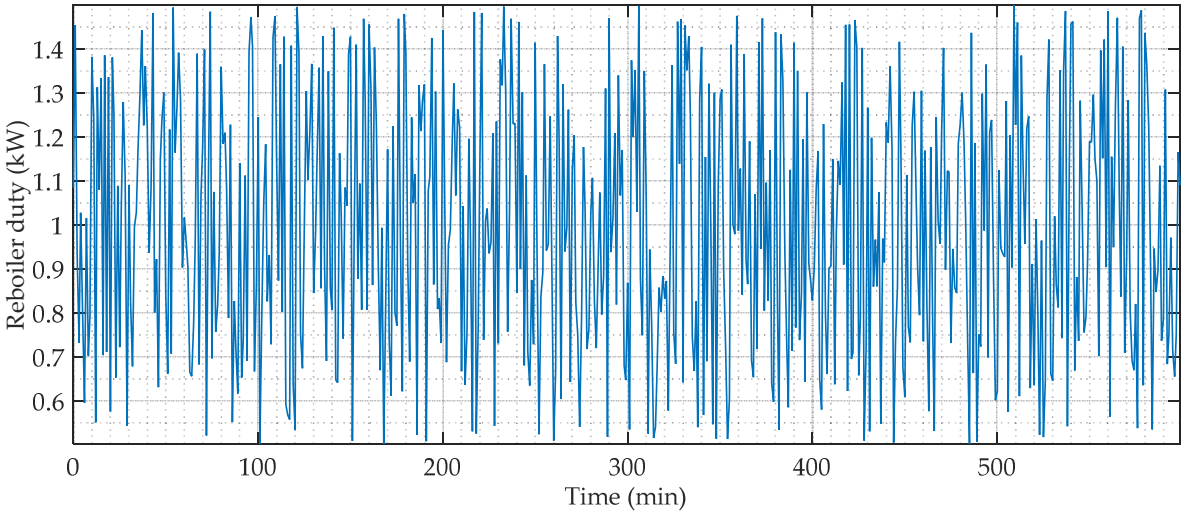

Figure 4. Reboiler duty (an input) of the Aspen HYSYS olefin metathesis reactive distillation process

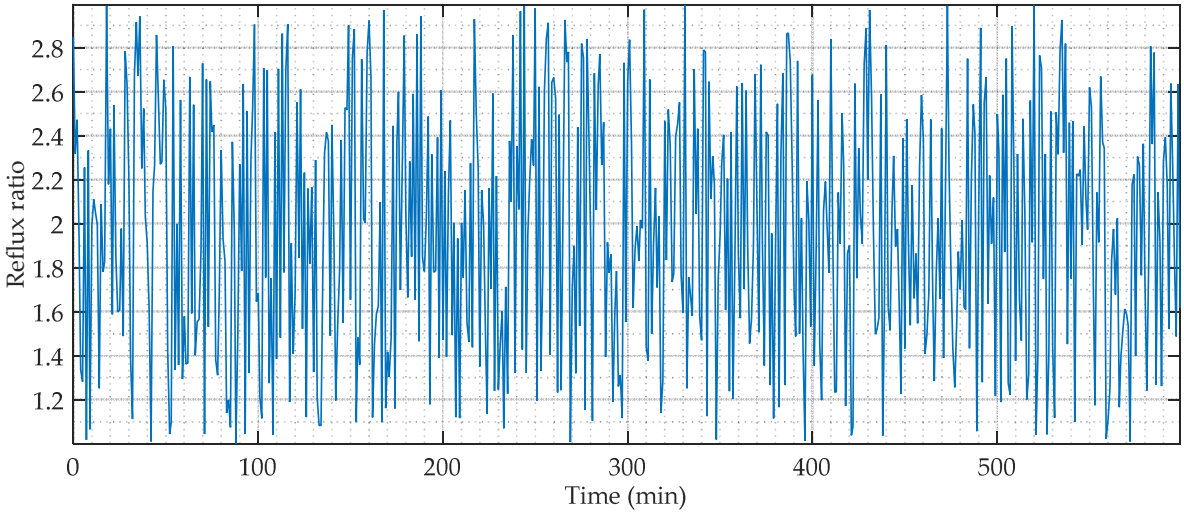

Figure 5. Reflux ratio (an input) of the Aspen HYSYS olefin metathesis reactive distillation process

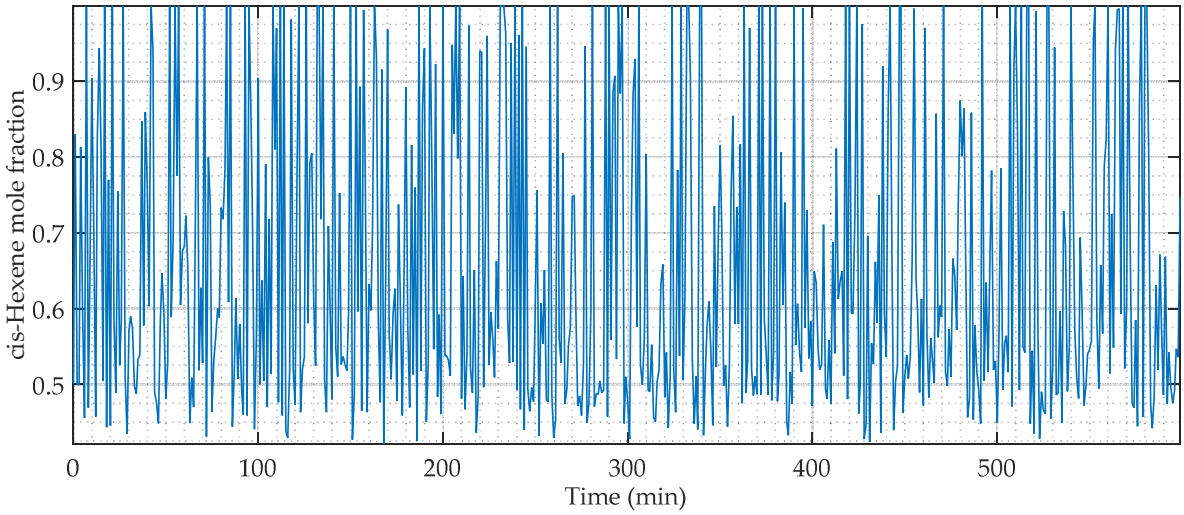

Figure 6. *cis*-2-Hexene mole fraction measured from the simulation of the Aspen HYSYS olefin metathesis reactive distillation process

Given in Figure 6 are the mole fraction data of the produced *cis*-2-hexene obtained upon the application of the input data generated and given in Figures 4 and 5. According to the figure, the mole fraction data of *cis*-2-hexene was found to occur in random manner because the data used to generate them were also in that manner. This has shown that the nature of the output obtained from the process was a function of the inputs. That observation was found to

be an indication of the fact that the output (the dependent) variable was a function of the input (the independent) variables, which is another evidence confirming the validity of the developed model.

Using the data obtained to formulate a transfer function model for the system, the obtained model was as given in Equation (15). The transfer function was relating the output of the process, which was the mole fraction of *cis*-2-hexene obtained from the reboiler section of the column, to the reboiler duty (input variable) and the reflux ratio (disturbance variable) of the column. As such, the transfer function had two parts. The part relating the output variable to the input variable and the other relating the output to the disturbance variable of the process. From the equation, it was observed that the disturbance was most likely to affect the process immediately upon start-up because its (the disturbance variable's) dead time was estimated to be zero (0).

Also, by considering the time constants of the two parts of the transfer function model, it was discovered that the process would be faster with respect to the disturbance (reflux ratio) than with respect to the main input (reboiler duty) of the process. This was seen from the comparison made between the values of the time constants of the two parts of the transfer function model of the process.

$$x_{cis-Hexene}(s) = \frac{0.3384e^{(-0.585s)}}{4.1156s+1}Q(s) + \frac{0.16088}{2.9863s+1}R(s) \quad (15)$$

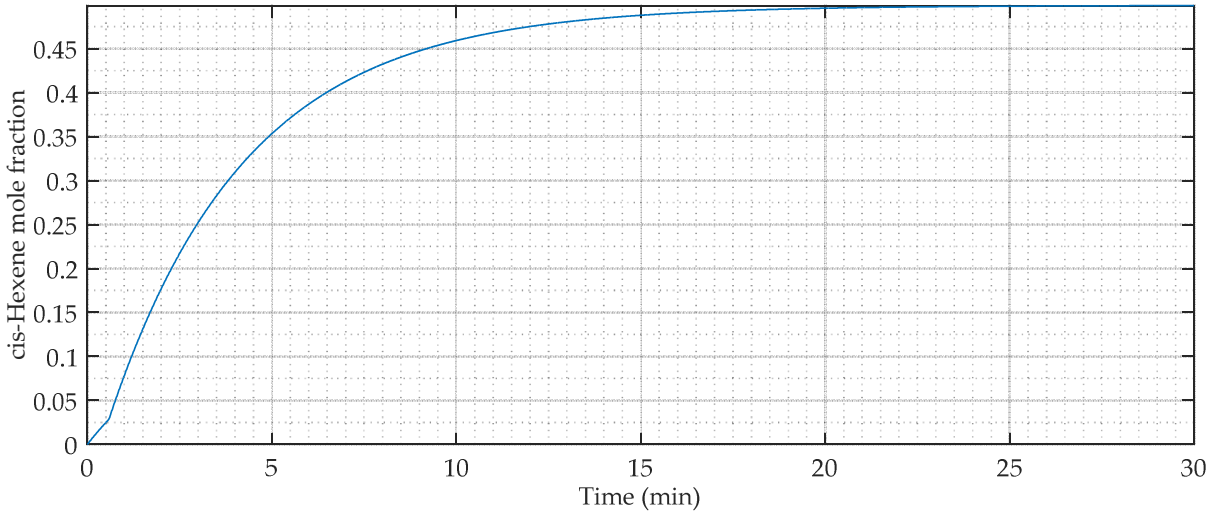

Figure 7. Open loop response of the developed transfer function model of process

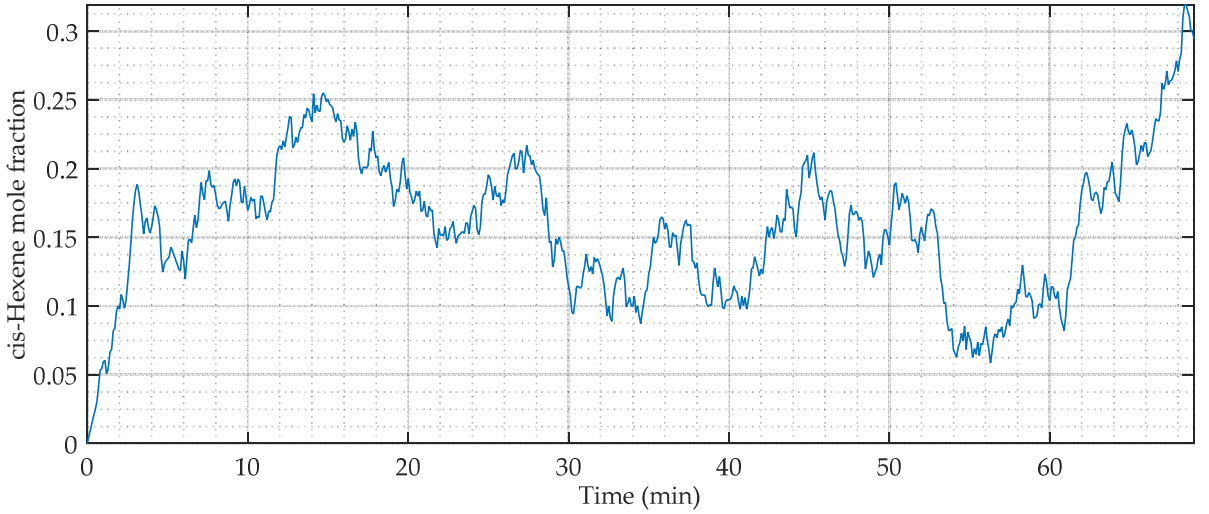

Figure 8. Data generated for ARX and ARMAX model formulation

Furthermore, the response obtained when the developed transfer function model of the process was simulated in order to test whether it would be stable or not by applying a unit step change to both the input and the disturbance variables is given in Figure 7. It was clear from the response shown in the figure that the system was able to get to a

steady state within 30 min. This observation was found to be very favourable because the settling time of the system was found not to be too much for a process like this.

After ascertaining that the transfer function model of the system developed was stable, it was used to generate the data required for the development of the ARX and ARMAX models by passing random number to them as the input, and the data generated were as given in Figures 8 and 9. Given in Figure 8 were the data used for the formulation of the models while the developed models were validated using the data given in Figure 9. It could be noticed from the data in the figures (Figures 8 and 9) that their trends were different. This was intentionally made like that so that the robustness of the models (ARX and ARMAX) would be clearly observed.

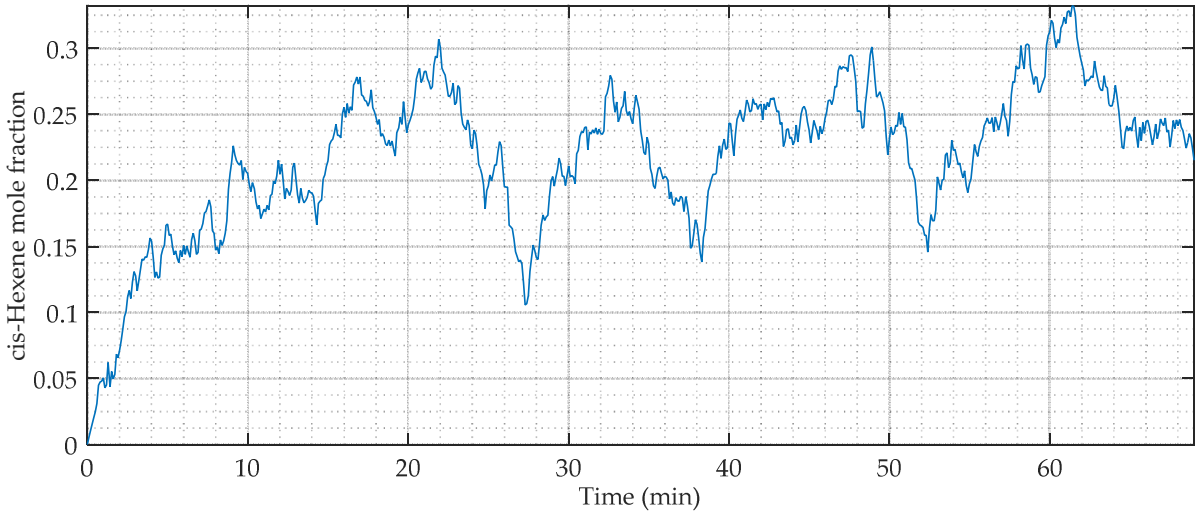

Figure 9. Data generated for ARX and ARMAX model validation

Using the data given in Figures 8 and 9 to develop the ARX and ARMAX models for the olefin metathesis reactive distillation process used for the production of *cis*-2-hexene in which the input variable of the model was the reboiler duty and the output variable was taken as the mole fraction of the *cis*-2-hexene obtained from the bottom section of the column, the developed model is given as:

ARX Model:
With reference to Equations (6) – (8),

Orders: $n_a = 3;\ n_b = 4;\ n_k = 6.$

$$A(z) = 1 - 2.953z^{-1} + 2.906z^{-2} - 0.9533z^{-3} \quad (16)$$

$$B(z) = 0.001218z^{-6} + 0.004496z^{-7} - 0.01246z^{-8} + 0.006744z^{-9} \quad (17)$$

ARMAX Model:
With reference to Equation (10) – (13),

Orders: $n_a = 3;\ n_b = 4;\ n_c = 4;\ n_k = 6.$

$$A(z) = 1 - 2.965z^{-1} + 2.931z^{-2} - 0.9654z^{-3} \quad (18)$$

$$B(z) = 0.001218z^{-6} + 0.004481z^{-7} - 0.01253z^{-8} + 0.00683z^{-9} \quad (19)$$

$$C(z) = 1 + 0.3763z^{-1} + 0.445z^{-2} + 0.4359z^{-3} - 0.5389z^{-4} \quad (20)$$

It was discovered from the equations of the models (Equations 16 and 17 for ARX and Equations 18 – 20 for ARMAX) given that their orders were moderate because too high orders might make the models to be cumbersome and too complex to be handled.

After obtaining the ARX and the ARMAX models of the process, they were simulated using the data generated for validation and shown in Figure 9, and the results of the simulation were as given in Figures 10 and 11 for the ARX model and the ARMAX model of the olefin metathesis reactive distillation process, respectively. It was observed from the figures that there were good agreements between the measured mole fraction of *cis*-2-hexene produced and the simulated ones using the developed ARX and ARMAX models.

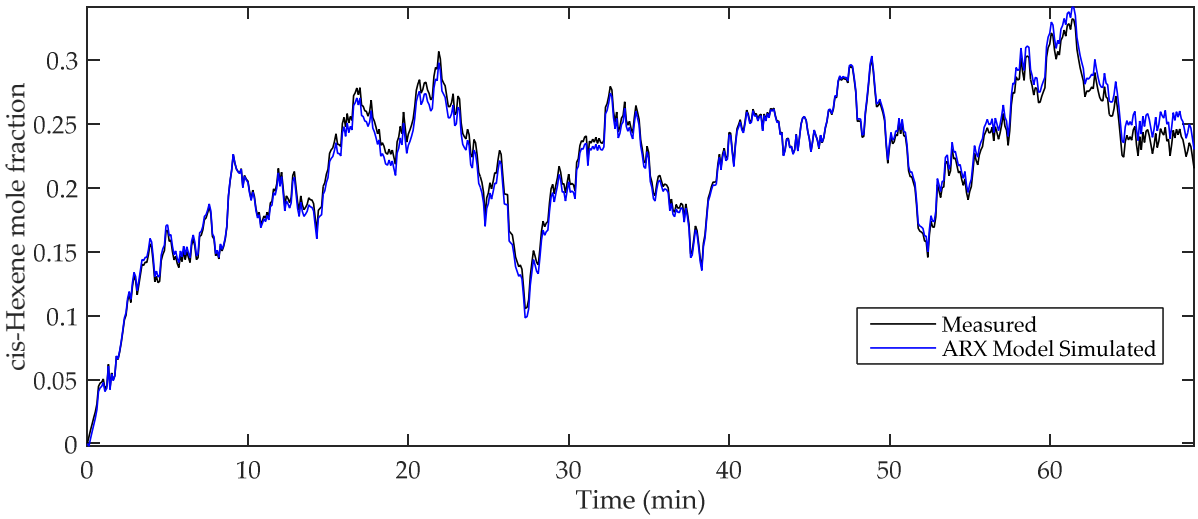

Figure 10. Measured and ARX model simulated mole fraction of *cis*-2-hexene

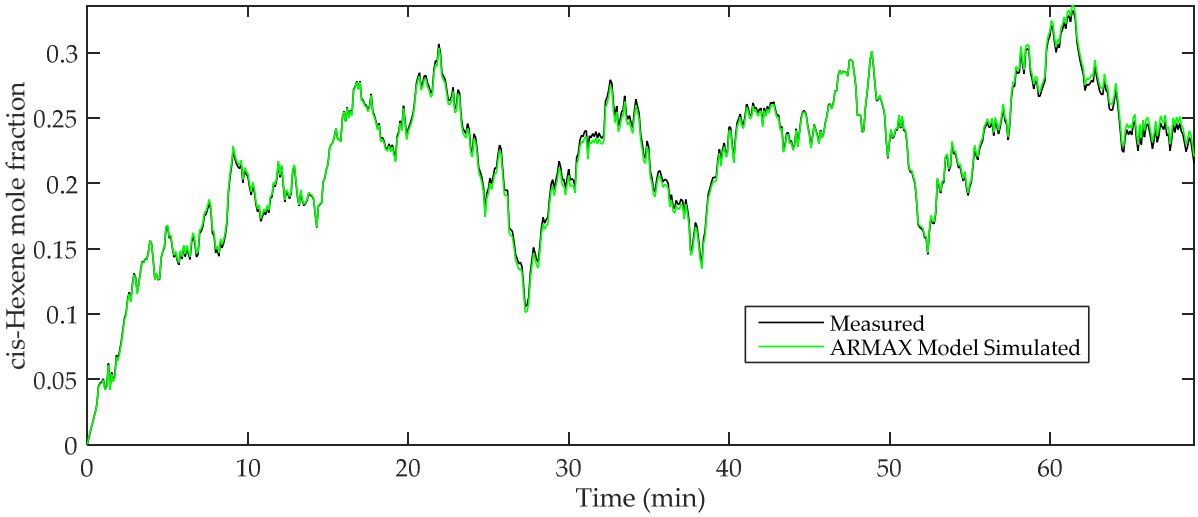

Figure 11. Measured and ARMAX model simulated mole fraction of *cis*-2-hexene

Furthermore, the performances of the two models were compared by calculating their mean of squared errors (MSEs) and the fit values that are given in Table 3. From the results of the calculated MSEs of the models, it was found that ARMAX model was better than ARX model for this process because its (ARMAX model's) mean of squared error was closer to zero than that of the developed ARX model.

Table 3. Performance values of the models

Model	MSE	Fit(%)
ARX	4.26E-05	88.31
ARMAX	9.31E-06	94.53

Apart from that, it was clear from the estimated fit values that the developed ARMAX model could account for 94.53% of the data used while ARX model could account for only 88.31%. Based on this, it has been seen that ARMAX model could account for more data of the process than ARX model could. This is another point that showed the superiority of ARMAX over ARX model for the olefin metathesis reactive distillation process.

CONCLUSION

The attainment of steady state by the output (bottom *cis*-2-hexene mole fraction) when the developed transfer function model of the olefin metathesis reactive distillation process was simulated showed that the process was a stable one. Also, the change that was noticed in the output when random input variable was passed into the system to generate data for ARX and ARMAX model was an indication that the chosen input variable was a valid one for the process. Furthermore, the developed ARX and ARMAX models were observed to have moderate orders, and the good comparison between the measured mole fraction and the simulated ones using the developed models revealed that they were able to represent the process well. However, the performance of the developed ARMAX model of the process was found to be better because its calculated MSE and fit values were estimated to be lower and higher respectively than those of the developed ARX model of the process.

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NOMENCLATURE

ΔG	Change in Gibbs free energy (kJ)
y_s	Simulated response
y_m	Measured response
$x_{cis\text{-}2\text{-hexene}}$	Mole fraction of *cis*-2-hexene obtained at the bottom section of the column
T_{dp}	Dead time of the main process model (min)
T_{dd}	Dead time of the disturbance process model (min)
T	Absolute temperature (K)
R_g	Universal gas constant (kJ/(kmol K))
RD	Reactive distillation
R	Reflux ratio
Q	Reboiler duty (kW)
MDL	Rissanen's Minimum Description Length
K_p	Static gain of the main process model
K_{eq}	Equilibrium constant
K_d	Static gain of the disturbance process model
ARX	AutoRegressive with eXogenous Inputs
ARMAX	AutoRegressive Moving Average with eXogenous Inputs
τ_p	Time constant of the main process model (min)
τ_d	Time constant of the disturbance process model (min)

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