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Reactive Batch Distillation

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Reactive batch distillation is a renowned unit operation that is used in industries. This process is an attractive process intensification scheme involving combined operations of reaction and separation in a single unit.

Advantages

1. Reducing capital cost
2. Reducing operational const: based on integrating producing and consuming energy
3. Different feeds and side streams may have

Disadvantages

Reaction and distillation should take place at the same temperature and pressure

Mathematical modelling

For the reboiler, total and species mass balance and energy balance are as follows (subscript *B*):

$$\frac{dM_B}{dt} = L_1 - V_B + M_B \Delta R_B$$

$$\frac{d(M_B x_{B,j})}{dt} = L_1 x_{1,j} - V_B y_{B,j} + M_B R_{B,j} \qquad j = 1, \ldots, Nc$$

$$\frac{d(M_B H_{L,B})}{dt} = L_1 H_{L,1} - V_B H_{V,B} + Q_B$$

First plate (subscript 1):

$$\frac{dM_1}{dt} = V_B + L_2 - V_1 - L_1 + M_1 \Delta R_1$$

$$\frac{d(M_1 x_{1,j})}{dt} = V_B y_{B,j} + L_2 x_{2,j} - V_1 y_{1,j} - L_1 x_{1,j} + M_1 R_{1,j} \qquad j = 1, \dots, Nc$$

$$\frac{d(M_1 H_{L,1})}{dt} = V_B H_{V,B} - V_1 H_{V,B} + L_2 H_{L,2} - L_1 H_{L,1}$$

Intermediate plates (subscript *i*):

$$\frac{dM_i}{dt} = V_{i-1} + L_{i+1} - V_i - L_i + M_i \Delta R_i$$

$$\frac{d(M_i x_{i,j})}{dt} = V_{i-1} y_{i-1,j} - V_i y_{i,j} + L_{i+1} x_{i+1,j} - L_i x_{i,j} + M_i R_{i,j}$$

$$\frac{d(M_i H_{L,i})}{dt} = V_{i-1} H_{V,i-1} - V_i H_{V,i} + L_{i+1} H_{L,i+1} - L_i H_{L,i}$$

Top plate (subscript *Nt*):

$$\frac{dM_{Nt}}{dt} = V_{Nt-1} + L_0 - V_{Nt} - L_{Nt} + M_i \Delta R_{Nt}$$

$$\frac{d(M_{Nt} x_{Nt,j})}{dt} = V_{Nt-1} y_{Nt-1,j} + L_0 x_{D,j} - L_{Nt} x_{Nt,j} - V_{Nt} y_{Nt,j} + M_{Nt} R_{Nt,j}$$

$$j = 1, \ldots, Nc$$

$$\frac{d(M_{Nt} H_{L,Nt})}{dt} = V_{Nt-1} H_{V,Nt-1} - V_{Nt} H_{V,Nt} + L_0 H_{L,D} - L_{Nt} H_{L,Nt}$$

Condenser (subscript *D*):

$$\frac{dM_D}{dt} = V_{Nt} - L_0 - D + M_D \Delta R_D$$

$$\frac{d(M_D x_{D,j})}{dt} = V_{Nt} y_{Nt,j} - D x_{D,j} - L_0 x_{D,j} + M_D R_{D,j}$$

The holdup of the trays are calculated based on Luyben (1990):

$$LV = L \times Mw_{Ave} / Density_{Ave}$$

$$HFOW = \left(\frac{LV}{999 \times WLS} \right)^{0.6667} \qquad MV = \left(HFOW + \frac{WHS}{12} \right) \times \frac{\pi d^2}{4 \times 144}$$

$$M = MV \times Density_{Ave} / Mw_{Ave}$$

The reboiler holdup at any time is calculated from an algebraic combination of the initial charge, the material in the column, and the total material removed up to that time:

$$M_B = M_{B0} + \sum_{i=1}^{Nt} M_{i0} - \sum_{i=1}^{Nt} M_i - \int D\,dt$$

Dynamic simulation of the reactive batch distillation column is performed by solving the system of Differential and Algebraic Equations (DAE) simultaneously using integration the Euler method.

For these purposes, the process is modeled using a tray-by-tray description resulting in 88 differential equations.

With this setup, the column operated at total reflux conditions for approximately 4.5 hours, following that distillate products removed .

Expriment

In order to design and implement the proposed state estimation scheme, a reactive batch distillation with an esterification reaction is considered. The reversible reaction is given by

$$CH_3COOH \quad + \quad C_2H_5OH \quad \Leftrightarrow \quad CH_3COOC_2H_5 \quad + \quad H_2O$$

Acetic Acid(1) Ethanol(2) Ethyl Acetate (3) Water (4)

The reaction mixture involves three azeotrops,

1. ethyl acetate–ethanol,
2. acetic acid–water and
3. ethyl acetate–acetic acid–water.

Reactive batch distillation can overcome azeotropes by changing or get rid of the conditions for azeotrope formation through the combined effect of reaction and separation.

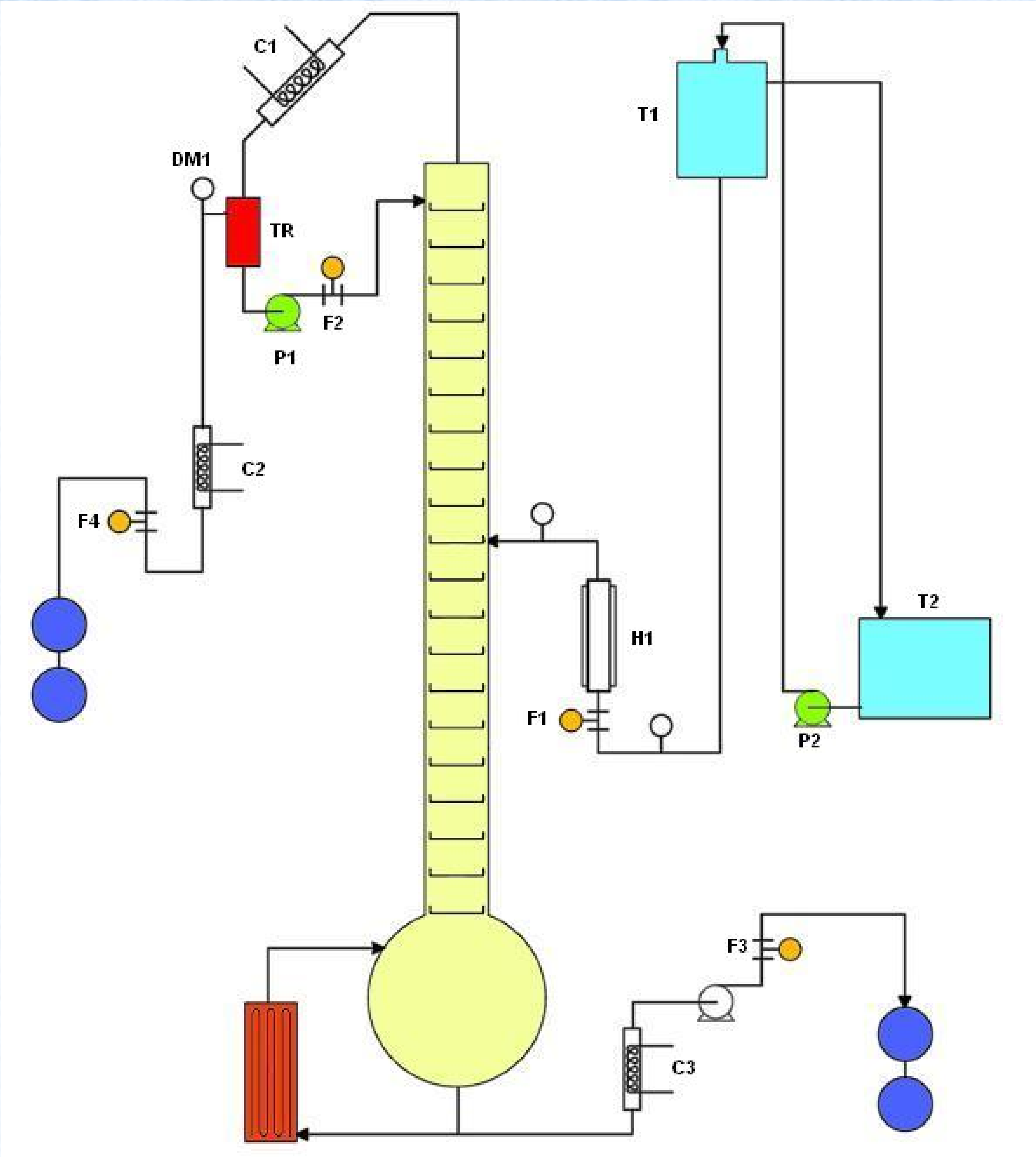
Because of the lowest boiling point of ethyl acetate, it is the main product in the mixture.

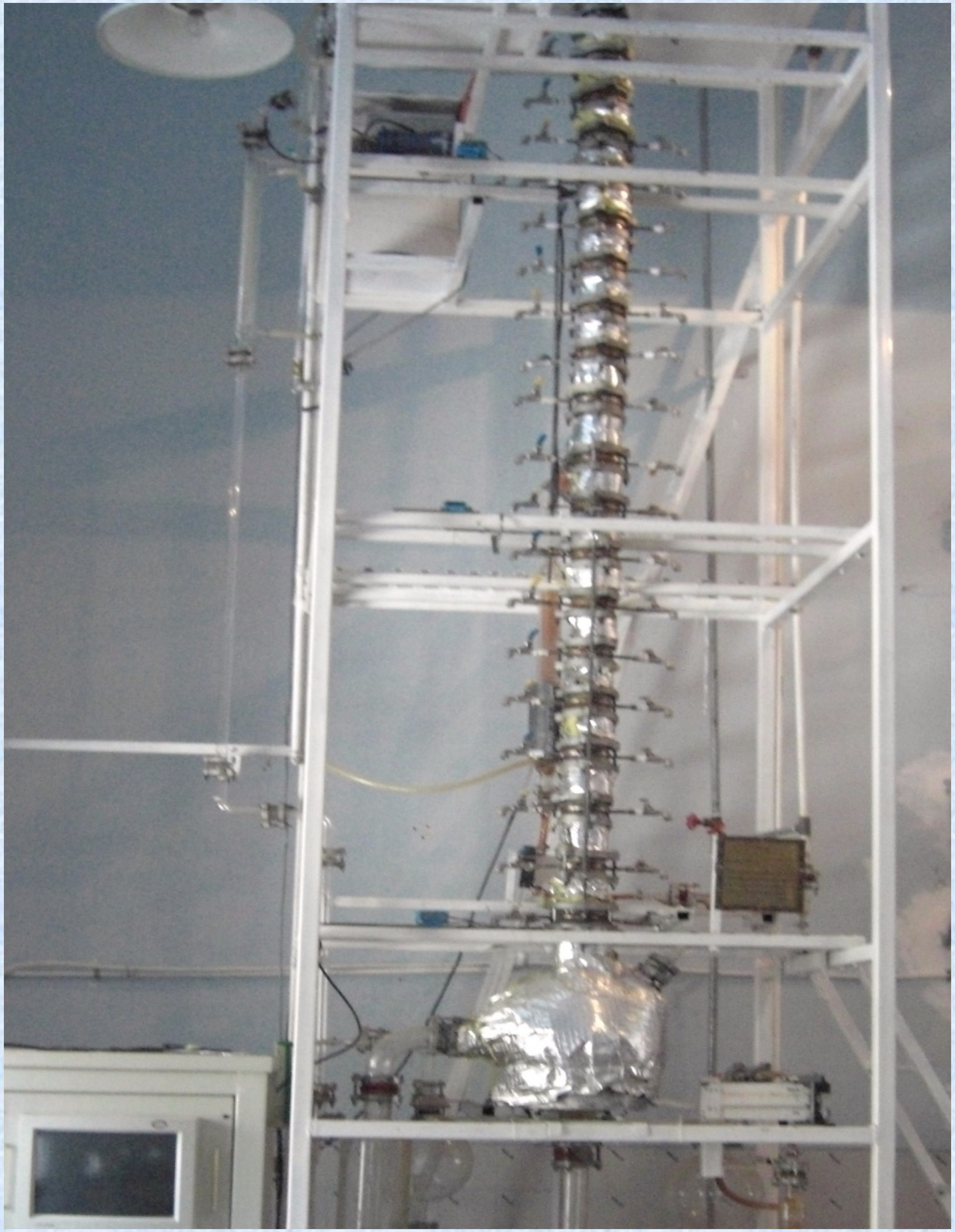
The chemical equilibrium has been being shifted to the right and improves conversion of the reactants by controlling the ethyl acetate removal.

Characteristics and operation conditions of the reactive batch distillation

No. of plates	19
Initial charge (kmol)	0.5
Initial mole fraction	
CH_3COOH (1)	0.35
C_2H_5OH (2)	0.45
$CH_3COOC_2H_5$(3)	0
H_2O (4)	0.2

Condenser Pressure (psi)	14.7
Column diameter (m)	0.08
WHS (m)	0.013
WLS (m)	0.0236
Q_B (kJ/hr)	3587.2
Kinetic data	
$r = k_1 C_1 C_2 - k_2 C_3 C_4$	
k_1 (gmol^{-1}.min^{-1})	4.76×10^{-4}
k_2 (gmol^{-1}.min^{-1})	1.63×10^{-4}


C1
T1
DM1
TR
F2
P1
C2
F4
H1
T2
F1
P2
F3
C3

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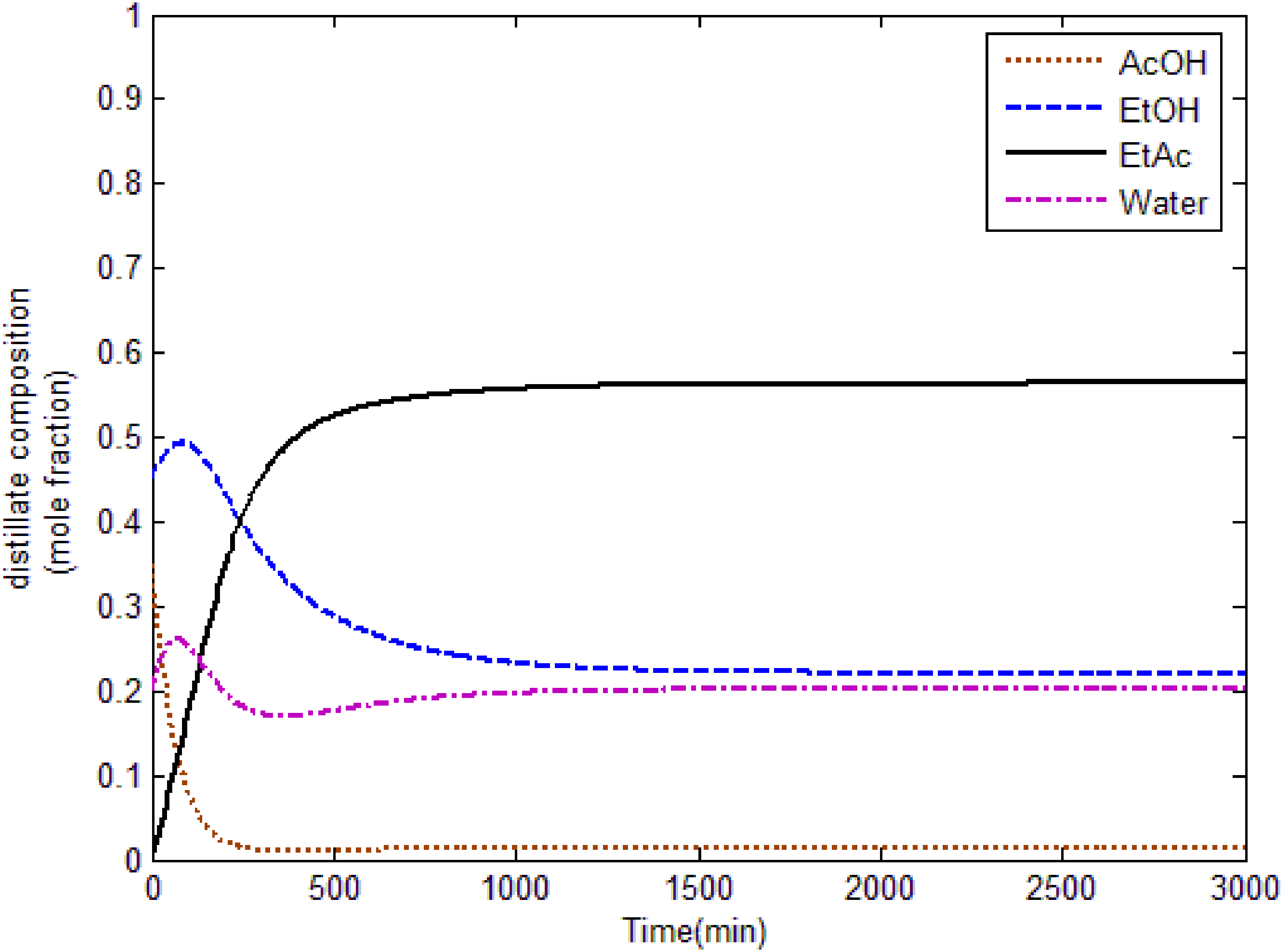


Fig. 2. Condenser profile for total reflux

Fig. 2 shows the composition profile of the condenser with respect to time for total reflux. This figure indicates that compositions reach to a steady state condition with total reflux after 900 minutes.

The condenser composition for reflux ratio equal to 4 is plotted in Fig. 3.

Since EtAc is the lightest distillate component, and its withdrawal from the column will cause the chemical equilibrium shift to the right hand, then its variation shows a maximum through out the operation.

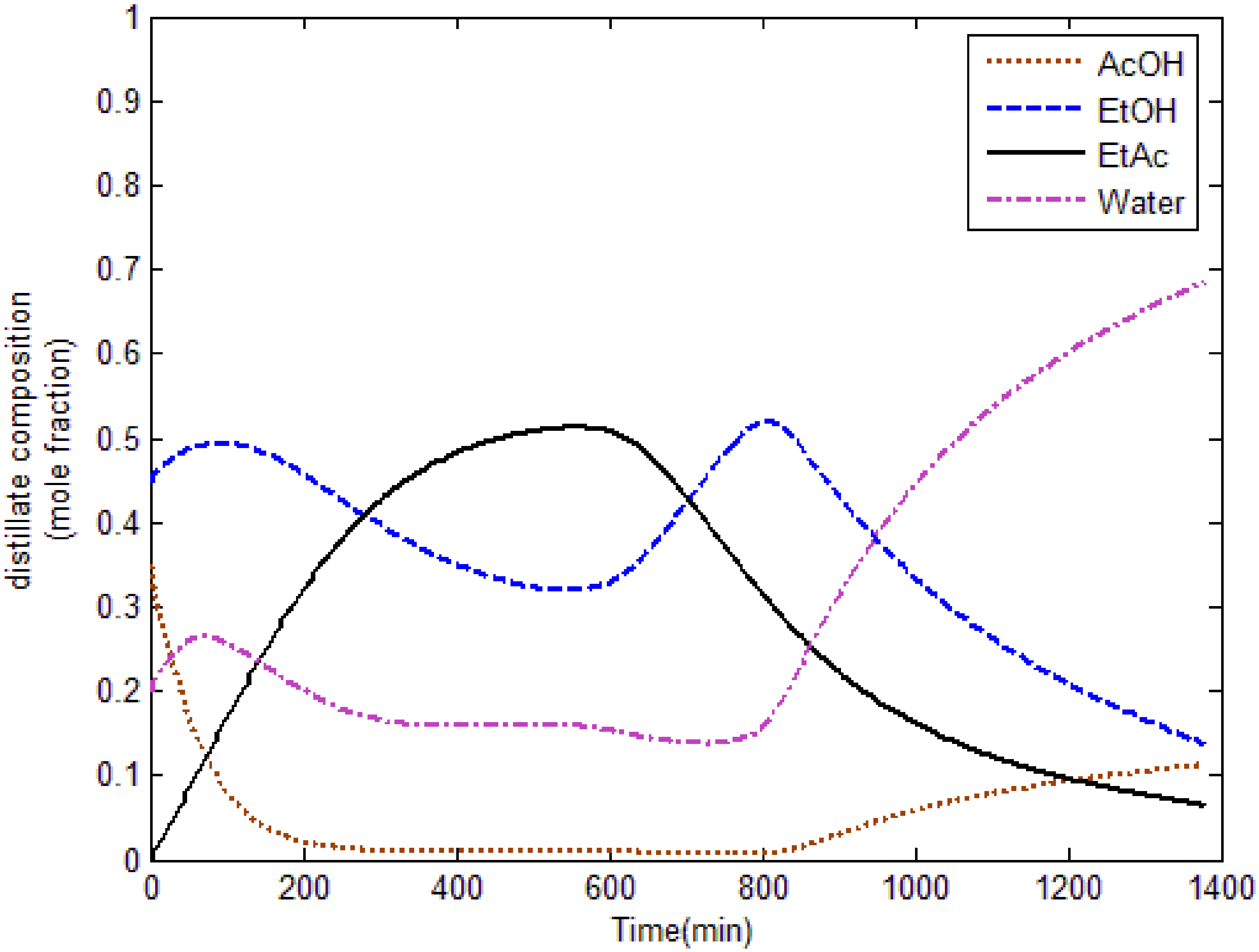


Fig. 3. Condenser profile for reflux ratio of 4 maintained throughout the batch.

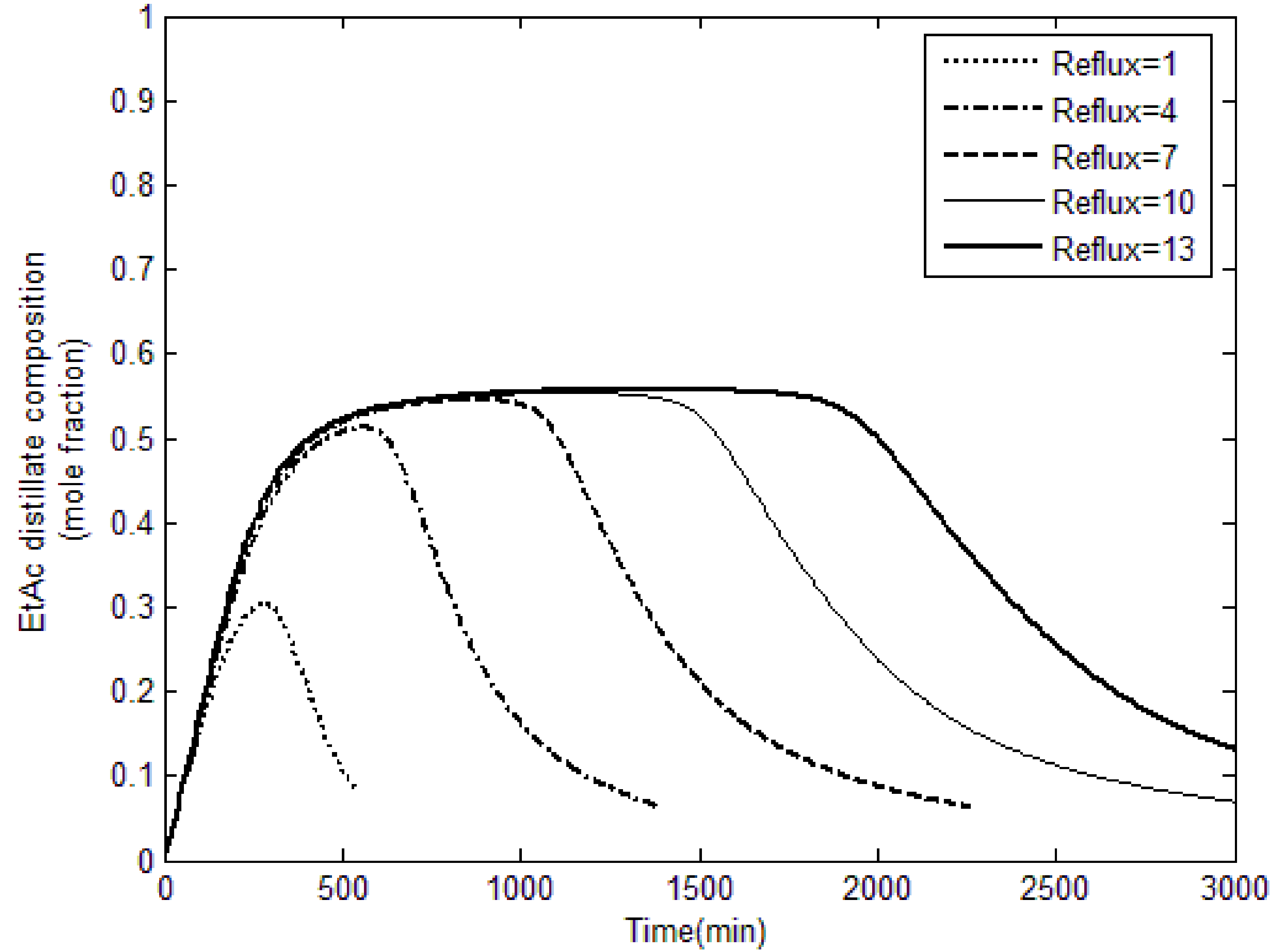


Fig. 4. EtAc mole fraction in the condenser for different reflux ratios with Q_B =3587.2 kJ/hr

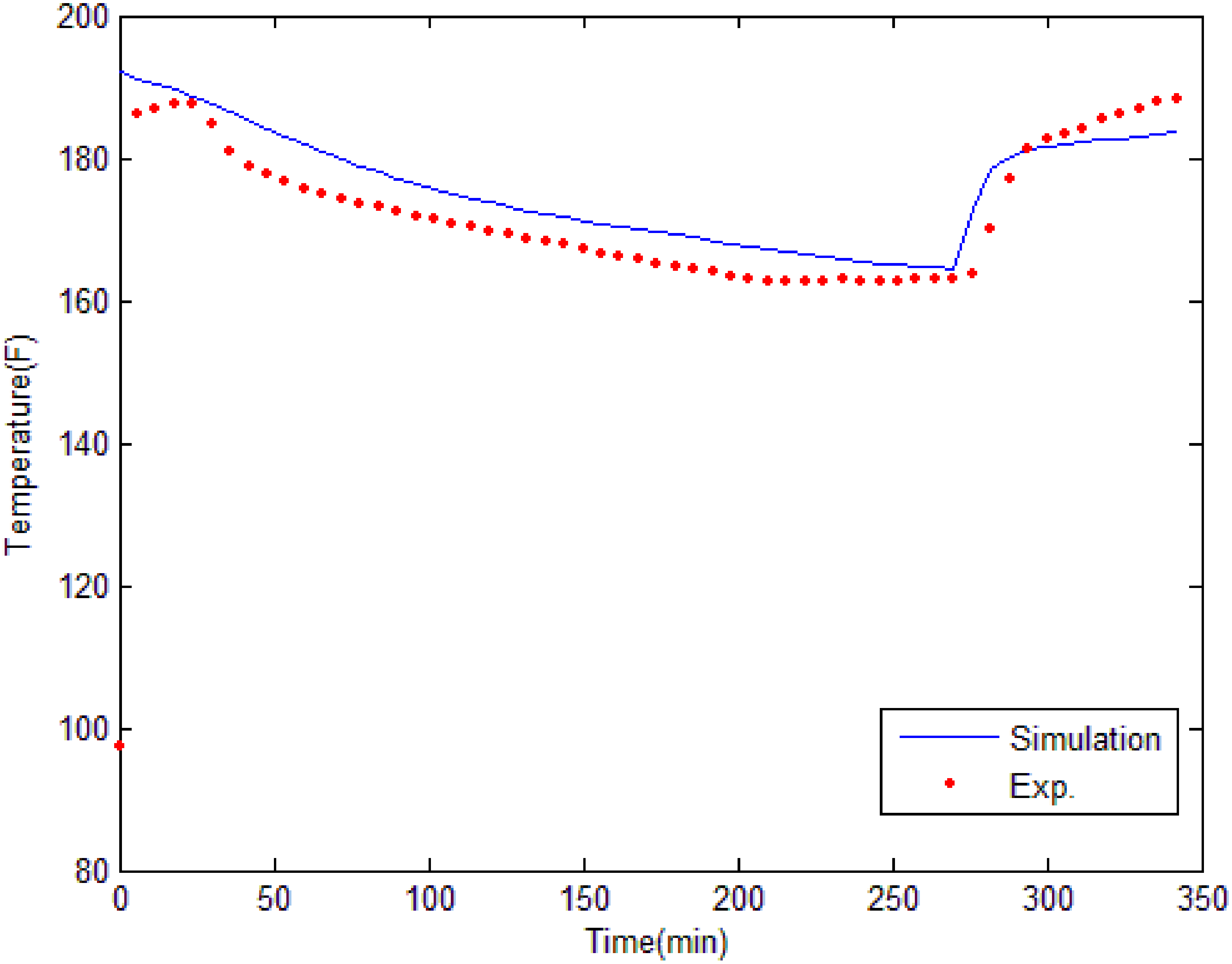


Fig. 5. Temperature profile of tray number3

Temperature profile is shown that for 250 minutes of the operation. Temperature is reducing because of the total reflux operation and produceing ethyl acetate that is the lightest component.

In the remaining 100 minutes the reflux ratio is equal to zero and all distilled products are left from the column and it causes the heavy components to come up through the column and makes trays warmer.

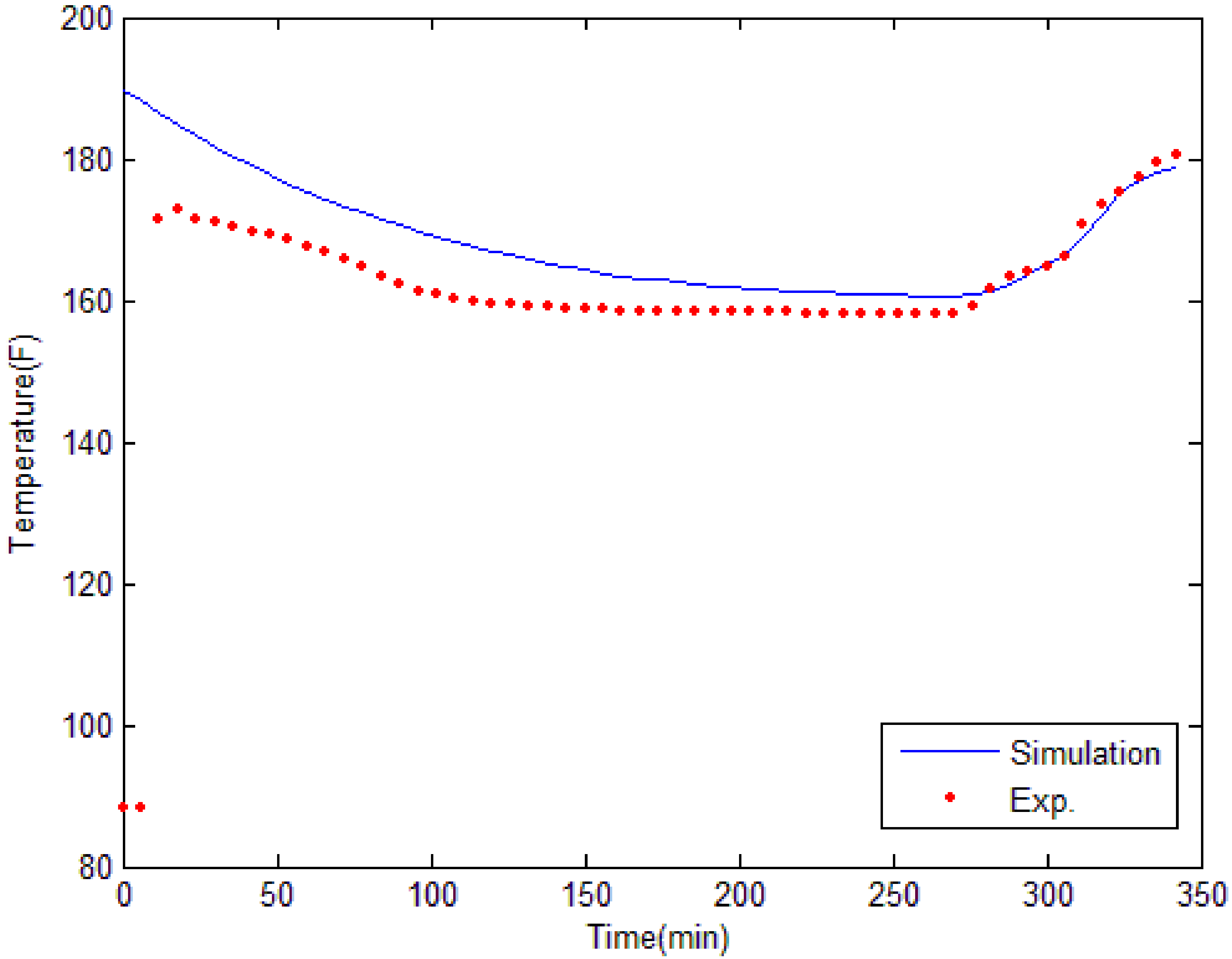


Fig. 6. Temperature profile of tray number 18

State estimation algorithm

Composition control of batch or continuous reactive distillation processes is very important. Especially, in order to meet the purity specifications, a batch column has to be operated as precisely as possible. If the current compositions are known, they can form a basis to improve the process performance through an operator decision making for the development of a closed loop control scheme.

Measurements of the compositions can be done using direct composition analyzers. However, composition measurement is not feasible, since, these analyzers, like gas chromatographs, involve large measurement delays as well as high investment and maintenance costs.

Oisiovici and Cruz (2000, 2001) have been developed a stochastic estimator for batch distillation columns, a discrete extended Kalman filter (EKF) for binary and multicomponent systems and tested them.

Venkateswarlu and Avantika (2001) used various methods based on Extended Kalman Filter (EKF), adaptive fading Kalman filter and steady state Kalman filter for inferential estimation of compositions from temperature measurements in multiple fraction multicomponent batch distillation.

An Artificial Neural Network (ANN) estimator to predict the composition values of a reactive batch distillation system inferentially is designed by Bahar and Özgen (2008).

Fernandez de Canete et al. (2008) have been described the application of an adaptive network based fuzzy inference system (ANFIS) predictor to estimate the product compositions in a binary methanol-water continuous distillation column from available on-line temperature measurements.

Number and Location of measurements

Yu *et al.* (1987) employed a degree-of-freedom concept to analysis distillation column observability and exhibited, if the number of measurements be at least (*NC* - 1), distillation column will be observable.

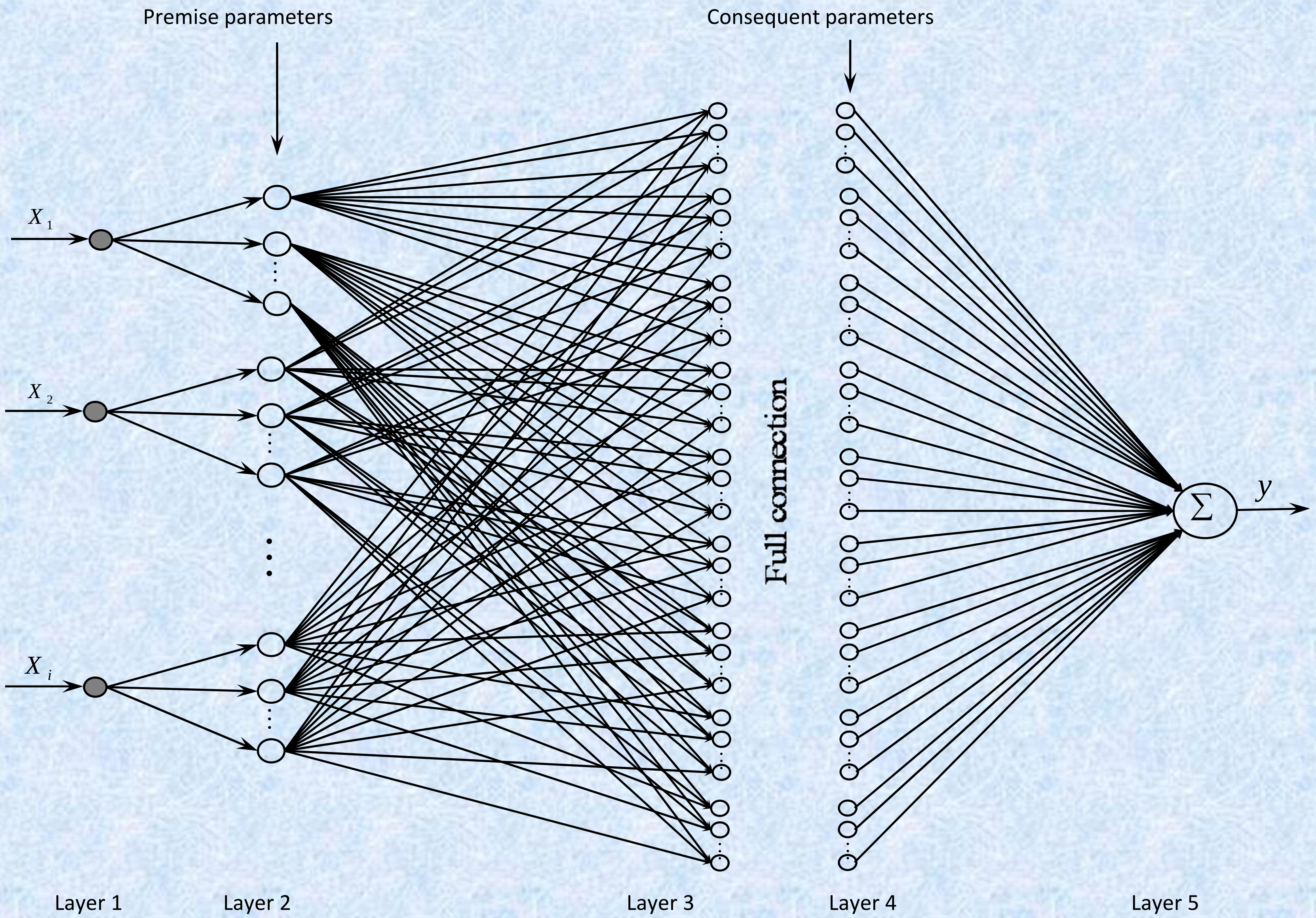
Other investigator (e. i, Quintero- Marmol *et al.* (1991) and Yildiz et al. (2005)) show the same results for multi-component batch distillation column, by using of an Extended Luenberger Observer and Extended Kalman Filter, respectively. They showed that at least (*NC*) thermocouples are needed for effective estimation.

The temperature measurement locations should be spread throughout the column homogeneously including the reboiler and the top tray (Yildiz et al. 2005). The reboiler and the top tray are the most sensitive temperature measurement locations for a multi-component batch distillation column (Venkateswarlu and Kumar 2006).

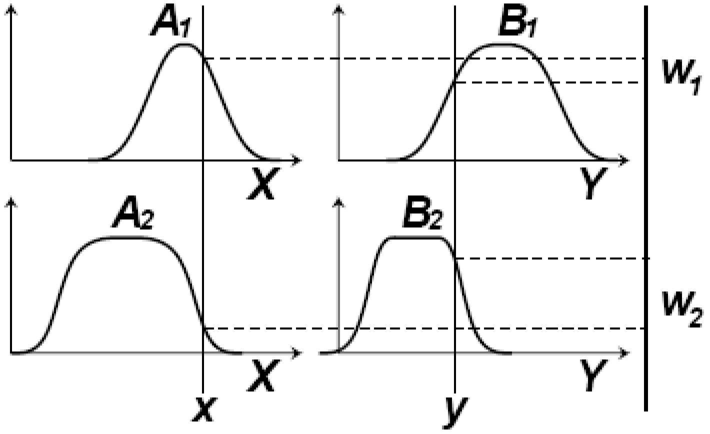
ANFIS-based state estimation

The basic idea behind the neuro-adaptive learning techniques is very simple. These techniques provide a method for the fuzzy modeling procedure to learn information about data set, in order to compute the membership function parameters that best allow the associated fuzzy inference system to track the given input-output data.

ANFIS constructs an input-output mapping based on both human knowledge (in the form of fuzzy if-then rules) and simulated input-output data pairs. It serves as a basis for building the set of fuzzy if-then rules with appropriate membership functions to generate the input-output pairs. We will describe primarily architecture and this learning algorithm for the Sugeno fuzzy model, with an application example of state estimation for reactive batch distillation.



Structure of ANFIS used for state estimation

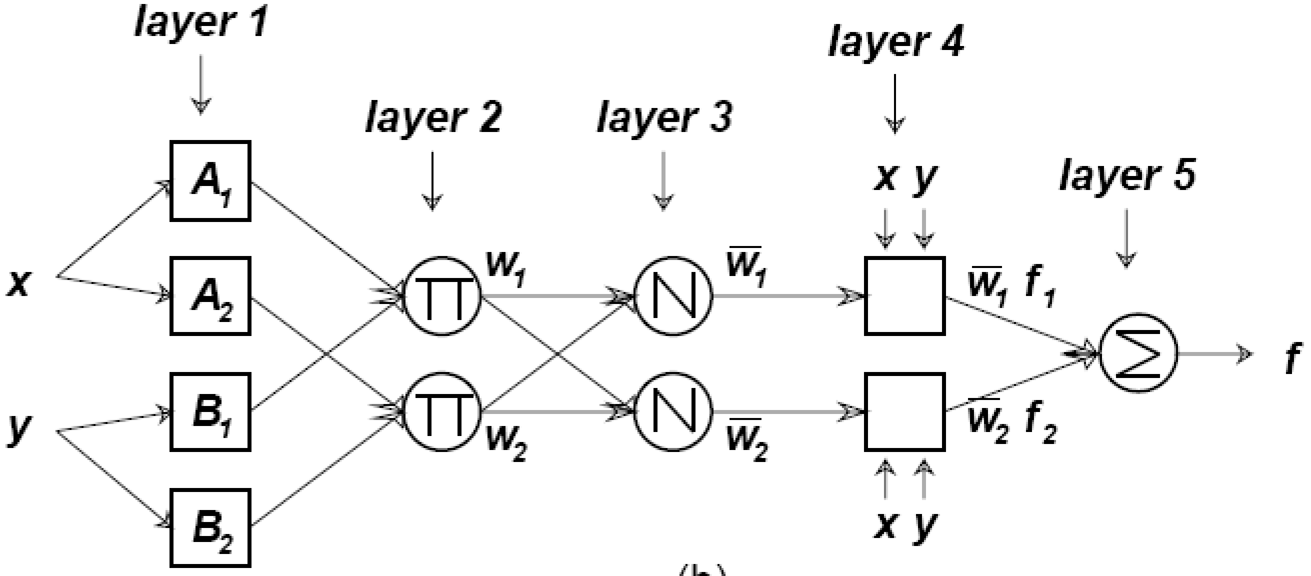


$$f_1 = p_1 x + q_1 y + r_1$$

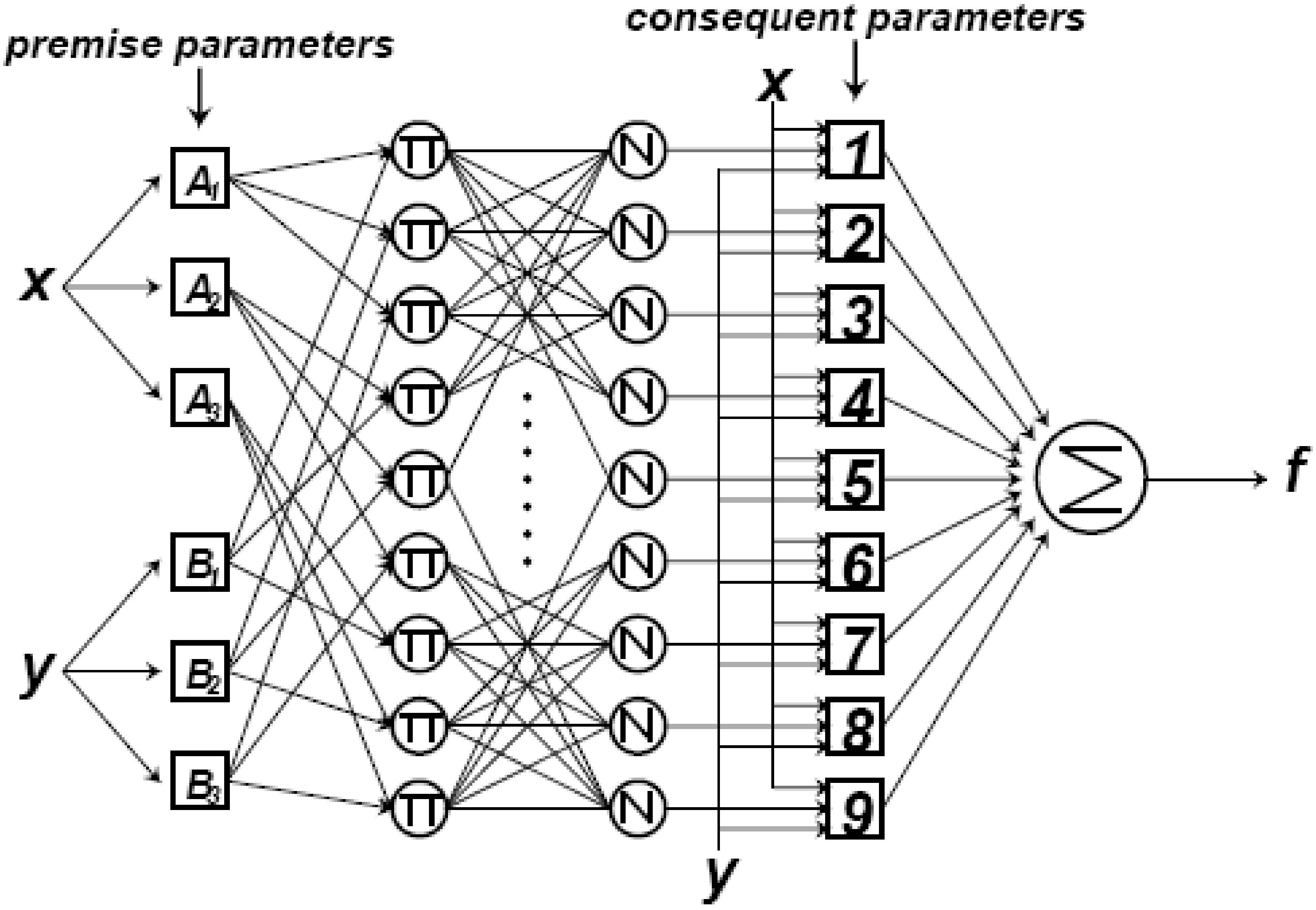
$$f_2 = p_2 x + q_2 y + r_2$$

$$f = \frac{w_1 f_1 + w_2 f_2}{w_1 + w_2} = \overline{w}_1 f_1 + \overline{w}_2 f_2$$

(a)



(b)


premise parameters
consequent parameters
x
y
A_1
A_2
A_3
B_1
B_2
B_3
Π
N
x
y
1
2
3
4
5
6
7
8
9
Σ
f

ANFIS architecture:

In this study, the artificial neural network and fuzzy logic are combined to establish a fuzzy neural network. The linear fuzzy rule in a Takagi and Sugeno's (TS) fuzzy model with 5 inputs has the form:

$$R^m : \ If \quad X_1 \ is \ A_{1j}, \quad X_2 \ is \ A_{2j}, \quad X_3 \ is \quad A_{3j}, \ X_4 \ is \ A_{4j}, \quad X_5 \ is \ A_{5j}$$

$$Then \quad y^m = \left(\sum_{i=1}^{5} r_i^m X_i \right) + r_0^m \qquad i = 1,2,\ldots,5; \quad j = 1,2,\ldots,n; \quad m = n^5$$

The ANFIS, in essence, integrates the basic elements and functions of a conventional fuzzy inference system (membership functions, fuzzy logic rules, fuzzification, defuzzification, and fuzzy implication) into a connectionist structure that has distributed learning ability to learn the membership functions and fuzzy logic rules. Each input has n membership function, and nodes of the same layer have similar functions as described below.

Layer 1: input layer

The input units in this layer are T_1, T_i, T_{20}, time and reflux ratio, and the output nodes just transmit these input values to the next layer.

Layer 2: linguistic term layer

This layer receives the signals from the input layer and uses the Gaussian 2 membership function to determine the relative contribution of the observed signals.

Layer 3: generate the firing strength

Layer 3 implements the links relating preconditions to consequences. Each output node represents the firing strength of a rule. The connection criterion is that each rule node has only one antecedent link from a linguistic variable.

Layer 4: consequent layer

Each node in this layer is an adaptive node with a node function

$$O_m^{(4)} = O_m^{(3)} f_m = \frac{w_m}{\sum_{m=1}^{n^5} w_m}\left[\left(\sum_{i=1}^{5} r_i^m X_i\right) + r_o^m\right], \quad i = 1,2,\ldots,5; \quad m = n^5$$

Parameters in this layer will be referred as consequent parameters.

Layer 5: output layer

The single node in this layer is a fixed node labeled Σ which computes the overall output as the summation of all incoming signals:

$$O^{(5)} = y = \sum O_m^{(4)}, \qquad m = n^5$$

four parallel ANFIS estimators are designed and combined to estimate the top product compositions from tray temperatures. Also, as it is stated in section 4 temperatures of three trays are utilized for estimation.

After training of the ANFIS structures, performances of estimators are investigated through the model.

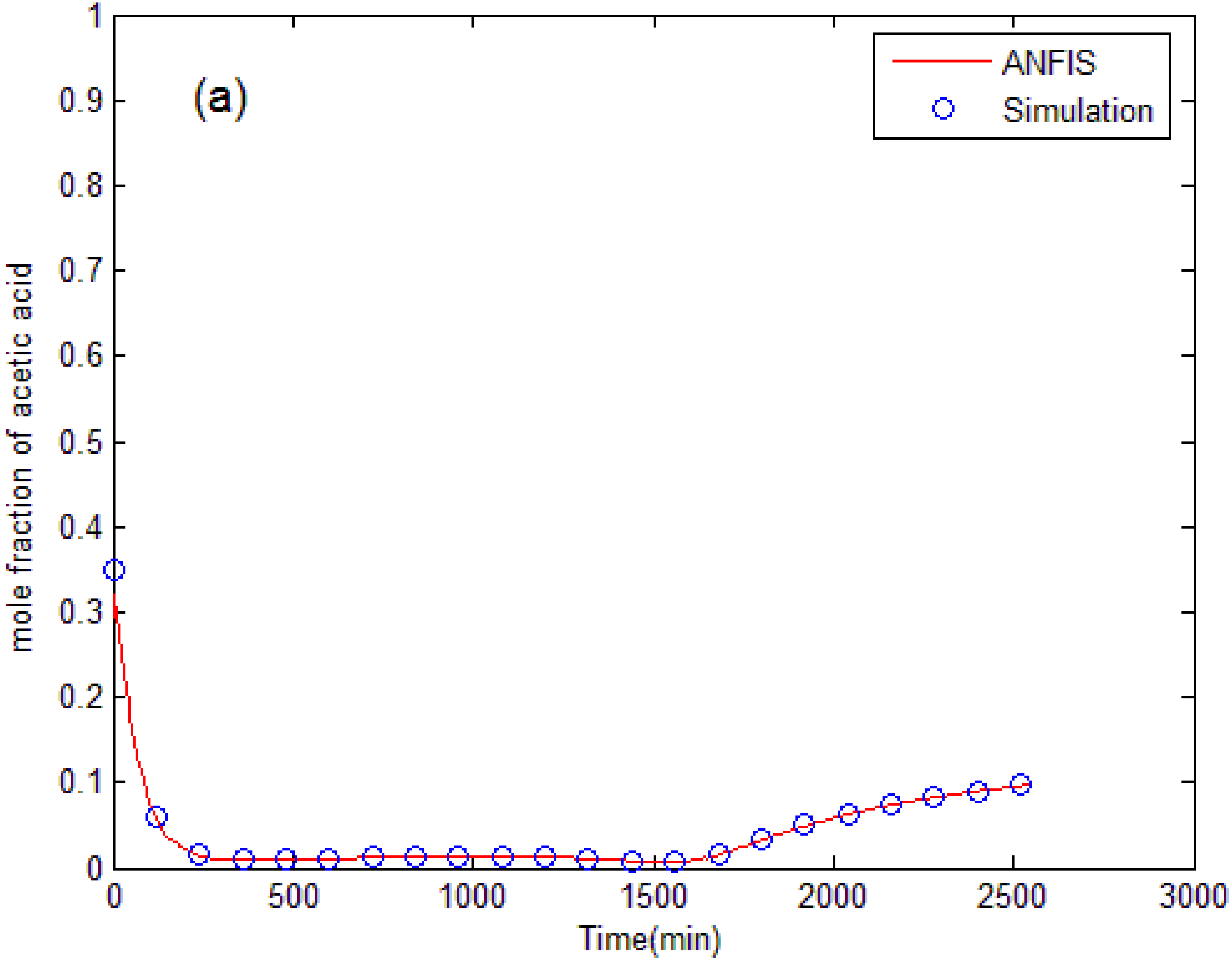
The temperature data of every sampling time is corrupted with a zero mean random Gaussian noise having a standard deviation of 0.2 °C. The temperature data with this noise is considered as the base case data.

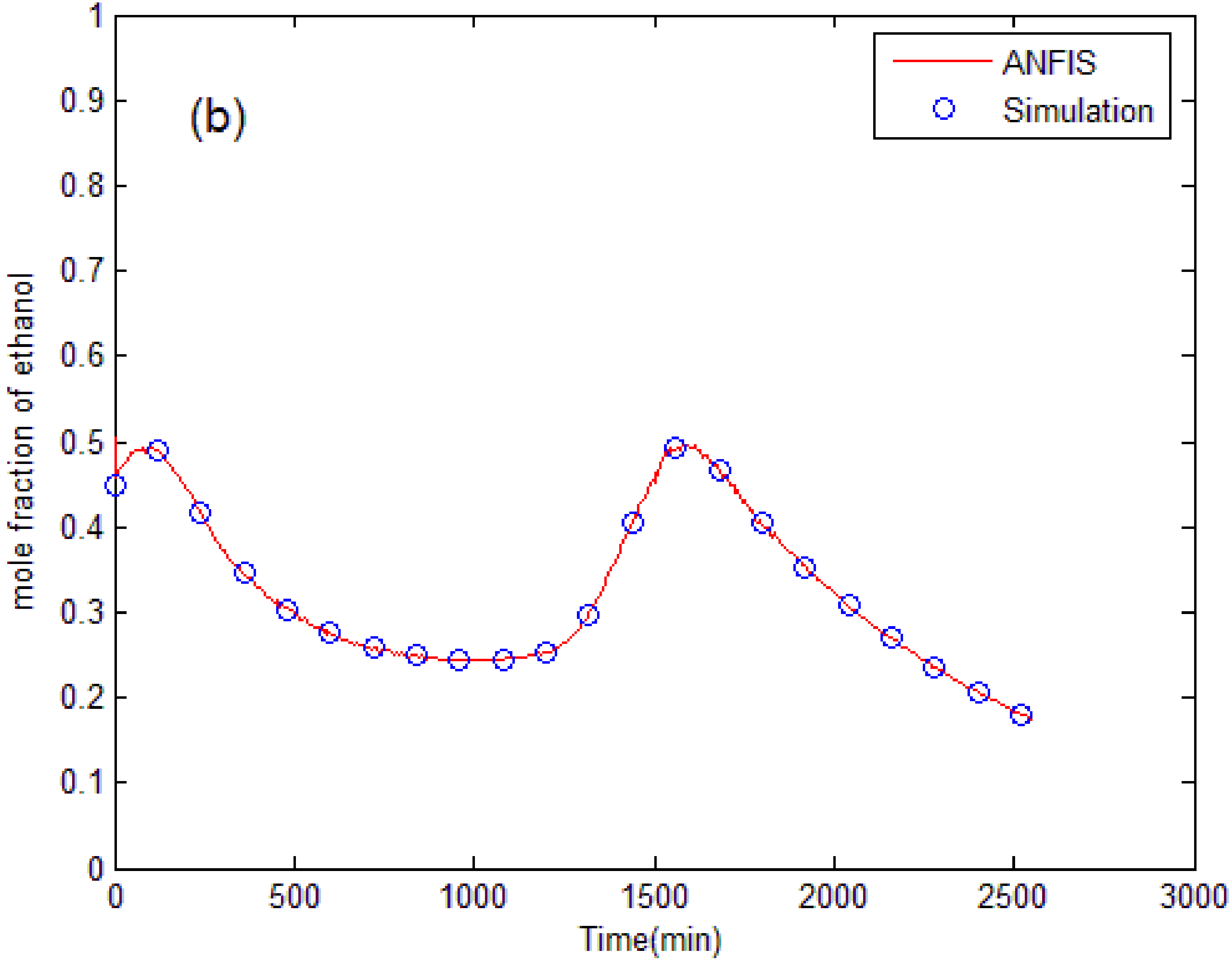
The accuracy of the state estimator is assessed by using an average mean integral squared error (AMISE), which is given by

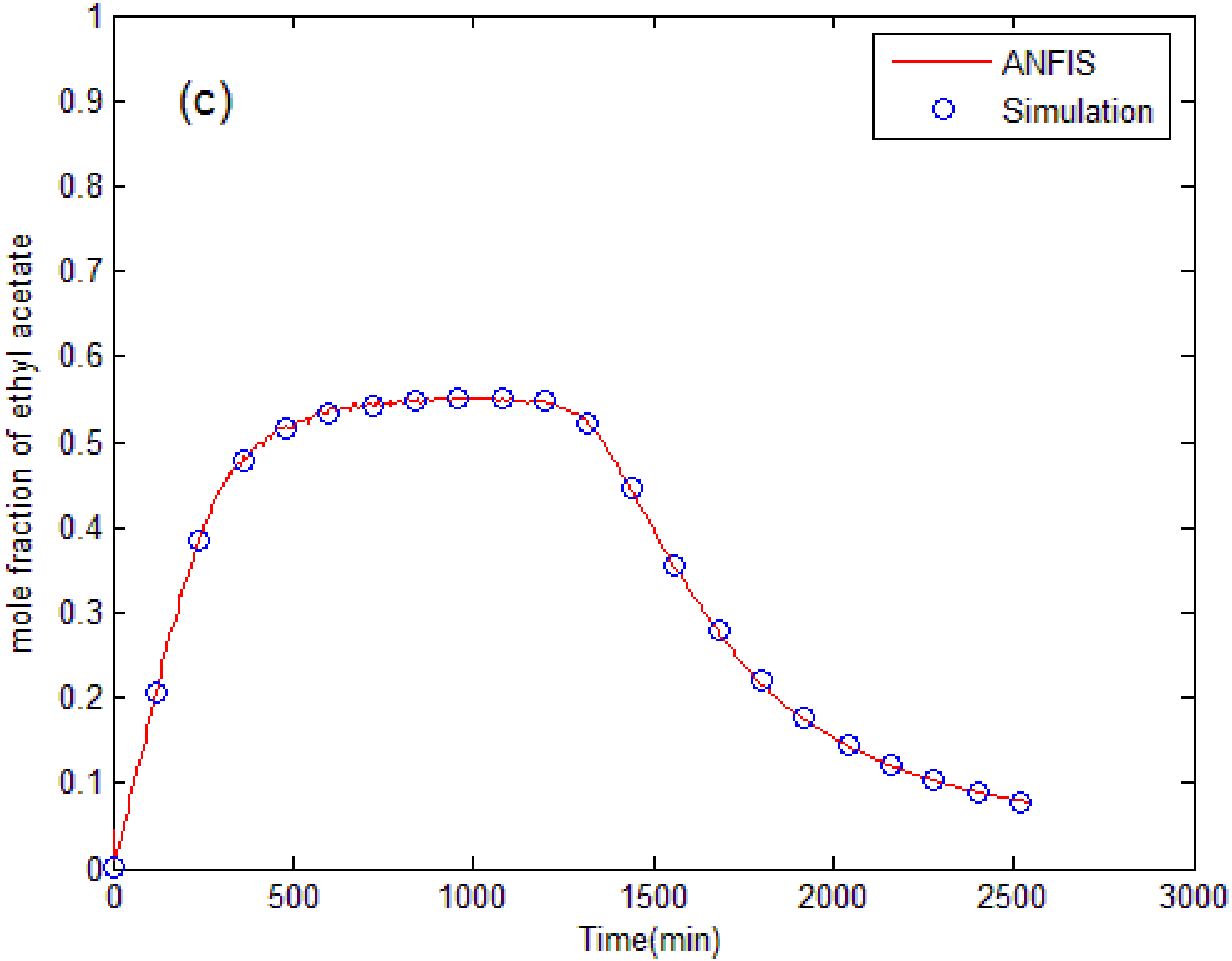
$$AMISE = \frac{1}{N_R} \sum_{j=1}^{N_R} \sqrt{\frac{\sum_{i=1}^{N_S} (X_{EtAc} - \hat{X}_{EtAc})^2}{N_S}}$$

Average mean integral squared error for sensor configuration T_1 and T_{20}

Measurement combination	AMISE	Measurement combination	AMISE
T_2	0.0030	T_{11}	0.0046
T_3	0.0033	T_{12}	0.0096
T_4	0.0030	T_{13}	0.0026
T_5	0.0025	T_{14}	0.0056
$*T_6$	**0.0021**	T_{15}	0.0042
T_7	0.0051	T_{16}	0.0026
T_8	0.0032	T_{17}	0.0045
T_9	0.0026	T_{18}	0.0024
T_{10}	0.0042	T_{19}	0.0024


(a)
ANFIS
Simulation
mole fraction of acetic acid
Time(min)
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
500
1000
1500
2000
2500
3000


(b)
ANFIS
Simulation
mole fraction of ethanol
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
500
1000
1500
2000
2500
3000
Time(min)


(c)
ANFIS
Simulation
mole fraction of ethyl acetate
Time(min)
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
500
1000
1500
2000
2500
3000

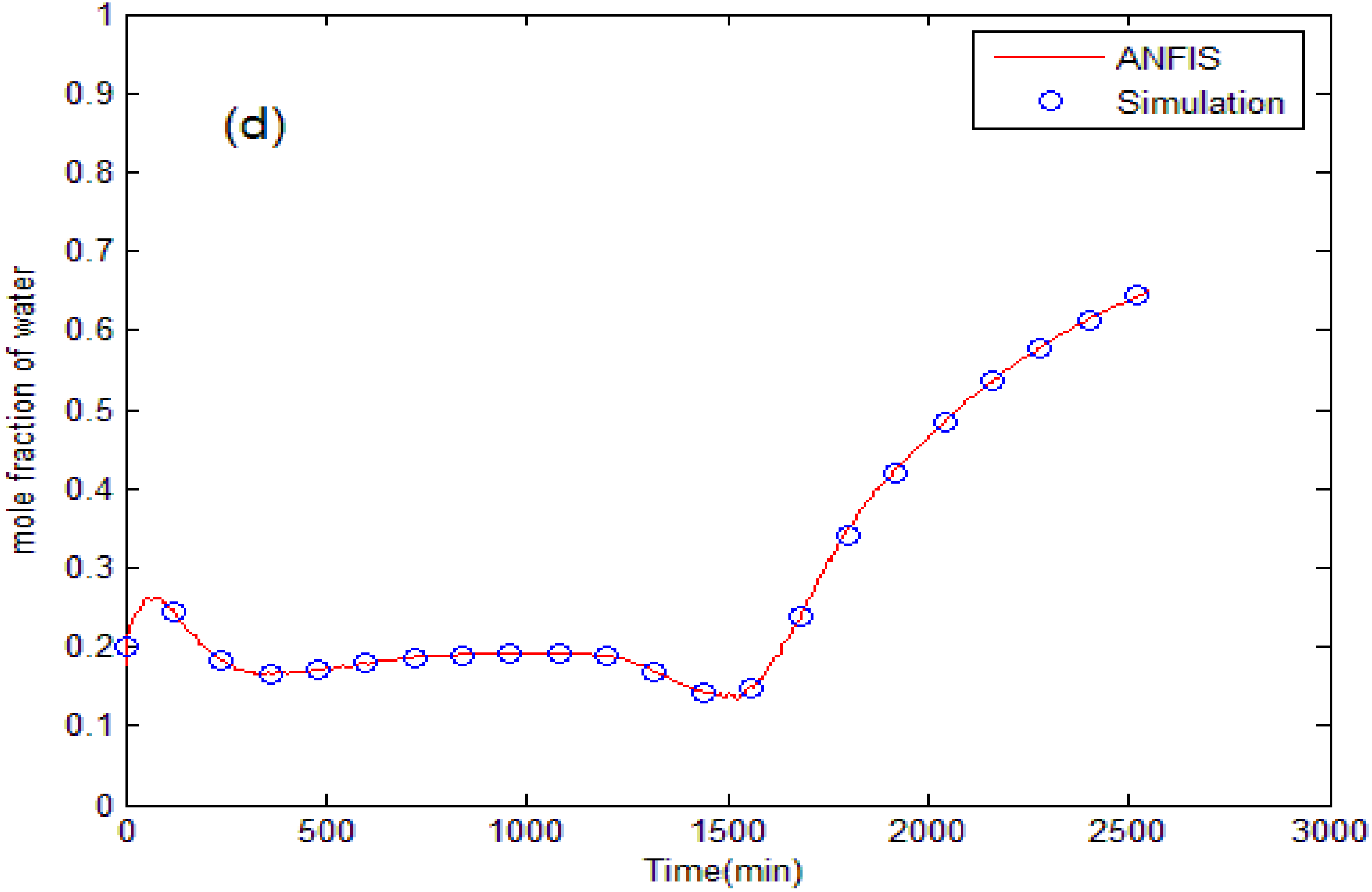


Fig. 7. Comparison of ANFIS with simulation results for (a) acetic acid, (b) ethanol, (c) ethyl acetate, and (d) water mole fraction by -3% step change in reflux ratio

Thank You For Attention