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Dynamic models for start-up operations of batch distillation columns with experimental validation

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Abstract

The simulation of batch distillation columns during start-up operations is a very challenging modelling problem because of the complex dynamic behaviour. Only few rigorous models for distillation columns start-up are available in literature and generally required a lot of parameters related to tray or pack geometry. On an industrial viewpoint, such a complexity penalizes the achievement of a fast and reliable estimate of start-up periods. In this paper, two “simple” mathematical models are proposed for the simulation of the dynamic behaviour during start-up operations from an empty cold state. These mathematical models are based on a rigorous tray-by-tray description of the column described by conservation laws, liquid–vapour equilibrium relationships and equations representative of hydrodynamics. The models calibration and validation are studied through experiments carried out on a batch distillation pilot plant, with perforated trays, supplied by a water methanol mixture. The proposed models are shown by comparison between simulation and experimental studies to provide accurate and reliable representations of the dynamic behaviour of batch distillation column start-ups, in spite of the few parameters entailed.

Keywords: Dynamic simulation; Batch distillation column; Start-up operation; Experimental validation; Water–methanol

1. Introduction

The simulation of batch distillation columns during start-up operations is a very challenging modelling problem because of the inherent and complex dynamic behaviour. The dynamic behaviour of distillation columns during start-up operations corresponds to the time from the initial state to a pseudo-steady state when a defined product composition is obtained. In this way, start-up operations represent very complex transient periods because of the simultaneous drastic changes in many state variables. As these dynamic transitions are always considered as non-productive periods, many researches have been performed with the objective of minimising the start-up time, the energy consumption or the amount of waste products and present the resulting start-up policies

(Gonzalez Velasco, Castresana Pelayo, Gonzalez Marcos, & Gutierrez Ortiz 1987; Ruiz, Cameron, & Gani, 1988; Fieg, Wozny, & Kruse, 1993; Barolo, Guarise, Rienzi, & Trotta, 1994; Sorensen & Skogestad, 1996; Baldon, Strifezza, Basualdo, & Ruiz, 1997; Han & Park, 1997; Furlonge, 2000). Although all the authors agree on the need of a reliable model in order to represent the dynamic behaviour, only few start-up models are reported in literature (Gani, Ruiz, & Cameron, 1986; Albet, Le Lann, Jouila, & Koehret, 1994a, 1994b; Wang, Li, Wozny, & Wang, 2003). In all these different start-up models, distillation columns are described by a rigorous tray-by-tray modelling related to the physical properties and the real geometry. The start-up behaviour is defined according to a start-up sequence that describes the transition on trays from a non-equilibrium a vapour–liquid equilibrium state. The switching conditions between the two states are determined by the bubble point temperature at operating pressure.

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Nomenclature

a	area (m ²)
A	heat transfer area (m ²)
C_p	specific heat (J kg ⁻¹ K ⁻¹)
e	Murphree efficiency
E	internal energy (J)
g	universal gas constant (J mol ⁻¹ K ⁻¹)
h	liquid molar enthalpy (J mol ⁻¹)
H	vapour molar enthalpy (J mol ⁻¹)
hw	weir height (m)
K	equilibrium constant
L	liquid flow-rate (mol s ⁻¹)
lw	weir length (m)
m	mass (kg)
mE	model of internal energy (J)
mh	model of liquid molar enthalpy (J mol ⁻¹)
mH	model of vapour molar enthalpy (J mol ⁻¹)
mK	model of equilibrium constant
μ	model of molar hold-up (mol)
$m\Delta P$	model of pressure losses (Pa)
P	pressure (Pa)
Q	thermal contribution (J s ⁻¹)
R	reflux ratio
t	time (s)
T	temperature (K)
u	molar hold-up (mol)
U	thermal transfer coefficient (J m ⁻² K ⁻¹ s ⁻¹)
v	volume (m ⁻³)
V	vapour flow-rate (mol s ⁻¹)
x	liquid molar fraction
y	vapour molar fraction

Greek letters

α	filling coefficient
β	constant related to physical characteristics of tray
ΔH	heat of condensation (J mol ⁻¹)
ρ	density (kg m ⁻³)

Subscripts

k	plate k
1	condenser
2	plate 2
n	vessel
cf	cooling fluid
hf	heating fluid
ext	ambient medium
kw	plate k wall

Superscripts

i	constituent i
b	bubble point
max	maximum

ml	molar
p	plate wall
tray	tray
*	equilibrium

In industry, many configurations of distillation columns are reported: tray columns (perforated plates, bubble-cap plates, etc.) and packed columns. All these configurations make difficult the set-up of start-up models which require a lot of parameters related to geometrical characteristics. To avoid this issue, we have developed two realistic but “simple” models for the simulation of batch distillation columns start-up. The purpose of the present work is then to thoroughly study, simulate and validate the dynamic behaviour of a batch distillation column during start-up operations using these two dynamic models.

Kister (1979) and Ruiz, Cameron, and Gani (1998) reported that the start-up operation consists of three phases according to the dynamic behaviour. The first stage, called the discontinuous stage, is characterised by its short time period and the discontinuous nature of all variables (hydraulic and transfer variables). The second stage, called the semi-continuous stage is characterised by the non-linear transient of the variables. At the end of this stage, hydraulic variables reach their steady state values. The third stage, called the continuous stage, is characterised by the linear transient responses of all variables. At the end of this stage, all variables reach their steady state values. The aim of this study is to develop accurate models in order to represent the dynamic behaviour during the discontinuous and semi-continuous stages, operated at total reflux. In this way, experiments have been carried out on a batch distillation pilot in order to calibrate and validate the proposed models.

This paper is divided into two main parts. In the first part, a detailed description of the developed models, their characteristics and the related dynamic behaviour is given. The second part describes the experimental device, the different experiments carried out and presents the comparison between experimental and simulated results.

2. General modelling

The treatment of the start-up dynamics requires to simulate a train of successive steps, related to the physical events occurring inside the column. Consequently, the transition from one step to another may lead to discontinuities in the general model structure. In a first part, the management procedure will be described then the general equations of the model will be written. Finally, the specific adaptations for each start-up model will be presented. Thus, a rigorous modelling may require modifications of the general equations model, deter-

mined by physical switching conditions: temperature reaches bubble point, condenser filled up, etc.

As the developed modelling is very complex, its description is divided in three different parts. In a first part, the management procedure of the start-up dynamics is described. In a second part, the general equations of the model are detailed. Finally, the specific adaptations for each start-up model are presented.

2.1. Management procedure of the column dynamics

The management procedure of the start-up steps consists of the following scheme:

1. Detection of an event entailing a new step (e.g. for boiler evaporation: temperature reaching bubble point in the boiler).
2. Resultant modifications of the mathematical model, according to the occurring step (e.g. for boiler evaporation, the equation describing the absence of vapour flow going out is substituted by an equation describing the vapour flow).
3. Accurate and consistent initialisation of the new mathematical model.
4. Solution of the new mathematical model.

This procedure requires to test the occurrence of each possible event at each solver step and so may be substantial calculation time consuming. This characteristic is however compensated by the velocity of the solver used (DISCO: Sargousse, Le Lann, Joulia, & Jourda, 1999) thanks to the use of operator sparse and to an automatic initialisation procedure of the new generated DAE system.

2.2. Mathematical model

The mathematical model results from the restriction to distillation column of a complex dynamic model developed for batch processes simulation (Elgue et al., 2001). In this framework, a tray-by-tray modelling approach of the column has been adopted. Thus, the dynamic model is described by a set of differential and algebraic equations (DAE system) written for each tray.

Ordinary differential equations (ODE) represent energy balances, total and component mass balances. Algebraic equations are composed of constitutive equations such as vapour liquid equilibrium relationships, summation equations, physical property estimations, etc. In order to reduce the complexity of the model, the following typical assumptions have been adopted for each tray:

1. Perfect mixing of vapour bubbles and of liquid phase.
2. Equilibrium relationship between liquid and vapour with possible introduction of the Murphree efficiency.
3. Negligible vapour hold-up compared to liquid one.
4. Constant volume of the liquid hold-up once filling up is completed.

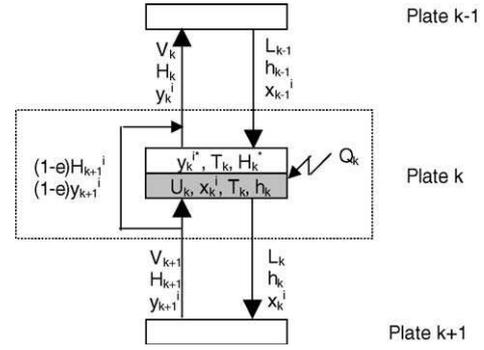


Fig. 1. Details of a tray representation.

The developed modelling offers a great flexibility regarding the kind of distillation columns to simulate: tray columns or packed columns. Packed columns can be modelled as tray columns by the way of the height equivalent of a theoretical plate or height of a transfer unit for a particular type of packing. For tray columns, the adopted description of a given plate (Fig. 1) allows to take into account the real geometry. In fact, the constant volume of the liquid hold-up (assumption 4) can be defined from the real plate characteristics for tray columns (diameter, active area, weir height) or from equivalent characteristics for packed and unknown columns. Nevertheless, the hydrodynamics of the column is not directly taken into account and so particular flow regimes (flooding, weeping, drying) cannot be rigorously represented, i.e. no specific events are defined to detect flooding, weeping or drying conditions. A standard flow regime, limited on one hand by the liquid seal and on the other hand by the downcomer critical velocity (Kister, 1979) is assumed, that ensures a good mixing.

The tray description (Fig. 1) involves that the liquid phase held-up on a plate (k) always comes down to the plate below ($k + 1$) through the downcomer (flow-rate L_{k-1}). This liquid flow is assumed to be instantaneous whatever the height between plates is. The vapour phase going up from the plate below (plate $k + 1$) is shared in two flows: one (flow-rate eV_{k+1}) is going up to the considered plate (plate k) and contributes to the plate equilibrium and the other (flow-rate $(1 - e)V_{k+1}$) is directly going up to the plate above (plate $k - 1$) and by-passes the considered plate. All these vapour flows are assumed to be instantaneous. The sharing constant (e) represents the Murphree efficiency (assumption 2). It can be adjusted to represent the non-ideality of the column plates.

The different plates of the column are numbered from top to bottom: plate 1 represents the condenser and plate n the boiler. The liquid reflux provided by the condenser is introduced at the top of the column (plate 2). Thus, the reflux vector of plate k (R_k) is equal to zero except for plate 2. Hence, each plate k of the distillation column (Fig. 1), including condenser and boiler, is represented by the following equations.

Total mass balance

$$\frac{du_k}{dt} = V_{k+1} + L_{k-1} - V_k - L_k + R_k L_1 \quad (1)$$

Component mass balances

$$\frac{d(u_k x_k^i)}{dt} = V_{k+1} y_{k+1}^i + L_{k-1} x_{k-1}^i - V_k y_k^i - L_k x_k^i + R_k L_1 x_1^i \quad (2)$$

Energy balance

$$\frac{d(u_k h_k)}{dt} = V_{k+1} H_{k+1} + L_{k-1} h_{k-1} - V_k H_k - L_k h_k + R_k L_1 h_1 + Q_k \quad (3)$$

Vapour–liquid equilibrium relationships

$$y_k^i - y_{k+1}^i (1 - e_k) - e_k K_k^i x_k^i = 0 \quad (4)$$

Vapour enthalpy balance

$$e_k H_k^* + (1 - e_k) H_{k+1} - H_k = 0 \quad (5)$$

Summation equation

$$\sum_i (x_k^i - y_k^i) = 0 \quad (6)$$

Hydrodynamic relationship

$$L_k = 0 \quad \text{or} \quad u_k - \mu_k(T_k, P_k, x_k^i) = 0 \quad (7)$$

Liquid–vapour constraint

$$V_k = 0 \quad \text{or} \quad T_k - T_k^b = 0 \quad (8)$$

Liquid enthalpy model

$$h_k - m h_k(T_k, P_k, x_k^i) = 0 \quad (9)$$

Vapour enthalpy model

$$H_k - m H_k(T_k^b, P_k, y_k^i) = 0 \quad (10)$$

Equilibrium constant model

$$K_k^i - m K_k^i(T_k^b, P_k, x_k^i, y_k^i) = 0 \quad (11)$$

Pressure model

$$P_{k+1} - P_k - m \Delta P_k = 0 \quad (12)$$

The thermal contributions of the different parts of the column are the following:

For condenser

$$Q_1 = U_{1,cf} A_{1,cf} (T_{cf} - T_1) - V_2 (H_2 - \Delta H_1) \quad (13)$$

For boiler

$$Q_n = U_{n,hf} A_{n,hf} (T_{hf} - T_n) + U_{n,ext} A_{n,ext} (T_{ext} - T_n) \quad (14)$$

For column plate

$$Q_k = U_{k,ext} A_{k,ext} (T_{ext} - T_k) \quad (15)$$

During start-up, the transition from one step to another may lead to modification of the equation models. The general modelling then entails a hybrid model including equations discontinuities determined by switching conditions related

to physical phenomena (boiling, tray filling up) and concerning hydrodynamic relationship and vapour–liquid constraint. Therefore, the formulations of Eqs. (7) and (8) vary according to the considered start-up step.

Correlations (Dream, 1999) are used to determine heat transfer coefficients. A complete physical property estimation system with associated data bank (Prophy[®]; Le Lann, Joulia, & Kohret, 1988) is also used for models determination: physical properties models, hydrodynamic relationship and, pressure drop. Physical properties (density, specific heat, enthalpies, molar volume, bubble point and vapour–liquid equilibrium constant) are estimated from typical laws: equation of state and activity coefficient models (UNIFAC or NRTL). Formulations of the other models are given in Eqs. (16)–(19). It has to be noted that different formulations can be adopted for each model, according to the desired level of complexity. For instance, hydrodynamic relationship can be defined by a simple volume constraint (Eq. (19)) or by typical formulations such as Francis formula (Eq. (20)).

Hold-up model

$$\mu_k = \frac{a_k^{\text{tray}} h w_k}{v_k^{\text{ml}}} \quad \text{simple formulation} \quad (16)$$

$$\mu_k = \frac{\rho_k}{m_k} a_k^{\text{tray}} \left[h w_k + 1.41 \left(\frac{L_k m_k}{\sqrt{g \rho_k l w_k}} \right)^{2/3} \right] \quad \text{Francis formula} \quad (17)$$

Pressure drop model

$$m \Delta P = \text{constant} \quad \text{simple formulation} \quad (18)$$

$$m \Delta P = \beta^{\text{tray}} V_k^2 \quad \text{formulation related to tray geometry} \quad (19)$$

DISCO (Sargousse et al., 1999), a general solver of DAE system based on the Gear method (Gear, 1971) allows to obtain the numerical solution of the developed mathematical model. Besides its accuracy and numerical robustness, DISCO allows to develop a complex management procedure of the column dynamic from its event detection facilities and its consistent initial condition calculations.

2.3. Start-up models

The aim of the present work is to represent the dynamic behaviour of batch distillation columns during start-up operations, particularly during the period described by Ruiz, Cameron, and Gani (1988) as the discontinuous stage. During this period, important changes (large dynamics) of thermodynamic variables take place inside the column. Two models describing the dynamic behaviour of start-up have been developed. The second model is more realistic and complex than the first one. Consequently, it is called “realistic model” by opposition to the first one, called “simple model”. These

models, with their advantages and drawbacks, are detailed in the following sections.

2.3.1. Simple model

This model is based on an improved version of the start-up model developed by [Albet Le Lann, Joulia, and Koehret \(1994a,b\)](#). The introduction of one additional parameter, the plate filling ratio, constitutes the main difference. During the column start-up, all the vapour flow going up from the lower plate is assumed to instantaneously condense on the considered plate, originally cold. As vapour continue to come from the plate below through the plate holes, the liquid is held-up on the plate. Thus, the liquid hold-up on the plate increases. Moreover, as fast as it fills up, the plate heats up. At the beginning of the filling up, the plate is assumed too cold to generate vapour flow. The filling ratio represents the value of the liquid hold-up from which the plate is hot enough to generate vapour flow that goes up to the plate above. In the present paper, this event is called local vapour generation event (LVGE). Consequently, the following steps sequence composes the modelling of the dynamic behaviour:

1. Heat is introduced into the boiler.
2. The boiler temperature reaches bubble point. Vapour starts going up and condensing on the upper plate which begins to fill up.

$$\text{Event : } T_n = T_n^b$$

Modification of Eq. (8) :

$$V_n = 0 \text{ replaced by } T_n - T_n^b = 0$$

3. Liquid hold-up reaches the filling ratio (α) in plate $n - 1$. Vapour starts to go up from the plate and to condense on the upper plate.

$$\text{Event : } v_{n-1} - \alpha v_{n-1}^{\max} = 0$$

Modification of Eq. (8) :

$$V_{n-1} = 0 \text{ replaced by } T_{n-1} - T_{n-1}^b = 0$$

4. From plate to plate, vapour reaches the top of the column. When the top tray generates a vapour flow, the condenser starts to fill up.
5. The condenser is completely filled up. Total reflux or fixed reflux is introduced into the column. The top tray ends to fill up, for a $(1 - \alpha)$ fraction.

$$\text{Event : } v_1 - v_1^{\max} = 0$$

Modification of Eq. (7) :

$$L_1 = 0 \text{ replaced by } u_1 - \text{mu}_1(T_1, P_1, x_1^j) = 0$$

6. The top tray is completely filled up. Liquid starts falling down the downcomer to the lower plate. In this way, step-by-step plates ends to fill up.

$$\text{Event : } v_k - v_k^{\max} = 0$$

Modification of Eq. (7) :

$$L_k = 0 \text{ replaced by } u_k - \text{mu}_k(T_k, P_k, x_k^j) = 0$$

7. The bottom plate is completely filled up. Discontinuous stage ends.

Simplicity is the main advantage of this model. In fact, apart from the typical characteristics of the column, only one parameter (the filling ratio) allows to describe the dynamic behaviour. According to the initial conditions of the column, this ratio varies in the range from 0 to 1. The filling coefficient tends towards 1 or 0 for start-ups, respectively, from cold state or hot state.

However, this model appears not able to represent the cases of trays filled up, with liquid falling down the downcomers before filling up of the condenser and reflux flow introduced inside the column. Such sequence may occur in the case of start-ups from cold state of columns with a high thermal inertia and/or high thermal losses. This drawback leads us to develop another more realistic model, suitable with the real physical behaviour of the column.

2.3.2. Realistic model

This model is partly based on the previous one. A new definition of the LVGE occurrence constitutes the main difference. In this model, the vapour going up from the lower plate is assumed to condense on the wall of the considered plate (which heats up) before falling down. The falling liquid is assumed without under-cooling (i.e. liquid at its bubble point temperature) and to hold-up on the plate below. Thus, as fast as the plate wall heats up, the plate below fills up. Once the plate wall temperature reaches the liquid bubble point temperature, the vapour goes up through the plate and starts to condense on the plate wall above, before falling down to the plate which starts to fill up. In this way, according to its thermal and geometrical characteristics, a plate may supply vapour to the plate above before or after the plate below is completely filled up. The dynamic behaviour of the start-up is then represented by the following step sequence:

1. Heat is introduced into the boiler.
2. The boiler temperature reaches bubble point. Vapour starts to go up and to condense on the upper plate wall which is heated.

$$\text{Event : } T_n - T_n^b = 0$$

Modification of Eq. (8) :

$$V_n = 0 \text{ replaced by } T_n - T_n^b = 0$$

Modification of Eq. (7) :

$$L_{n-1} = 0 \text{ replaced by } L_{n-1} = V_n$$

3. The plate wall reaches the liquid bubble point temperature. Vapour goes up and starts to heat the upper plate. The resultant condensation phenomenon starts to fill up the plate.

$$\text{Event : } T_{n-1}^p = T_n^b$$

Modification of Eq. (8) :

$$V_{n-1} = 0 \quad \text{replaced by} \quad T_{n-1} - T_{n-1}^b = 0$$

Modification of Eq. (7) :

$$L_{n-2} = 0 \quad \text{replaced by} \quad L_{n-2} = V_{n-1},$$

$$L_{n-1} = V_n \quad \text{replaced by} \quad L_{n-1} = 0$$

4. Vapour gradually reaches the top of the column. When the LVGE occurs at the top, the condenser starts to fill up.
5. The condenser is completely filled up. Total reflux is introduced inside the column. The top tray finishes to fill up (except if it has already been done).
Event : $v_1 - v_1^{\max} = 0$

Modification of Eq. (7) :

$$L_1 = 0 \quad \text{replaced by} \quad u_1 - \mu u_1(T_1, P_1, x_1^i) = 0$$

6. The top tray is completely filled up. Liquid starts to fall down through the downcomer to the lower plate. In this way, plates end to fill up step-by-step (except if it has already been done).
Event : $v_k - v_k^{\max} = 0$

Modification of Eq. (7) :

$$L_k = 0 \quad \text{replaced by} \quad u_k - \mu u_k(T_k, P_k, x_k^i) = 0$$

7. The bottom plate is completely filled up. Discontinuous stage ends.

In consequence of this specific modelling, two new equations with the associated variables have been added to the model (Eqs. (20) and (21)). The plate wall internal energy is estimated from the tray geometry by Eq. (22).

Plate wall energy balance

$$\frac{dE_k^p}{dt} = V_{k+1}H_{k+1} - L_k h_k + U_{kw,ext}A_{kw,ext}(T_{ext} - T_k^p) \quad (20)$$

Plate wall internal energy model

$$E_k^p - mE_k^p(T_k^p) = 0 \quad (21)$$

Plate wall internal energy

$$mE_k^p = m_k^p C_p^p T_k^p \quad (22)$$

In comparison to the previous one, this model is more complex and needs additional data: the plate wall mass (related to the plate geometry) and the plate wall specific heat. In return,

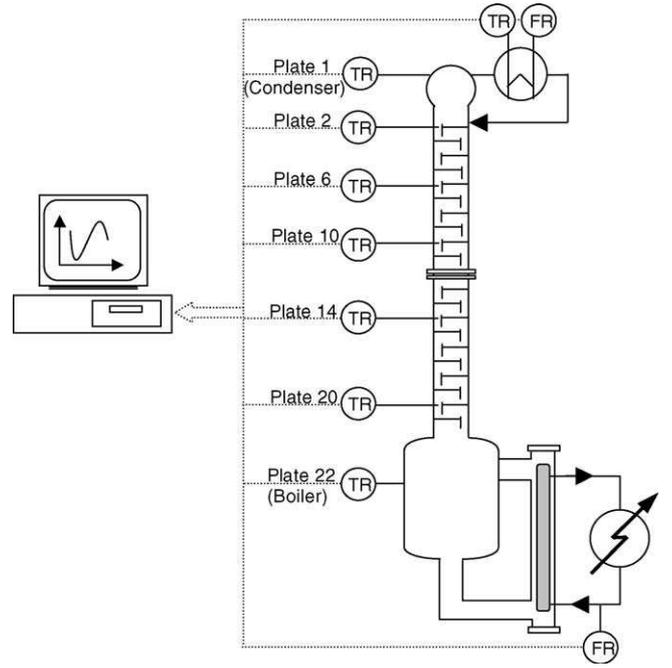


Fig. 2. Batch distillation pilot plant.

it allows to simulate complex sequences of start-ups from cold and empty state. Moreover, the thermal inertia due to vapour condensation on the column wall between two plates can also be considered, through a modification of the plate wall energy model.

3. Experimental device

For the purpose of the present study, a batch distillation column, composed of two glass elements of 1 m height and 100 mm diameter, has been used (Fig. 2). Each element is packed with 10 glass perforated plates of Oldershaw type. Oldershaw plate is provided with circular, vertically disposed holes. The vapour rising through these holes prevent the condensate to pass through in the opposite direction. A weir and overflow pipe, mounted in the centre of the plate, keeps a constant low level of liquid on the plate and ensures a good internal reflux distribution. Temperatures inside the column are recorded every other plate. Samples of the liquid hold-up can be withdrawn on every plate.

The bottom of the column is composed of a glass vessel (volume: 50 l, diameter 400 mm) connected to a thermosiphon. The heat transfer area of the thermosiphon is 0.18 m². Thermosiphon is supplied by a heat transfer fluid

Table 1
Details of experiments carried out

Experiment	Heat transfer fluid flow-rate (m ³ h ⁻¹)	Initial vessel volume (l)	Initial column temperature (°C)	Room temperature (°C)
1	1.5	39.0	27	27
2	1.2	38.5	30	30
3	0.9	37.5	25	25

(Gilotherm) circulating in a boiler at about 160 °C. A control valve, between the boiler and the thermosiphon, regulates the heat transfer fluid flow-rate. The temperature of the medium inside the vessel is also recorded.

The condenser is located 2.5 m up from the column bottom. It is composed of a spiral coil heat exchanger (0.75 m² heat transfer area) provided with cooling water. The flow-rate, input and output temperatures (around 20 °C) of the cooling water are recorded. A control valve allows to control the water flow-rate. The condenser is operated as a total condenser with the vapour condensing at the top of the column and flowing back to a reflux divider located below. An adjustable timer controls periodic switching between distillate tanks and reflux to the column. A full SIEMENS numerical system (DCS) allows to record all pertinent information.

As plates are numbered from top to bottom, the different parts of the pilot column are indexed as follows (Fig. 2):

- plate 1: condenser,
- plate 2: top of the column,
- plate 21: bottom of the column,
- plate 22: thermosiphon.

4. Application results

The purpose of this experimental study is to obtain an accurate measurement of the dynamic behaviour during the start-up of the column and particularly during the discontinuous stage. In this way, only the total reflux period has been studied. In order to increase the time of the dynamic behaviour, experiments have been carried out from cold state and at atmospheric pressure.

The vessel is fed with approximately 40 l of water-methanol mixture, with a composition of around 82% molar of water. The exact composition is determined before each experiment by gas chromatography. This well-known binary mixture has been chosen in order to avoid sample taking in the column during experiments. In fact, temperature measurements allow to know the exact tray composition by the way of the vapour-liquid equilibrium diagram. A water-methanol mixture has also been chosen for the volatility variations it presents. Furthermore, this binary mixture is in agreement with assumption 3 of the mathematical model set-up (vapour density is negligible compared to the liquid density).

The column start-up has been studied through three experiments, with different heating powers and hence different dynamic behaviour. Therefore, three different flow-rates of the heat transfer fluid have been supplied to the thermosiphon by the way of the control valve. Table 1 gathers these flow-rates and the other operating conditions of each experiment.

The three different experiments carried out have been simulated. In order to verify the accuracy of the developed models, experimental results are compared to the associated simulations. NRTL model has been chosen to estimate the phys-

Table 2
NRTL parameters of water-methanol mixture

A_{12}	-243.55
A_{21}	872.81
α_{12}	0.2994

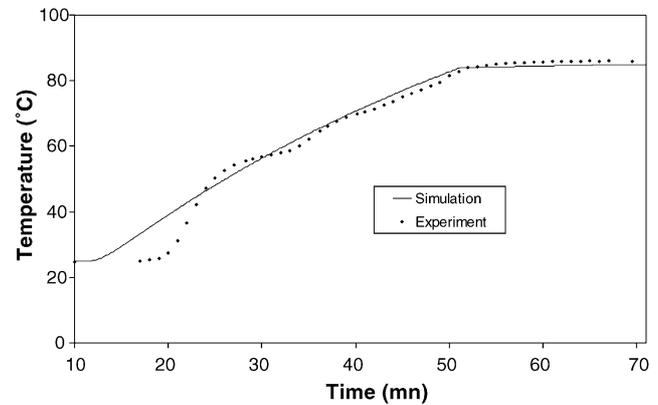


Fig. 3. Variations of vessel temperature for experiment 1.

ical properties of the water-methanol mixture. The NRTL numerical parameters used, according to DECHEMA data, are given in Table 2.

Considering the simulation calibration and validation, two points appear particularly critical: the column behaviour and the thermosiphon representation. Consequently, in order to highlight the benefits of the developed models, the vessel temperature variations on one hand and the temperature variations of the different column plates on the other hand are presented.

4.1. Thermosiphon simulation

With regard to the thermosiphon representation, the temperature measured inside the vessel is directly compared to the simulation results, as shown in Figs. 3–5 for experiments

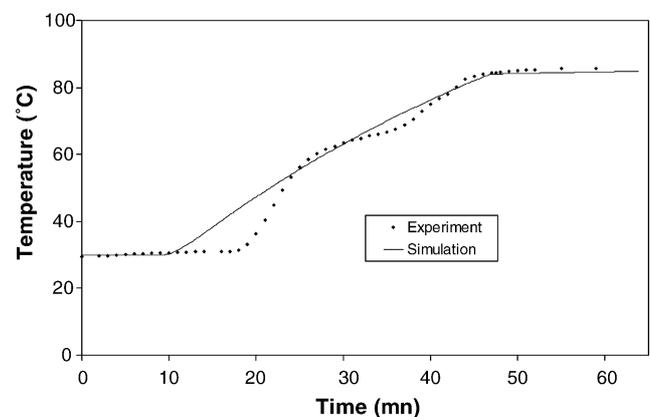


Fig. 4. Variations of vessel temperature for experiment 2.

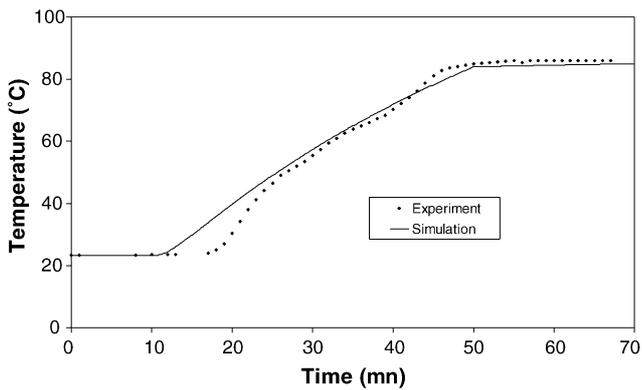


Fig. 5. Variations of vessel temperature for experiment 3.

1–3, respectively. As can be seen, simulations show a good agreement with experiments. Nevertheless, experimentally at the beginning of the start-up, the thermosiphon heat supply presents an oscillatory behaviour. This behaviour is directly related to the thermosiphon technology. In fact, the liquid heated by the thermosiphon only flows to the vessel when its temperature is high enough. Therefore, the heat internal reflux inside the vessel shows an oscillatory behaviour, decreasing until the vessel mixture reaches bubble point. As the aim of this study is not to obtain an accurate representation of thermosiphons, this oscillatory behaviour has not been integrated in the mathematical model and then does not appear in simulations.

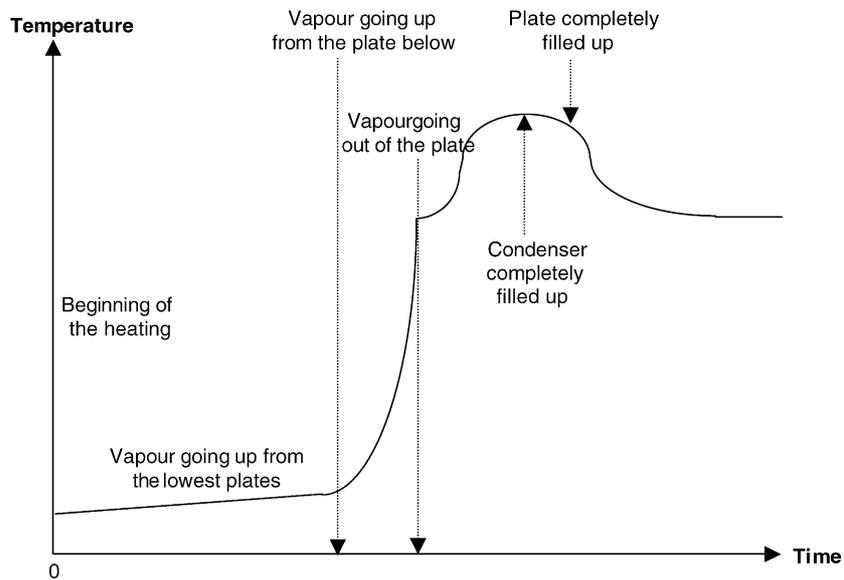


Fig. 6. Analysis of tray dynamics.

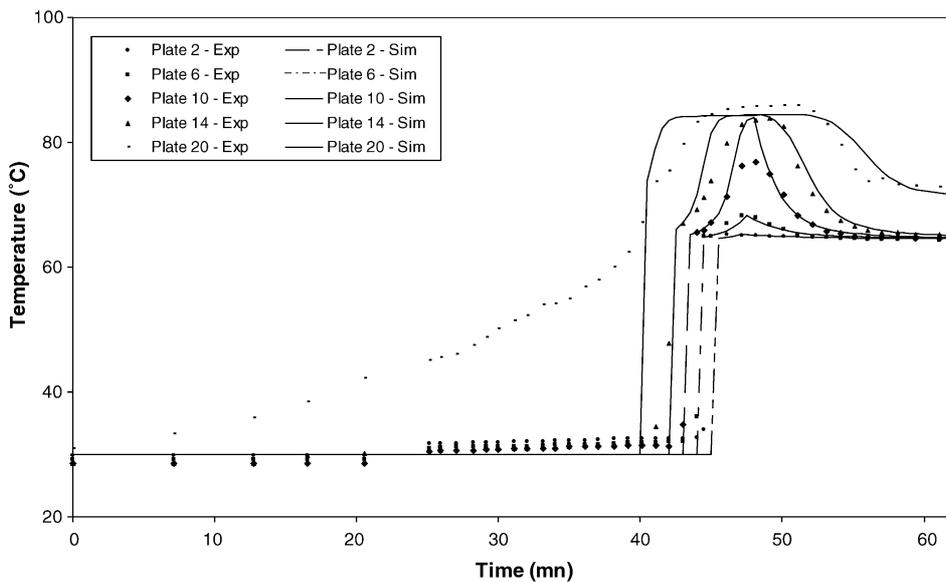


Fig. 7. Temperature profiles, experiment 1 conditions, simple model.

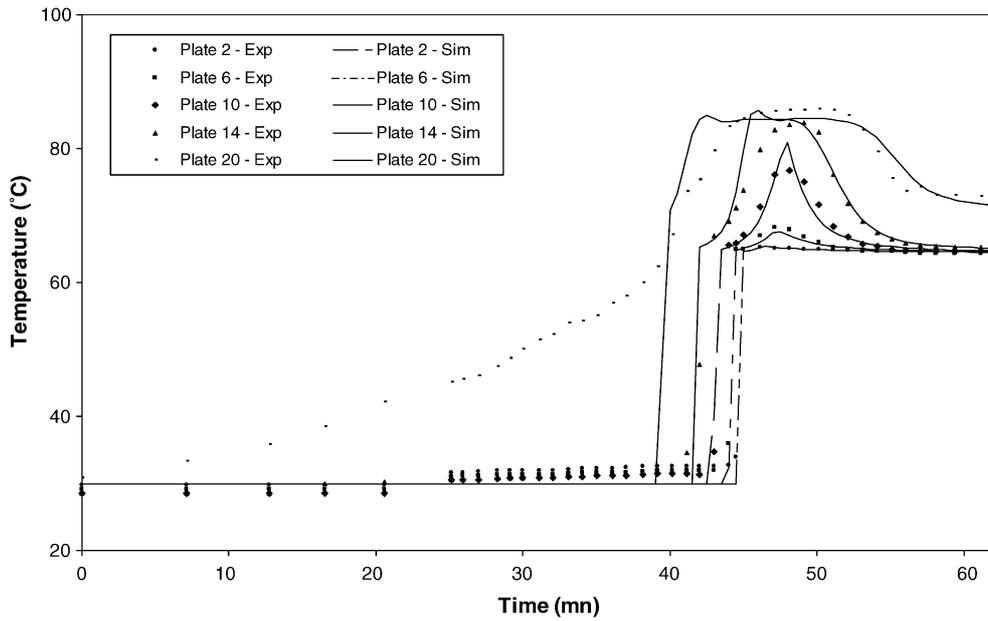


Fig. 8. Temperature profiles, experiment 1 conditions, realistic model.

4.2. Start-up simulation

The study of the temperature profiles inside the column appears as a good way to validate the developed start-up models. In fact, temperature sensors are easy to implement and offer a fast and reliable records. Measurements of hydrodynamic variables (such as liquid flow-rate, liquid level or pressure drop) are very difficult to set-up and cannot offer stable records because of the constant local variations on plates. Moreover, temperature measurements represent the standard measurement in industry. Finally, the analysis of the temperature records offers dynamic information about both hydrodynamic and thermodynamic variables, as shown in Fig. 6:

bubble points, filling up of plates and condenser, total start-up period, etc.

With regard to the column start-up, the validation approach differs from the thermosiphon study. In a first part, using experiment 1 results, the specific parameters of each model have been estimated from temperature profiles:

- the filling ratio (α), for the simple model,
- the mass added to each plate wall to represent the thermal inertia of the column, for the realistic model.

In a second part, experiments 2 and 3 have been simulated with the same parameters value in order to exemplify the models accuracy.

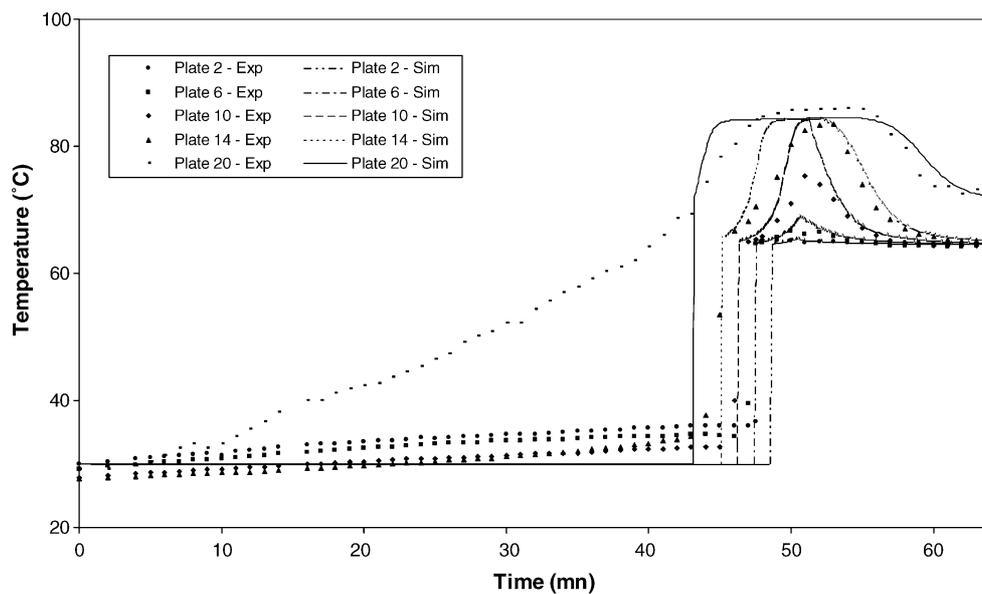


Fig. 9. Temperature profiles, experiment 2 conditions, simple model.

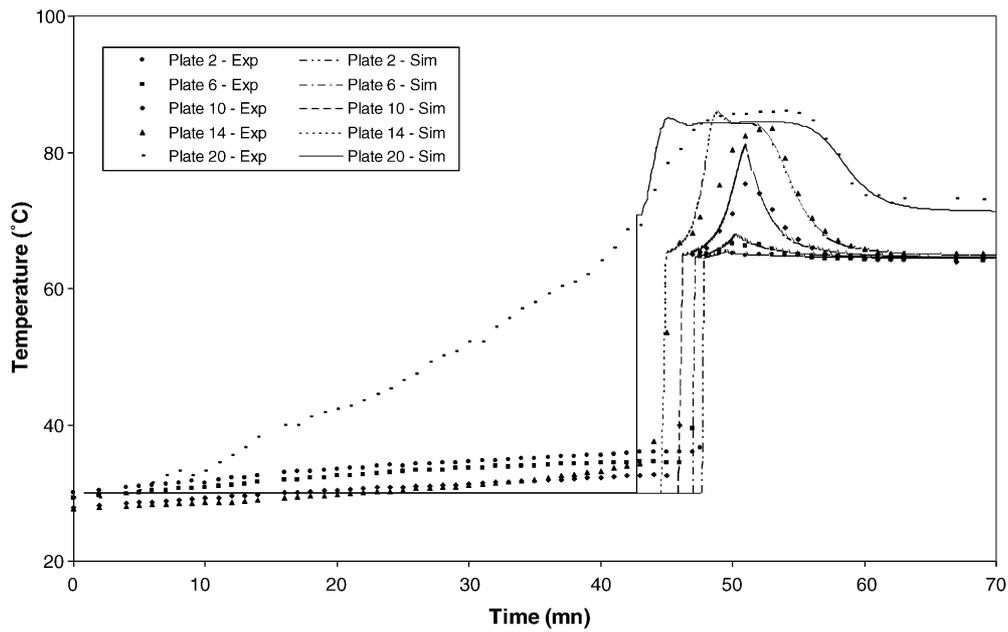


Fig. 10. Temperature profiles, experiment 2 conditions, realistic model.

For the simple model, experiment 1 allows to evaluate the filling ratio. Initially, the column is cold, and so the filling coefficient is set to a value close to 1, equal to 0.95 (Fig. 7). In order to simplify its determination, the filling coefficient is assumed to be constant all along the column (i.e. equal for each plate of the column).

For the realistic model, the thermal characteristics of each plate have been defined from the plate geometry: specific heat and mass of the plate wall. In order to take into account the thermal inertia, due to the cold wall of the column, a mass addition has been defined in the internal energy model. This

mass addition represents the thermal environment of each plate. In this way, the addition depends of the plate position inside the column, the more a plate is high, the less thermal inertia is felt (Fig. 8).

Figs. 7 and 8 emphasize a limitation peculiar to our model. Experimentally, bottom plates (particularly plates 21 and 20) appear to heat up from the beginning of the start-up. In fact, even before temperature reaches bubble point, a slight vapour flow, due to the thermosiphon heat, escapes from the vessel and begins to heat bottom plates. In our model, vapour flows are only generated when bubble point is reached. Therefore,

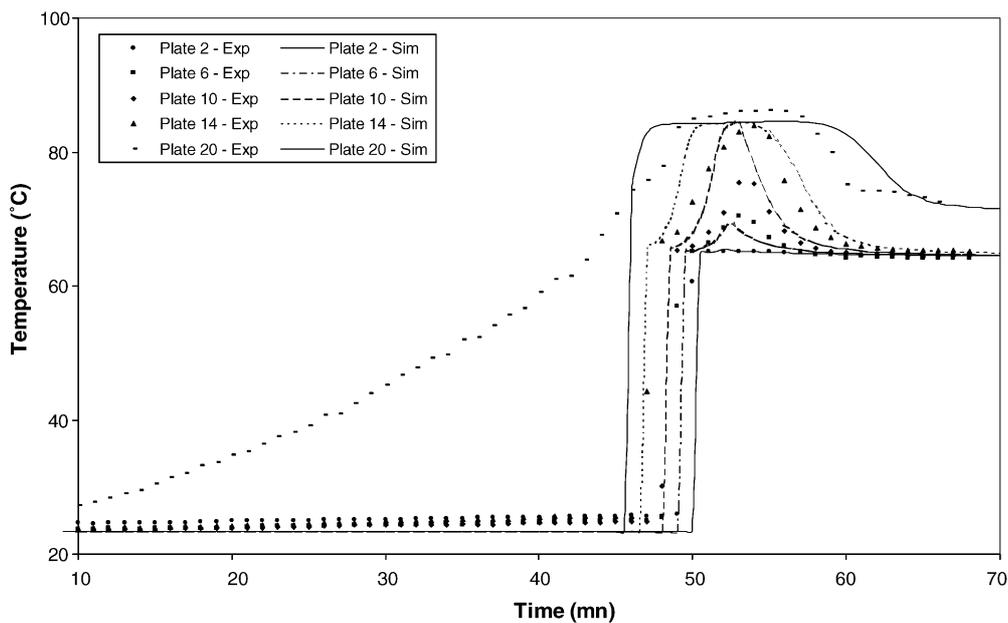


Fig. 11. Temperature profiles, experiment 3 conditions, simple model.

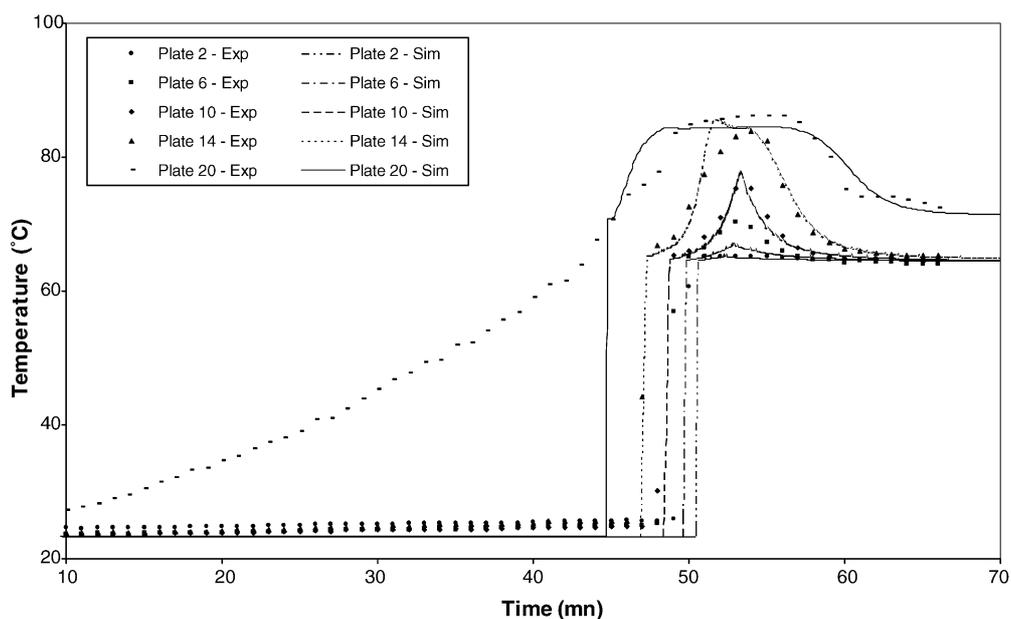


Fig. 12. Temperature profiles, experiment 3 conditions, realistic model.

in the period before the water–methanol mixture reaches bubble point (the first 40 min), experiment and simulation temperatures of bottom plates differ.

The set of estimated parameters has been used for simulation of experiments 2 and 3. Figs. 9–12 show, for experiments 2 and 3, the temperature variations inside the column, respectively, for the simple and the realistic model. In Figs. 10 and 12, the realistic model appears more accurate than the simple one. Nevertheless, the good agreement between experimental and simulated results in both cases allows to validate the two models of column start-up.

The developed start-up models need a preliminary tuning phase in order to obtain fully satisfactory perfor-

mances. In fact, the specific parameters of each model require to be adjusted to the configuration of the considered batch distillation column. Once this preliminary tuning phase has been performed, the results show that the models offer accurate and reliable representations of column start-ups, as can be seen on the different comparison realised between experimental and simulated results.

Hydrodynamic variables are significant of start-up modelling since they dictate the transition states of the trays. To emphasize the changes involved by the developed models, the liquid hold-up variations occurring on trays 10 and 20 are reported in Figs. 13 and 14. These figures allow to assess the

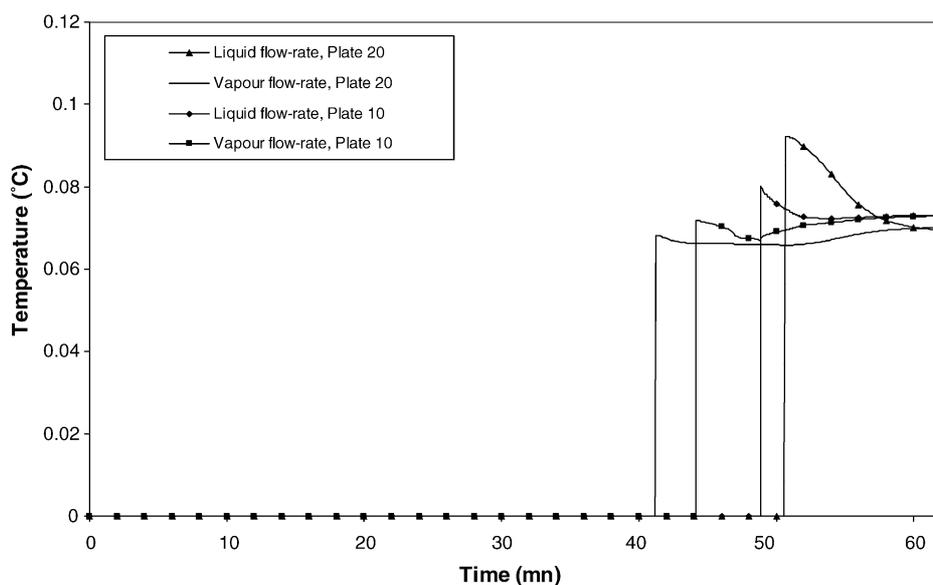


Fig. 13. Flow-rate profiles, experiment 1 conditions, simple model.

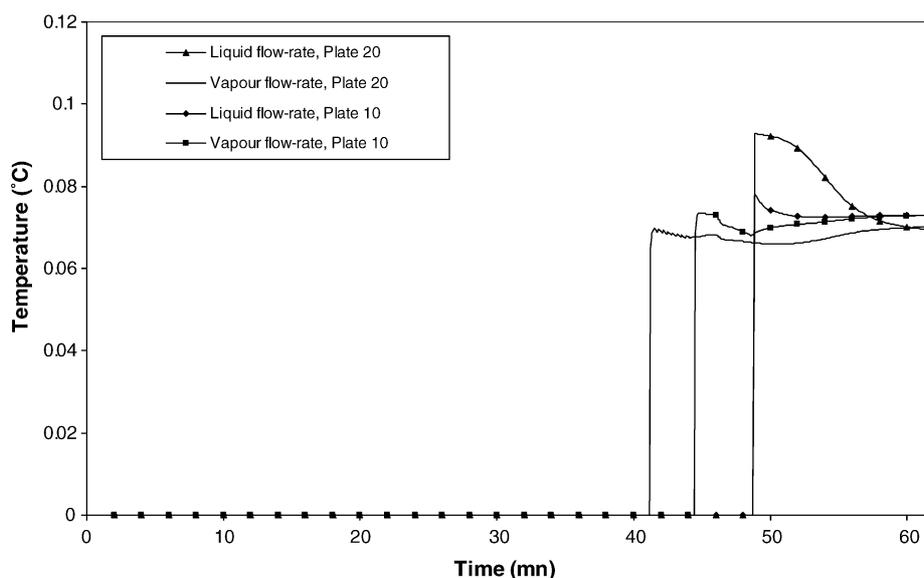


Fig. 14. Flow-rate profiles, experiment 1 conditions, realistic model.

modelling sequences defined to represent the column start-up and to highlight the models differences.

5. Conclusion

Two models have been developed for the representation of start-up operations of batch distillation columns: a simple model and a realistic model. The proposed models are defined by a succession of steps based on the real behaviour. Therefore, two start-up sequences are proposed, starting from an empty cold state. The train of steps is determined by switching conditions related to physical phenomena, that leads to a hybrid formulation of the model equations. On a resolution viewpoint, a specific solver is used to clear up event detection, system initialisation and calculation time issues.

The two mathematical models have been successfully applied for the simulation of a batch distillation column start-up. Three application cases allow to emphasize the advantages and drawbacks of each model. Thus, the used of one model rather than the other depends on the desired representation. The simple model allows, with only one adjustable parameter, a fast and reliable representation. The realistic model gives a more accurate representation but needs more parameters and entails additional tuning efforts and so further tuning time.

The proposed models offer a simple but realistic tray-by-tray description of the start-up. In fact, only one specific parameter per tray for the simple model and two for the realistic one, are required. Moreover, the level of the modelling complexity can be adjusted to the desired solution (simple and fast, accurate) by the introduction of a more complex hydrodynamic description. Furthermore, the modelling is not directly linked to a specific tray geometry and so is suitable for various column configurations: perforated plate, bubble-cap

plate, packed, etc. On an industrial viewpoint, such simplicity and flexibility appear as significant features.

In the field of column optimisation, a correct estimation of the period from empty and cold state to steady state is of prime necessity. In this way, the developed models offer significant improvements compared to the traditional modelling based on an initial state derived from a steady state at total reflux. Therefore, in the case of optimisation problems including time-dependant objective function, the accurate estimation of start-up times provided by the proposed models appears very interesting. Perspectives are not only focused on the optimisation of batch distillation start-up policy but also on optimisation of global processes integrating batch distillation. In this way, the developed modelling has been satisfactory used for the optimisation of the solvent changing policy of a batch pharmaceutical application (Elgue et al., 2001).

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