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Fuzzy Hybrid Modeling of a Reactive Distillation Column for Ethyl Acetate Process

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ABSTRACT

The complex interactions between vapour-liquid equilibrium, mass and energy transports and reaction kinetics in reactive distillation (RD) process have constituted serious computational challenges to its modelling and control. This is because large numbers of nonlinear differential - algebraic equations are needed to represent the process when using first principle modelling approach. This work developed simplified model RD with the essential nonlinear characteristics relevant to control design purpose but with reduced computational demand. A Continuous RD flow sheet for ethyl acetate process was designed using a commercial simulator (ASPEN) to generate relevant dynamic data for model development. A hybrid Fuzzy Hammerstein (FH) model consisting nonlinear fuzzy model and linear state space model was then developed. The developed model was compared with linear autoregressive input exogenous (ARX) and nonlinear autoregressive input exogenous (NARX). The developed FH model has the order of [8, 8, 8, 8, 8] for dynamic linear part and 300 parameters consisting of 192 premise parameters and 108 consequent parameters for static nonlinear part. The output variables were temperature on trays 9 and 29 (T_9 and T_{29}) of the column. The values of root mean square error (RMSE) for the three models RMSE (ARX: 0.951; NARX: 0.75 and FH: 0.011) and RMSE (ARX: 0.961; NARX: 0.79 and FH: 0.015) obtained for T_9 and T_{29} respectively show that FH model outperforms the other two models. Correlation tests showed that FH model mostly described the dynamic nature of the process as it passed both whiteness and independent test criteria at $p < 0.01$.

Keywords: *Fuzzy system, Hybrid, Reactive distillation, Nonlinearity, Process dynamics*

1. INTRODUCTION

Reactive distillation (RD) belongs to a class of process intensification in chemical engineering in which chemical reaction and product separation are combined in a single column. This process offers the advantages in significant reduction on both capital and operating costs (Doherty and Buzad, 1992; Wang et al., 2003 and Talwalkar et al., 2007). The recent experimental works in which the advantages of reactive distillation are exploited are Zubair and Chin, 2010 and Araromi *et al*., 2011.

The increased interest in reactive distillation in the chemical process industries has led to a lot of research activities in this area in the last two decades resulting in the publications of many journal articles, many of which dealt with modelling of the process. Model, which gives adequate representation of the process, is needed for design and analysis in the area of control, optimization, fault detection and many real time operations. (Jafari *et al*.,2008). First principle knowledge often expressed in terms of material and energy balances and kinetics of the reaction system are used to model RD (Baur *et al*., 2003; Almeida-Rivera and Grievink, 2004). The dynamic model of reactive distillation obtaining from the first principle approach gives rise to large number of differential algebraic equations (DAE) and also there are complex interactions between vapour-liquid equilibrium, mass and energy transports and reaction kinetics. These have constituted serious computational challenges to its modelling and control due to large number of nonlinear differential - algebraic equations. Using the model in that form will be highly computational expensive especially for control design, online control and optimization purpose (Wittgens, 1999).

System identification which makes use of input output data generated through process excitation (Ali, 2011) can provide simplified approach to modelling complex system. Hybrid modelling which is also referred to as gray box modelling, semi-mechanistic modelling or polytopic modelling is a system identification approach in which first principle modelling and black-box modelling techniques are combined to describe highly nonlinear behaviour over a large operating domain with less use of computational resources (Van Lith et al., 2002). Some of the earlier works in this direction are Costa et al. (1999), who developed a hybrid neural model for bioprocesses and Pearson and Pottmann (2000), who focused on gray-box identification for three classes of block-oriented models (Hammerstein models, Wiener models, and the

feedback block-oriented models) for process with output multiplicities. Other works are Kahrs and Marquardt (2008); Chen et al., 2009 and Abonyi *et a*l. (2009).

In recent years, fuzzy modelling and identification have been used effectively in approximation of nonlinear process (Hosseinpourtehrani and Ghahraman, 2011). This is based on its capability to combine expert knowledge and measured data. When a model of a real process is created using only precise information, most often a lot of unsolvable difficulties due to the highly complex and nonlinear nature of the world will be encountered. Fuzzy modelling is a useful technique for the description of non-linear systems. In fuzzy modelling, non-linear process behaviour is approximated by multiple linear models with fuzzy transitions. In the modelling, fuzzy sets are employed to describe the continuous domains of input and output variables by dividing these domains into a small number of overlapping regions which constitute so-called linguistic values (for example High, Medium, Low) of the input and output variables (Betlem et al., 2005; Nithya et al., 2008).

The four basic components of fuzzy system are fuzzification, fuzzy rule base, fuzzy output engine (inference mechanism) and deffuzification (Tareghian and Kashefipour, 2007).The process of converting crisp values into membership functions is called fuzzification. Inference mechanism or inference engine is the computational method which calculates the degree to which each rule *fires* for a given fuzzified input pattern. Defuzzification converts the resulted fuzzy sets defined by the inference engine to the output of the model to a standard crisp signal. The two commonly used fuzzy models are Mamdani models and Takagi-Sugeno (TS) models (Tareghian and Kashefipour, 2007). They differ in their ability to represent information. While Mamdani models are based on collections of IF–THEN rules with vague predicates and fuzzy reasoning. Takagi-Sugeno (TS) models are formed by logical IF–THEN rules that have a simple linear relationship as consequent part.

The aim of this work is to develop simplified model for reactive distillation column with essential nonlinear characteristic that is suitable for real time application such as online nonlinear control design and online dynamic optimization.

2. STEADY-STATE DESIGN FOR REACTIVE DISTILLATION PROCESS

The process considered in this work is reaction between acetic acid and ethanol to produce ethyl acetate. Acetic acid reacts with ethanol in the presence of an acid catalyst to give ethyl acetate and water. This is an exothermic reversible reaction (Eq. (1)):

$$CH_3COOH + C_2H_5OH \xrightarrow{\text{sulpuric acid}} CH_3COOC_2H_5 + H_2O \quad (1)$$

Lai *et al*. (2008) presented rate expression for catalyzed reaction using Amberlyst 35 as given in Eq. (2)

$$R = m_{cat}\left(K_f[CH_3COOH][C_2H_5OH] - K_r[CH_3COOC_2H_5][H_2O]\right) \quad (2)$$

$$K_r = 1.24x10^9 \exp\left(\frac{-6105.6}{T}\right)$$

$$K_r = 1.34x10^8 \exp\left(\frac{-5692.1}{T}\right)$$

where, *R* is the reaction rate in mole/min, m_{cat} is the mass of catalyst in g, K_f is the forward reaction rate constant in $cm^6mol^{-1}g^{-1}s^1$and K_f is the backward reaction rate constant in $cm^6mol^{-1}g^{-1}s^1$ Ethyl acetate has the lowest boiling point (77.2 ^oC) of all the reacting species (Table 1) and this can be separated to its pure form in principle by using distillation. However, nonideal nature of the reacting mixture makes this impossible - the phase behaviour of the system gives three binary minimum-boiling azeotropes and one ternary minimum-boiling azeotrope.

Table 1: Boiling point of pure components and azeotropes

Pure components/azetropes	Boiling point (^oC)
CH_3COOH	117.90
H_2O	100.00
C_2H_5OH	78.31
$C_2H_5COOCH_3$	77.20

C_2H_5OH /H_2O	78.17
$C_2H_5COOCH_3$/ C_2H_5OH	71.81
$C_2H_5COOCH_3$/H2O	70.38
C_2H_5OH/$C_2H_5COOCH_3$/H_2O	70.23

Source: (Tang *et al.*, 2005**)**

The process flow diagram (PFD) for the optimal design shown in Fig. 1 was achieved through a rigorous optimizing procedure using ASPEN PLUS. The PFD has a heterogeneous catalytic RD column with two decanters and a stripping column. RD column base was included as part of reactive section as suggested by Araromi *et al*., 2008. The RD column has a diameter of 3 m and the weir for each tray is 0.1524 m high. The equilibrium model stage with bubble cap was used in the simulation. The holdup volume used in each tray is 1 m^3 (Hung *et al*.,

2006). Since reaction majorly takes place in bottom of the column, a much larger holdup is expected in the bottom of the RD column. Therefore, in this work 10 times tray holdup as used by Tang et al., 2005 for bottom of the RD was adopted. In the heterogeneous catalyst system, the kinetics is catalyst weight-based. Reaction kinetics was applied by assuming that 50% of the tray holdup volume would be occupied by the solid catalyst. The volume was converted into catalyst weight using a catalyst density of 770 kg/m^3 (Hung *et al.*, 2006)

In order to simulate waste water in Petrochemical and fine chemical industries (Radulescu *et al*., 2009), acetic acid feed of 30w/w% solution was used. Ethanol feed solution of 0.99 mole fraction was used (assay compositions). The approach presented by Huang *et al*. (2004) was adopted for optimal design of RD process in order to find feed tray locations, to determine the number of plates for given specifications and other steady state values needed for dynamic simulation. The values for steady state operation and associated cost obtained from the approach are presented in Table 2. The values in Table 2 were considered as the nominal situation, at which the dynamic data for model identification were generated.

3. IDENTIFICATION TESTS

The steady state parameters and design variables obtained from steady state design was used in identification test. Identification tests involved process excitation and data collection. Data generation was achieved by exporting the PFD in Fig. 1 from Aspen Plus environment to Aspen Dynamic environment. In the dynamic simulation, ethanol fresh feed and reboiler duty were used as input variables, and ±10 mol% acetic acid feed composition was used as disturbance. Tray temperatures and compositions are favourable candidate for controlled variables in RD process. However, due to slow in response and error associated with composition measurement, compositions are not usually used for controlled variables. Temperature measurements are certainly preferred because of their reliability and fast response (Hung *et al.*, 2006**)**. Measurement location was determined by carrying out sensitivity of the tray temperatures to the three inputs, namely, reboiler duty (Qr), fresh acetic acid feed (FAcOH) and acetic acid composition (disturbance) at the base-case reflux ratio. Data was generated using pseudo-random binary signal (PRBS) to vary reboiler heat duty and feed rate of ethanol while acetic acid concentration (disturbance variable) was varied using Gaussian noise signal. Pseudo-random binary signal (PRBS) signal generator was connected to the column to vary reboiler heat duty and feed rate of ethanol in order to obtain tray temperatures for all the stages. Disturbance was introduced by connecting the random signal generator to acetic acid stream in order to vary acetic acid composition.

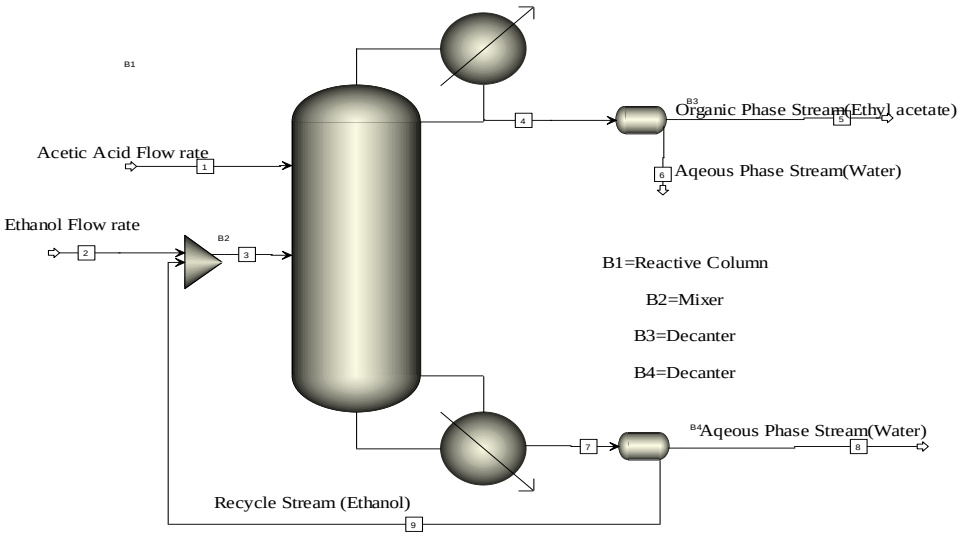


Fig. 1: Process flow diagram (PFD) for the optimal design

4. HYBRID MODEL DEVELOPMENT FOR RD PROCESS

The methodology for this work was based on a solution to Hammerstein structure called Fuzzy Hammerstein model. The structure has a nonlinear static gain function that feeds a linear dynamic function. Input vector **u** passes through the static gain function to obtain **f**(**u**) and then passes through the linear dynamic map to produce the output vector **y**. A discrete state space model was used to represent the linear dynamic element and a fuzzy model was used to describe the steady-state behaviour of the system. The structure (the rules) of the fuzzy model was generated based on linguistic and mechanistic knowledge about the steady-state behaviour of the system. Input-output data obtained from Aspen flowsheet of RD process was used to estimate the parameters of the rule consequences.

5. MODEL FORMULATION

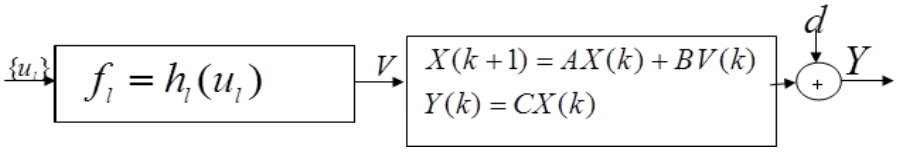
The Fuzzy Hammerstein (FH) model consists of the cascade connection of a static (zero-memory) nonlinearity and a linear time invariant (LTI) system (Fig. 2), where Y is the output vector with length n_y, $\{u_l\}$ is input vector for l =1,…,n_u with n_u equals to number of input, V represents the transformed input vector and d is the vector of additive disturbance variables. Fuzzy Takagi-Sugeno (TS) was adopted in this work because of its ability to give good approximation of most nonlinear system. Functions $f_l = h_l(u_l)$ are represented by zero-order Takagi-Sugeno (TS) fuzzy models formulated as a set of rules (Abonyi *et al.*, 2000) given as

$$Rule^{nr}_{i_1,\ldots,i_{nu}} : \text{if } u_1 \text{ is } A_1^{i_1}(u_1) \text{and} \cdots \text{and } u_{nu} \text{ is } A_{nu}^{i_{nu}}(u_{nu}) \text{ then } h_l = c_l^{nr} \quad (3)$$

$A_j^i(u_j)$ is the ij^{th} antecedent membership function of the fuzzy set for jth input of ith linguistic value and c_l^{nr} is the consequent parameter (crisp number). Membership functions (MFs) used in the characterization of each fuzzy set was Gaussian MFs. A Gaussian MF given in Eq. 3 is characterized by two parameters: c (centre) and σ_{ij} (width)

$$A_j^i(u_j) = \exp\left\{-\frac{1}{2}\left(\frac{u_j - c_{ij}}{\sigma_{ij}}\right)^2\right\} \quad (4)$$

Fuzzy subtractive clustering method (FSCM) was used for rule extraction. Given a set of input and output data, this technique assumes that each data point is a potential cluster centre and computes a measure of the potential of each data point and based on the density of surrounding data points this technique extracts a set of rules that models the data behaviour. An important parameter is the cluster radius for each input variable. It specifies a cluster range of influence in each of the data dimensions, assuming that the data fall within a unit hyperbox. This was achieved using Fuzzy logic tool box in MATLAB software.



Where

$f_l = h_l(u_l)$ is static nonlinearity function

u_l is input and $l = 1, \cdots, n_u$

n_u is number of input

V= input vector with length n_u

X is a vector representing state variables
A (n_y x n_y) is the process (plant) matrix,
B(n_y x n_u) is the input matrix,
C(1 x n_y) is the output matrix, and
D is the feed-through matrix.

d is disturbance variables.

The defuzzification strategy used to provide a means to choose a single output is center-average. A crisp output is chosen using the centers of each of the output membership functions and the maximum certainty of each of the conclusions represented with the implied fuzzy sets. The output of the fuzzy model h_l was obtained from Eq. 5

$$h_l = \frac{\sum_{i_l=1}^{N_R} \beta_{i_1}(u)\, c_{i_l}^l}{\sum_{i_l=1}^{N_{nu}} \beta_{i_l}(u)} \quad (5)$$

where $_{N_j}$ is the number of fuzzy sets in the j_{th} input domain. The weight $0 \le \beta_{i_1}(u) \le 1$ which is the overall truth value of the $_{i_1,\ldots i_{nu}}$ th rule was calculated using Eq. 6

$$\beta_{i_1}(u) = \prod_{k=1}^{n_u} A_k^{i_k}(u_k) A_{k,i_k}(u_k) \quad (6)$$

After obtaining the static nonlinear model, a multivariable linear dynamic ARX model was then constructed. The nonlinear ARX representation of the MIMO Hammerstein model is of the form:

$$\mathbf{y}(k+1) = \sum_{i=1}^{\mathbf{na}} \mathbf{A}_i y(k-i+1) + \sum_{i=1}^{\mathbf{nb}} \mathbf{B}_i \mathbf{f}(\mathbf{u}(k-i-\mathrm{n_d}+1)) \quad (7)$$

where

$$f_k(u(k-1-n_d-n_b)) = \sum_{j=1}^{N_R} \beta_j \left(u(k-1-n_d-n_b)\right) c_j^k \text{ (k=1...3)} \quad (8)$$

At steady state,

$$y(k+1) = y(k-i+1) = y_s \text{ and } \quad f(u(k-1-n_d-n_b)) = f_s \quad (9)$$

Therefore, the steady state behaviours of the Hammerstein model was obtained as

$$\mathbf{y_s = Gf(u_s)} \quad (10)$$

Where

$$G = \sum_{i=1}^{\mathbf{nb}} \mathbf{B}_i \left(I - \sum_{i=1}^{\mathbf{na}} \mathbf{A}_i \right)^{-1} \quad (11)$$

According to Pearson and Pottmann (2000), for singe-input, single-output SISO system, if G =1 then the static nonlinearity **f(.)** is simply the steady state curve for the Hammerstein model.

$$\mathbf{\Phi}_\mathrm{d} = \begin{bmatrix} \sum_{i=1}^{nb} b_{i,1,1} & \sum_{i=1}^{nb} b_{i,1,2} \\ \sum_{i=1}^{nb} b_{i,2,1} & \sum_{i=1}^{nb} b_{i,2,2} \end{bmatrix} \begin{bmatrix} \beta_1 & \beta_2 & \cdots & \beta_{N_R} \\ \beta_1 & \beta_2 & \cdots & \beta_{N_R} \end{bmatrix} \quad (14)$$

The aggression problem defined was solved using least square solution.

$y(k), \ldots, y(k-i+1)$ are the lagged outputs of the linear dynamic system

$u(k), \ldots, u(k-i-n_d+1)$ are the lagged inputs to the linear dynamic system

n_a denotes the maximum lags for the past outputs

n_b denotes the maximum lags for the past inputs,

n_d is the discrete-time delay

$A_1, \ldots, A_{na}$ are n_y x n_y matrices

$B_1, \ldots, B_{nb}$ are n_y x n_x matrices

where

Parameters Estimation Algorithm

Linear subspace parameters were first assumed, then the parameters of the nonlinear subspace was estimated by solving the regression problem formulated as

$$\mathbf{y}_d = \mathbf{\Phi}_d \mathbf{d} + \boldsymbol{\varepsilon} \quad (12)$$

where $\boldsymbol{\varepsilon}$ denotes the zero-mean, normally-distributed modeling error. For data pairs N, matrices $\mathbf{y}_d$ and $\mathbf{\Phi}_d$ are given by:

$$\mathbf{y}_d(k) = \mathbf{y}(k) - \sum_{i=1}^{n_a} \mathbf{A}_i^1 \mathbf{y}(k-1) \quad (13)$$

Estimation of parameters of the linear dynamic model was done by solving the following regression problem:

$$\mathbf{y}_l = \mathbf{\Phi}_l \boldsymbol{\theta}_l + \boldsymbol{\varepsilon} \tag{15}$$

$$\theta_l = \left[A_1, A_2, \cdots, A_{n_a}, B_1, B_2, \cdots B_{n_b} \right]^T \tag{16}$$

$$\Phi_l(k) = [y_1(k-1), \cdots, y_1(k-n_a), y_2(k-1), \cdots, y_2(k-n_a), \sum_{j=1}^{N_R} \beta_j d_j^1(u(k-1-n_d)) \cdots$$

$$\sum_{j=1}^{N_R} \beta_j d_j^1(u(k-n_b-n_d)), \sum_{j=1}^{N_R} \beta_j d_j^2(u(k-1-n_d)) \cdots \sum_{j=1}^{N_R} \beta_j d_j^2(u(k-n_b-n_d))] \tag{17}$$

Linear parameters were estimated by using constrained quadratic programming (QP) function. This was done by minimizing the function given in Eqns. 19 - 21

$$J(\theta) = \frac{1}{2}\theta_l^T H \theta_l + C^T \theta_l \tag{19}$$

with

$$H = 2\Phi_l^T \Phi_l \tag{20}$$

$$C = -2\Phi^T y_l \tag{21}$$

Parameters estimations were done by using a standard alternating optimization procedure proposed by Narendra and Gallman (Babuˇska, 1998).

6. RESULTS

When PFD transported to Aspen Dynamic environment was simulated, the following information was displayed on the message pane of the module:

- A total of 1290 equations (39.0%) were eliminated
- Simulation has 2840 variables, 2020 equations.

The input variables used for simulation are ethanol feed rate, reboiler heat duty and acetic acid concentration. All the stage temperatures generated were considered as possible candidates for output variables. Sensitivity of the stage temperatures to the three inputs, namely, reboiler heat duty (Qr), fresh acetic acid feed (FAcOH) and acetic acid composition was carried out using non-square relative gain (NRG) of Hung *et al.* (2006) to determine the correct output variables. Based on the largest row sum of NRG temperature of stage 9 (T_9) and temperature of stage 29 (T_{29}) were selected as output variables (Fig. 3). These two temperatures correspond to the locations either with large temperature breaks (Fig. 3) or having high sensitivity. T_9 is located in the rectifying section reactive column (RD) while T_{29} is located in the reactive section of RD. Out of 5393 data set collected, 2500 sample was used as estimation data set and 2893 was used as validation data set.

The order of linear model was obtained by using Akaike's final prediction error (FPE) criterion. Training data set was used to estimate the linear parameters and nonlinear parameters. Iteration was carried out to search for model orders that gave least FPE. The model orders found are [8, 8, 8, 8, 8] with the true FPE criteria as 0.04342 and 0.04694 for T_9 and T_{29} respectively. Static part of the FH model was identified by carrying out several simulations using different clustering radii values. The smaller the radium, the lager the number of rules extracted and the more accurate the model is in prediction. However, the larger the number of rules the more complex the model becomes and the more computational power is required to simulate the model. Therefore there is need to make compromise between accuracy and complexity of the model. The plot of RMSE for both training and checking at different radii is showing in Fig. 6. In order to avoid over prediction, RMSE for both training and checking must be close enough and this was obtained at *radium*=0.35 with corresponding 9 rules. The total number of fitting parameters of the static part of the FH model is 300 consisting of 192 premise parameters and 108 consequent parameters.

The first validation test carried out is model fit; this was done by simulating the identified model with validation data set. Linear autoregressive input exogenous (ARX) and non-Linear autoregressive input exogenous (NARX) model structures were identified with the help of identification toolbox in MATLAB using the same data set and they were compared with the proposed Model. This was done in order to have performance evaluation of the proposed model. The simulated or predicted model outputs for the three models are shown together with the validation data used in Fig. 7. The performance was also measured by RMSE. The RMSE obtained for T_9 were

0.951, 0.75 and 0.011 for ARX, NARX and FH models respectively and for T_{29} were 0.961, 0.79 and 0.015 for ARX, NARX and FH models.

Model fit validation is not enough to ascertain the quality of identified model for dynamic process, there is need know how much" left over" dynamic is unaccounted for during the identification exercise. Residual analysis is the validation tool used to account for this. Residual autocorrelation and cross correlations functions within the 99% confidence intervals are shown in Fig. 8 and 9 respectively.

7. DISCUSSION

The orders [8, 8, 8, 8, 8] obtained implies that past eight values of each input and past eight values of each output are needed to predict current outputs. The implication of this is that for 3 by 2 MIMO system considered in this work, each input and each output require eight parameters to formulate the dynamics part of the model. This is in addition to 300 parameters needed to formulate the static part of the model. This means that total number of 620 parameters is needed to characterize the process. The result obtained from ASPEN simulation which is based on first principle approach indicates that as much as 2020 nonlinear equations and with as much as 2840 variables are needed to model the process.

The ASPEN result is in line with observation made by Katariya *et al*. (2008) that modelling and simulation using first principle approach model is a computationally rigorous activity as it involves large number of highly non-linear equations like pressure drop correlations, packing holdup correlations, etc. The resulting models are generally of high order mostly not suitable and computationally very costly when used for on-line and real-time applications such as model-based control and model-based dynamic optimization (Balasubramhanya and Doyle III, 2000).

With this information, it can be said that the computational load of the identified model is less when compared with model obtained from first principle approach. The model can be more suitable for real time applications such as online controller setting and online optimization than using model obtained from first principle because less computational resources will be required to implement it. Besides, because of hybrid nature of the proposed model, it has better physical interpretation and generalization capability than model resulting from purely black-box modelling (Van Lith *et al., 2002;* Tian *et al*., 2004*)*. Also, the model can give allowance for trade-off between complexity and model performance in practical application (Libei *et al*., 2006).

From the model fit validation test carried out on identified models. It can be said from visual observation (Fig. 7) that NARX and the proposed Model (FH) give better fit when compared to ARX. The poor performance of ARX model can be attributed to the nonlinear nature of the process which linear model cannot handle. The model fit, which represents percentage of the output variations reproduced by the model for each model is shown in the figure legend. A high number means a better model. NARX has the best value (90.22%) followed by FH (84.04%) for stage temperature T_9. However the situation is not the same for stage temperature T_{29}, FH has highest value (81.16%) followed by NARX (74.43%). The impressive fit of the NARX model especially for T_9 comes as no big surprise as the unit of wavenet used as nonlinear estimator in its model structure was extra ordinary high (30 units). This will of course tell on computational load during simulation because the larger the unit the larger the numbers of parameters to represent the model. Expectedly, the worst fit is obtained for ARX (34.67%). On the average it can still be said that FH gives best fit. The RMSE results indicate that the proposed model is much more accurate than the other two modelsb.

Fig. 8 shows that only ARX model produces residuals outside the confidence interval. This implies that the model has worst performance of all the three models in predicting dynamic behavious of the process. The plots of residual autocorrelation functions for NARX and FH models fall within the confidence intervals. However, residual autocorrelation function for FH has greater fluctuation i.e. it is centered around zero (non-biased) while NARX residual function appears to display a systematic pattern. The implication of this is that FH model fits the data more correctly than NARX model because it residual approximates the random errors. Before conclusion can be made on the best model, there is need to carry out complementary analysis the cross-correlation of the residuals with the inputs. Fig.9 shows the plot of cross-correlation of the residuals with the inputs. For a model to be considered good model, residuals should be uncorrelated with past inputs. Any observation of correlation implies that the model does not describe how the output is formed from the corresponding input. Ideally for an acceptable model, the cross correlation function between input and residuals curves should lie within the confidence intervals. A peak outside the confidence interval for lag *k* indicates that the contribution to the output *y(t)* that results from the input *u(t-k)* is not properly described by the model. Looking at the Fig. 9, except for HF model which has all its cross correlation function within the confidence level, the other two models display some correlations at some lags. Thus the best model among the three estimated models is FH model, indicating that this model is the best of all the

three models in predicting the future values of output, given the past input and output measurements.

8. CONCLUSION

This work has considered identification of hybrid Fuzzy Hammerstein (FH) model to effectively characterize nonlinear behaviours of reactive distillation process. The model consists of nonlinear fuzzy part and linear state space part. A continuous RD flowsheet for ethyl acetate process was designed using a commercial simulator (ASPEN) to generate relevant dynamic data for model identification. Parameters estimations were done by using a standard alternating optimization procedure proposed by Narendra and Gallman (Babuska, 1998). The results have shown that FH model can provisionally capture the nonlinear dynamic behaviours of RD system and the model can be found suitable for real time applications.

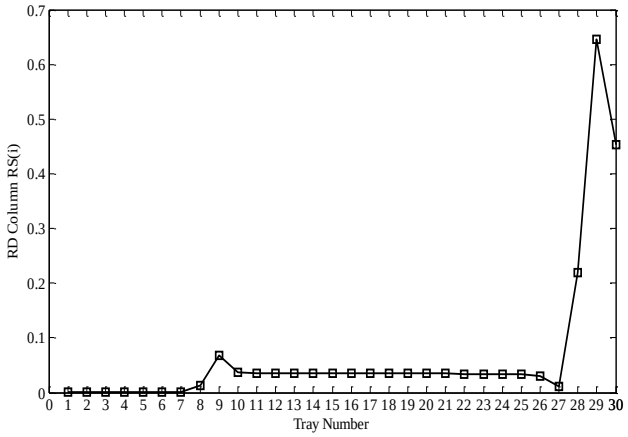


Fig. 3 : NRG and selected temperature control trays

Table 2: Nominal operating conditions and the column parameters for ethyl acetate Reactive Distillation

Parameters	Values
Total no. of trays (*NT*)	28
Stripping(*Ns*)/reactive(*Nrxn*)/rectifying (*NR*)	8/12/8
Acetic acid solution feed tray (NF1)	11
*Etha*nol solution feed tray (NF2)	15
Flow rate of acetic acid solution (Kmol/hr)	30
Composition of acetic acid	30 g/g % (0.114 mol fraction)
Flow rate of ethanol solution (Kmol/hr)	10.5
Composition of ethanol	0.5 mol fraction
Heat duty Condenser (MJ/h)	-4118.271
Reboiler (MJ/h)	4000
Tray Column diameter (m)	1.219
Reflux rate (Kmol/hr)	100.386
Reflux drum hold up (Kmol)	0.050
Reboiler hold up (Kmol)	0.010

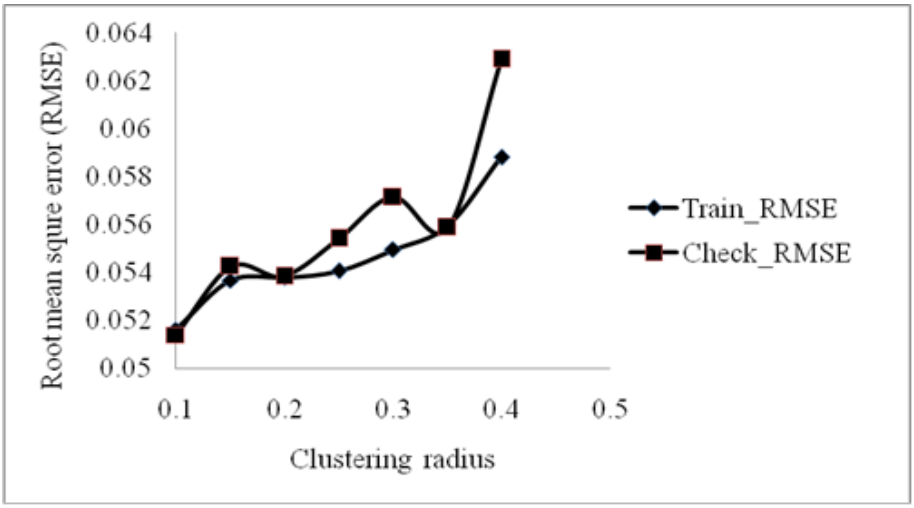


Fig. 6: Plot of clustering radius versus root mean squre error (RMSE)

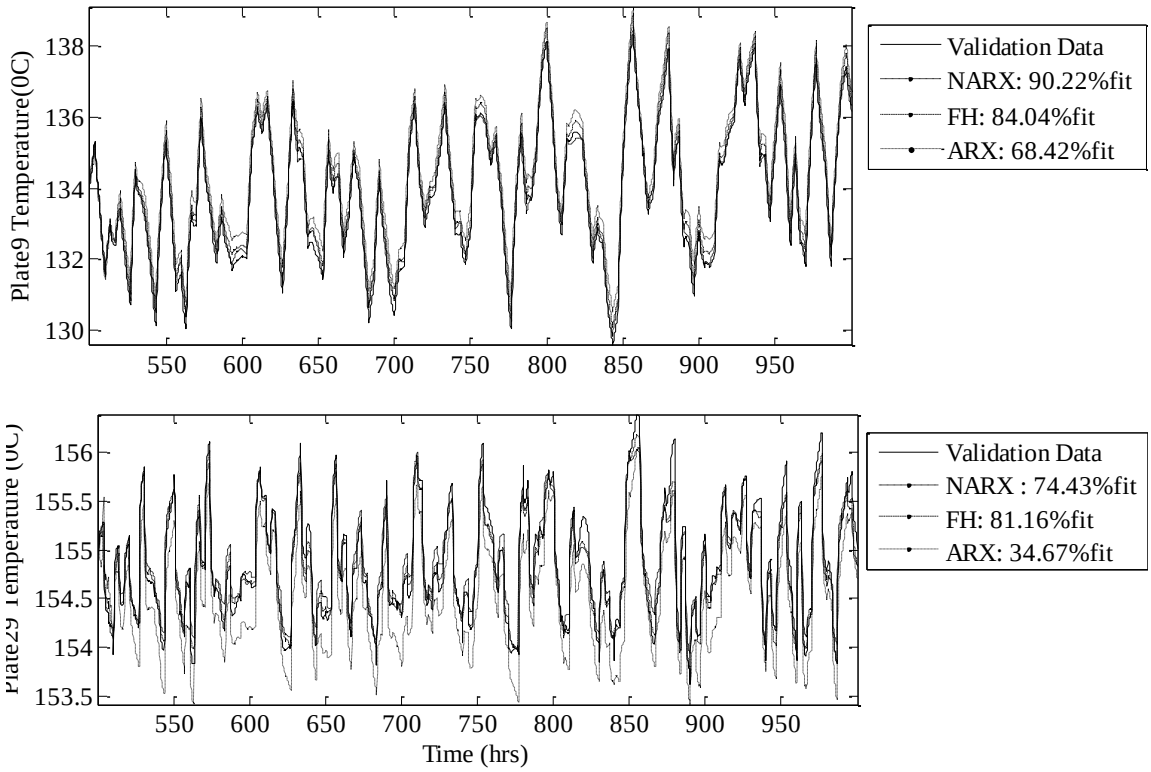


Figure 7: Model fit plots for the identified models

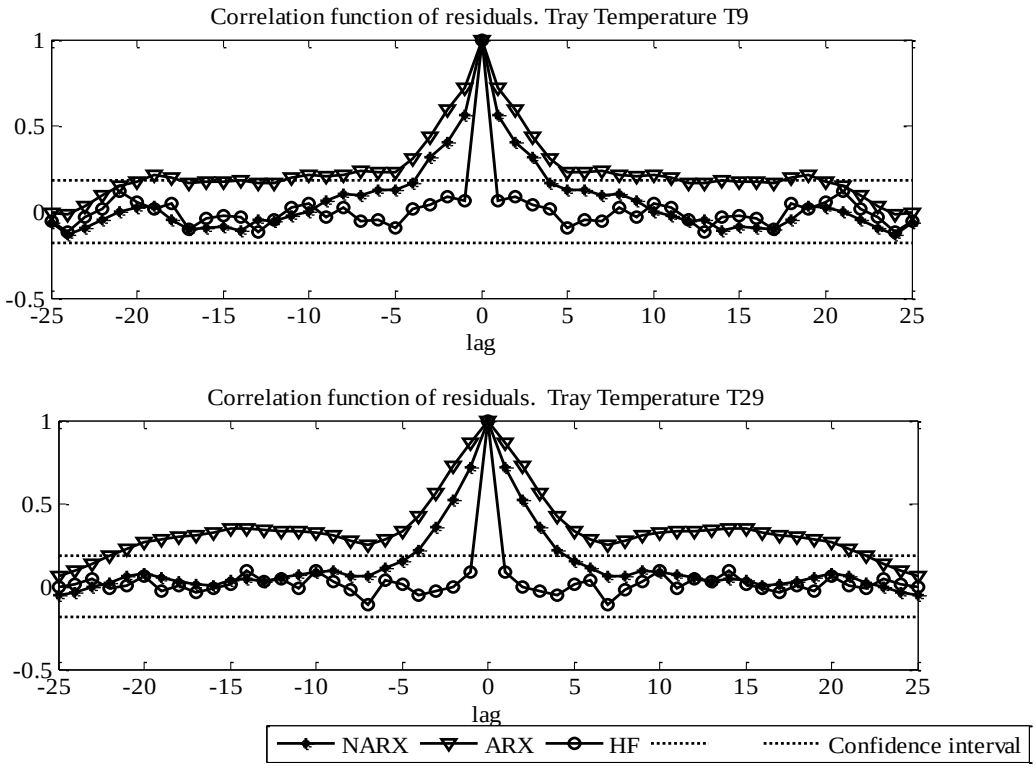


Figure 8: Plots of auto correlation function of residuals

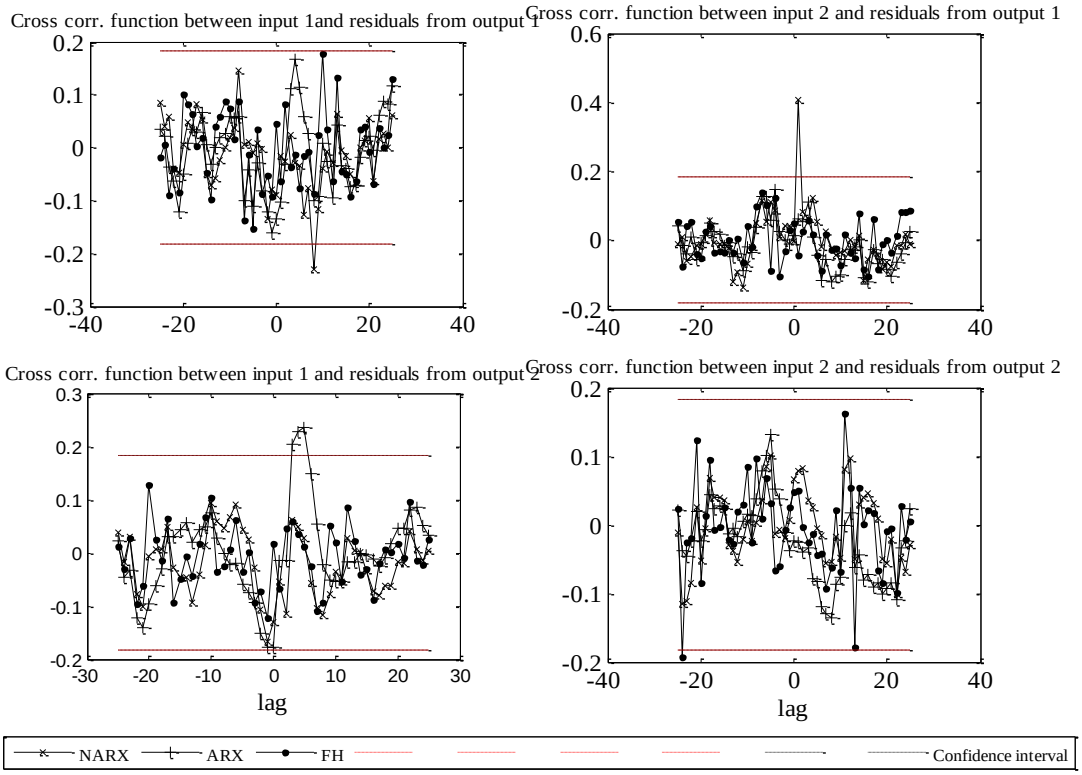


Figure 9: Plots of cross correlation function of residuals

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