Web-based Modelling of Chemical Processes Using MOSAIC : A Reactive Distillation Case Study

Chieng Tiew Hing
Process Systems Engineering Group (PSERG)
FKKSA, Universiti Malaysia Pahang
Kuantan, Malaysia
thing90@live.com

Mohamad Rizza Othman
Process Systems Engineering Group (PSERG)
FKKSA, Universiti Malaysia Pahang
Kuantan, Malaysia
rizza@ump.edu.my

***Abstract* - Equation oriented modelling of chemical processes can be done using different modelling environments such as Matlab, Fortran and gPROMS. However, it is a time consuming process and require a lot of programing effort. Since the modelling is done using their own programming languages, it has the poor readability. Thus, users prone to make the mistakes during code writing and error identification will be tedious. In this study, a new modelling environment called MOSAIC will be used for modelling of chemical processes. MOSAIC is a web based modelling software and it introduces a novel approach to equation based modelling. To demonstrate its capability, a reactive distillation (RD) model is used as a case study. The RD results from the modelling work in MOSAIC fitted well with the results found in literature. Using MOSAIC, the modelling effort is possible even without the knowledge of programming languages. In addition, it is fond to be less errors and saves time. A comparison with other modelling software is also presented indicate the advantages and disadvantages of MOSAIC. Such assessment is important to help in deciding to use MOSAIC for any modelling works.**

Keywords—modelling, reactive distillation

I. INTRODUCTION (*HEADING 1*)

MOSAIC is a web-based equation oriented application for modelling of chemical processes. It is designed for minimization of modelling errors and programming effort, avoid programming errors, enhance file documentation and encouragement of cooperative work. As explain in Kuntsche et al. [1], one of the important features of MOSAIC is modelling at the documentation level. It means that MOSAIC able to perform modelling work at the documentation level. The model equations in the documents i.e. books, journals etc. can be written easily in MOSAIC using LaTeX. By using that, the documented equations in MOSAIC have the same readability as documentation level. Therefore, modelling of equations can be done even without knowing any programming languages.

In MOSAIC, modelling is done by creating objects separately. These objects include notation object, equation objects, function objects, equation system object, evaluation object and parameter object. Each objects can be reused by another models. Thus, it will reduce the effort of modelling and also allow reuse of model elements effectively.

The next important features of MOSAIC is code generation to other modelling environments i.e. Matlab, gPROMS etc. This is particularly usefull especially for soving complex mathematical equations. MOSAIC itself can be a code generator and solver. However, MOSAIC is not designed to be a full solver as complicated models cannot solved directly. Therefore, with the code generation function in MOSAIC, allow the model to be solved in in other modelling environments. In addition to that, MOSAIC also has the feature of centralized cooperation on internet. It allows users to share their models easily. The shared model can be read or write by another user on internet.

One of the most studied chemical process is reactive distillation (RD). The equation oriented modelling of RD has been done by many researchers by using different modelling environments, these include Matlab, FORTRAN and so on. Modelling of complex models using these software will be time consuming process and it requires a lot of programming efforts especially to troubleshoot syntax error. Besides that, using these software users often experience the problem of readability since it is written based on its own programming language. Hence, it tends to cause modelling of chemical processes to be difficult and frustrating. Other tools for the solution of models will be sequential based approach such as AspenPlus. Although the simulation can be done easily, it lacks in transparency of equations systems involved and additional programming software such as Aspen Custom modeler (ACM) will needed to build customize equation oriented model.

To overcome some of the above mention modelling limitations, MOSAIC could provide a good alternative. Therefore, the objective of this paper is twofold. First is to explore the modelling of RD for MTBE production by using MOSAIC. Second is to compare the advantages and disadvantages of MOSAIC with other similar software. This could help in deciding to use MOSAIC for any modelling work.

II. MATHEMATICAL MODELLING OF RD

Reactive distillation (RD) technology has brought many advantages in chemical production, these include heat

integration, elimination of azeotropes, 100% conversion of reactants with suppression of side products and lower capital cost. Although not all the chemical manufacturing are suitable by using RD, there are still a lot opportunity in application of RD technology in chemical industries. The production of MTBE is one of chemical processes can be fully benefited from RD technology.

The first step of RD modelling begins with mathematical modelling by formulation of equations to describe phenomenon occurring at the steady state. These formulated equations namely material balance equations, phase equilibrium equations, summation equations and heat balance equations or known as MESH equations will be required in finding solution of the model.

A. MESH Equations

There are several models in designing of RD among others include steady-state equilibrium (EQ) stage model, dynamic EQ stage model, steady-state EQ stage model with stage efficiencies and dynamic EQ stage model with stage efficiencies [2]. In this study, modelling of RD will be based on steady state EQ model. In equilibrium (EQ) stage model, the vapor and liquid phases are assumed to be in thermodynamic equilibrium [3]. The MESH equations used in this work are modified from work of Murat et. al as in [4] and represented as below:-

Overall material balance

$$\frac{dM_j}{dt} = F_j - L_j - V_j + V_{j+1} + L_{j-1} + \delta_j R_j \tag{1}$$

Components material balance

$$\frac{dM_j x_{j,i}}{dt} = F_j z_{j,i} - L_j x_{j,i} - V_j y_{j,i} + V_{j+1} y_{j+1,i} + L_{j-1} x_{j-1,i} + \delta_j \sum_{r=1}^{R} \left(v_{i,r} r_{j,r} \right) \tag{2}$$

Since the model is assumed as steady state, and thus the derivative of material balance will be equal zero. Both *j* and *i* are subscripts to represent the stages numbers and components respectively. *F* represents feed flow rate, *L* represents liquid flow rate then *V* will represent vapor flow rate. *x* and *y* are the mole fraction of liquid and vapour respectively. Furthermore, for integrated reactive part of equation, *r* represents the reaction rate and *v* represents the stoichiometry of chemical components. The value of δ will be either *0* or *1* to decide whether reaction occurring at the stages or not.

Phase equilibrium equations

$$y = \frac{\gamma P^o x}{\phi P} \tag{3}$$

Phase equilibrium relation equation describes the relationship between liquid mole fraction and vapor mole fraction of chemical components when vapour and liquid at equilibrium state. For the ideal condition, the value of activity coefficient, γ and fugacity, ϕ will be equal to 1. Saturated vapor pressure, P_o can be calculated using of Antoine equation and P is pressure of reactive distillation column.

Summation equations

$$\sum_{i}^{c} x_{j,i} = 1.0 \qquad \text{(Liquid phase)} \tag{4}$$

$$\sum_{i}^{c} y_{j,i} = 1.0 \qquad \text{(Vapor phase)} \tag{5}$$

The summation equation states that sum of mole fraction of each components in liquid phase and vapor phase of each stages will be equal to 1.

Enthalpy balance equations

The energy balance of each stages of RD can be described by equation as follow

$$\frac{dH_j}{dt} = F_j H^F + L_{j-1} H^L_{j-1} + V_{j+1} H^V_{j+1} - V_j H^V_j - \delta_j \sum_{r=1}^{R} \left(\Delta H^R_{j,r} \right) r_{j,r} \tag{6}$$

Again, for the steady state, the derivative of energy balance will be equal to zero. For the reactive part of the equation, ΔH is enthalpy change of chemical reaction, *r* is the rate of reaction and δ will decide whether the reaction taking place at each stages or not. For RD modelling of MTBE production, it is assume only a single reaction, and thus the summation of enthalpy change can be ignored.

B. Kinetics Model of MTBE Production

For the production of MTBE, the kinetics model is obtained from work of Rehfinger and Hoffmann as in [5]. The activity based rate model is shown as follow:

$$r_j = Wqk_f \left(\frac{a_i^{IB}}{a_i^{MeOH}} - \frac{a_i^{MTBE}}{K_{eq} a_i^{2MeOH}} \right) \tag{7}$$

Where,

$a_i = \gamma_i x_i$

$k_f = 3.67 \times 10^{12} \exp(-\frac{11110}{T})$

$K_{eq} = 284 \exp[f(T)]$

$$f(T)=A_1\left(\frac{1}{T}-\frac{1}{T_0}\right)+A_2\ln\left(\frac{T}{T_0}\right)+A_3(T-T_0)+A_4(T^2-T_0^2)$$
$$+A_5(T^3-T_0^3)+A_6(T^4-T_0^4)$$

T0	=		298.15 K
A1	=	-	1.49277×103 K
A2	=	-	77.4002
A3	=		0.507563 K-1
A4	=		9.12739×10-4 K-2
A5	=		1.10649×10-6 K-3
A6	=	-	6.27996×10-10 K-4

From equation (7), *W* is weight of catalyst of each stages. *q* is the amount of acid groups on the resin per unit mass with value of 4.9 equiv/kg. The activity of each components is the product of its activity coefficient and liquid mole fraction. k_f is reaction rate constant and K_{eq} is equilibrium constant.

C. RD Process Conditions for MTBE

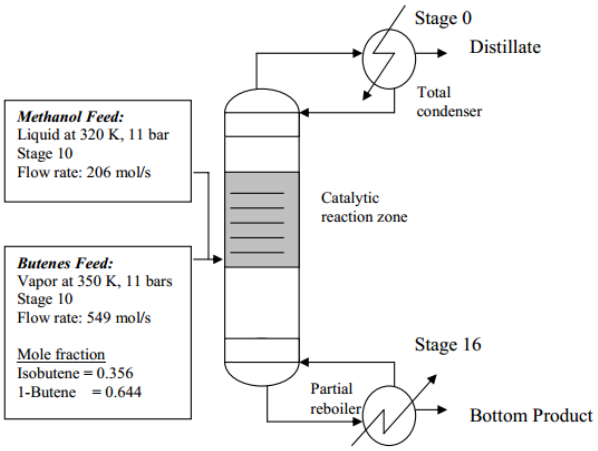

Fig. 1. Column Configuration and Feed Specifications for MTBE synthesis [4]

Figure 1 shows the RD column configuration and feed specifications. The production rate of MTBE assumed to be 197 mol/s or 500,000 metric tons/year. 6400 kg of catalyst is used for 8 reactive trays with amount of 800 kg catalyst each reactive tray. The reactive trays are located in the middle of column from the stage 3 until stage 10. There are in total 17 stages with condenser as stage 0 and partial reboiler as stage 16. The operating pressure is 11 bar or 10.87 atm. The reflux ratio is set to 7. There are two feed streams, methanol feed stream and mixed butenes feed stream with a small stoichiometric excess of methanol [6].

In the modelling work, it is assumed that MTBE synthesis system only consists of main components, namely methanol, isobutylene, MTBE, and n-butene. However, in the actual situation, a minimum of 12 components is involve in the MTBE synthesis. For model simplification, the inert C4 components is assumed as pseudocomponent which is represented by n-butene since they have quite similar physical and chemical properties. The small quantities of by-products can be regarded as negligible.

III. RD MODELLING IN MOSAIC

The process of RD modelling using MOSAIC can be summarized as shown in Figure 2. Overall there are 7 steps. Details explanation will follow next.

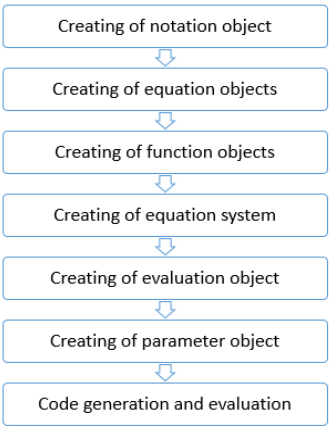

Fig. 2. Procedure of modelling using MOSAIC

Step 1: Creating of notation

In this first step, the symbols with description are created in notation to represent the variables of equations. Each variables can be represented by base name alone or together with subscripts and superscripts. By introducing subscripts and superscripts, it allows two or more variables to have similar base name. Besides the creating of symbols for each variables, the indices required for modelling are created as well in notation.

Step 2: Creating of equation objects

Next step involve creating the equations. The modelling equations can created by using of LaTex. LaTex is a documentary language which allows equations to be expressed in documentation level.

Step 3: Creating of functions objects

Step 3 is almost the same as creating of equation objects, in which function objects are created by using LaTeX as well. However, the method of creating functions are not as simple as creating equations. It may require creating parameter list object if parameter set index in involve. Parameter set index allows users to set the index on output variable and parameters. The specification of output variable and input variables will be required. The output variable will be the variable that is calculated while the input variables are almost similar as design variables where the setting of its values will be required. The formula which leads to output variable will be written in LaTex.

Step 4: Creating of equation system

All equations and functions of a model are connected by equation system for evaluation. Adding of equations to equation system can be done easily, but for adding functions, it is required to set the output variables and input variables. Preview of all the added equations and functions in equation system can thus be made.

Step 5: Creating of evaluation object

In order to create the evaluation object, the equation system first has to be loaded. Once it is loaded, indexing can be made by specifying the max value of each indexes. After indexing, all the equations and functions involved in modelling can be displayed.

Step 6: Creating of parameter object

Specifications of variables are now ready to be made by assigning the variables as iteration variables or design variables. The degree of freedom will be automatically calculated when assigning of variables. In order to solve the model, degree of freedom must be zero. Once it is achieve, the value of each design variables is given for calculation. Furthermore, good initial values of iteration variables are important in solving the models.

Step 7: Code generation and evaluation

Since MOSAIC is not designed to be full solver, it is not able to solve complex models. Hence, the solving of model can be made by code generation to other modelling environments i.e. Matlab, ACM, gPROMS, C++ etc. The generated codes can be run at their own corresponding environment for the solving of model.

IV. Results and Discussion

A. Model Validation and Comparison

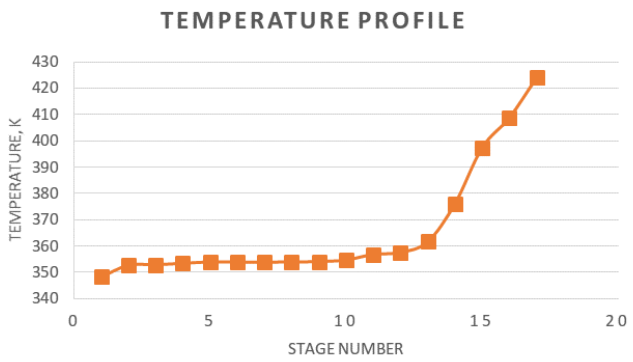

Fig. 3. Temperature versus Stage number

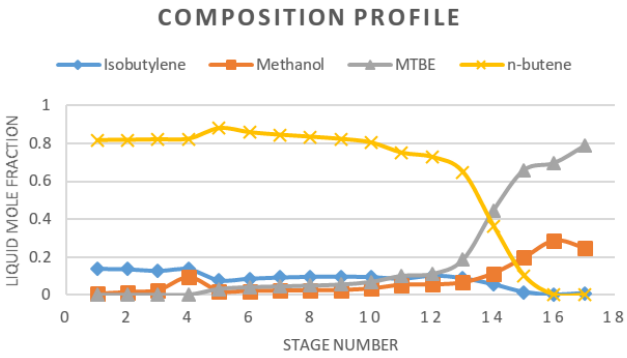

Fig. 4. Liquid mole fraction of each components versus Stage number

The temperature profile of the modeled RD is shown in Figure 3, it can be seen that temperature is maintained at around 353 K from condenser to feed stage. After the feed stage, the temperature starts to increase rapidly until it achieves the temperature of 424 K at the reboiler. The temperature of each stage is dependent on the component composition. Since the relative high mole fraction of n-butene from condenser stage to feed stage, the temperature is maintain at around boiling point of pure n-butene (348K). After the feed stages, the mole fraction of MTBE increases rapidly until it reaches nearly 80% purity of MTBE at reboiler stages. Thus, it causes the temperature of each stages to increase instantly until it hits the highest temperature of 424K which is nearly boiling point of pure MTBE. For composition profile (see Figure 4), the system nearly reach 80% purity of n-butene and MTBE at condenser stage and reboiler stage respectively. There is only a significant drop of n-butene mole fraction only after feed stage. Other than that, the mole fraction of MTBE increase at significant amount after the feed stage. From the table I, it can be seen that results of proposed model are still quite satisfactory compared to Nijhuist's. Note that is this work it is assumed the liquid and vapor phase behave ideally.

TABLE I. Comparison of Results between Proposed Model and Nijhuist's Model

		Nijhuist's model		Proposed model	
Quantity	Units	Top	Bottom	Top	Bottom
x_{IB}		0.01	0.00	0.14	0.01
x_{MeOH}		0.04	0.00	0.00	0.25
x_{MTBE}		0.00	0.98	0.00	0.79
$x_{n\text{-butene}}$		0.95	0.02	0.82	0.00
Temperature	K	348	424	348	424
Product flow	mol/s	366	197	340	254
Heat duty	MW	49.7	35.1	45.6	35.1
MTBE purity	%	98		79	
Reflux flow	kmol/s	2.60		2.38	

B. Software comparison

Generally modelling tools for modelling of chemical processes can be categorized into two groups. The first group consists of readily made models with preprogramed equation

systems and an appropriate numerical solution algorithm. Also known as sequential modelling approach, Aspen Plus, Aspen HYSY and CHEMCAD, are some example of software belong to this group. Using these software modelling of chemical processes can be done easily, however it lacks in transparency of equations involved. Another software may be needed to build the equations model before embedding it into those software. One such example is Aspen custom modeler (ACM).

The second group or the equation oriented modelling provide modelling environment based on their own programming languages. The users are free to define their own equation systems by writing their own code using specifically define programming language. Moreover, these tools are suitable for the creation of customized models. Some examples of these tools are gProms, Aspen Custom Modeler (ACM), GAMS and Matlab [1].

TABLE II. COMPARISON OF MOSAIC WITH TWO GROUPS OF MODELLING TOOLS

	MOSAIC	EO	SMA	Notes
Modelling at documentation level	Available	-	-	Modelling by using MOSAIC can be done at documentation level. Since the equations are written in Latex documentary language, it has similar readability as documentation level. In addition, coding is not required for modelling using MOSAIC.
Reuse of model elements	Available	Partially available	-	The reuse of model elements by MOSAIC is more systematic and organized. The users can either reuse the notation, equations, functions and variables settings easily.
Code generation	Available	Not complete	-	Code generation of MOSAIC are much more complete. MOSAIC allows users to generate the code and run their model in many different environments.
Centralized internet database capability	Available	-	-	MOSAIC allows users to share their works on internet more easily. The shared work can be viewed or write by the person that users shared to.
Degree of freedom analysis	Available	-	-	Degree of freedom analysis can be done automatically by MOSAIC. In order to generate code for evaluation, degree of freedom must be zero. Hence, it is necessary for the model to have correct equations in equation system and all the errors must be corrected.
Error identification	Excellent	Difficult	-	Since modelling by MOSAIC is written in Latex documentary language, it has high readability. Hence, users can identify the errors more easily if they are making mistakes while key in modelling equations. In addition, coding is not required by MOSAIC, hence it results in less errors and less effort required.
Language	Documentary	Programing	-	MOSAIC is using Latex documentary language in writing of modelling equations. Latex has high readability, unlike programming language, it is difficult to read and understand the coding.
Readability	Excellent	Poor	-	Latex documentary language has much higher readability than programming language.
Difficulty	Medium	Hard	Easy	Although without knowledge of coding in modelling, MOSAIC still allows users to do modelling without a doubt. The stages of modelling using MOSAIC are creating of notation, creating of equations, creating of functions, creating of equation system, creating of parameter object and creating of evaluation object. The most difficult part will be creating of functions, however it can overcome easily once users know how it works.
Transparency of model	Excellent	Excellent	Poor	All equations involved in modelling are known for MOSAIC. Besides that, given value of variables also can be determined.
Building of customized model	Capable	Capable	Not capable	MOSAIC is similar to software like Matlab which is designed to build the customized model.

Table 2 shows the comparison between MOSAIC, EO (equation oriented programming) and SMA (sequential modelling approach). This table can be use a guideline for choosing MOSAIC for any modelling and simulation works.

V. CONCLUDING REMARKS

Modelling by using MOSAIC can be done even without knowing of any modelling programming languages. MOSAIC uses the LaTeX documentary language in documentation of all modelling equations. LaTeX is much easier to learn than programming languages. Modelling equations created using LaTeX almost have the same readability as documentation level. This is useful for error identification and it makes the modelling at documentation level to be possible. Besides that, reuse of model elements can be done effectively by using MOSAIC. Since modelling by using MOSAIC is done by creating of objects separately, each objects can be reused by another model. Different models with different settings can be created easily. Thus, it is time saving and reduce the efforts of modelling. In order to obtain accurate results, it is important to ensure all the errors and mistakes during modelling are corrected. MOSAIC definitely can help to identify the errors and mistakes more effectively. In addition, good initial values of iteration variables are important for solving of the model.

This work is sponsored by Malaysia Ministry of Education under grant no. RDU130601.

Acknowledgment

This work is supported by Malaysia Ministry of Education under grant no. RDU130601. Special thanks to Markus Illner for his kind assistance and advice.

References

[1] S. Kuntsche, T. Barz, R. Kraus, R., H. Arellano-Garcia & G. Wozny, "MOSAIC a web-based modeling environment for code generation". Computers & Chemical Engineering, 2011, 35(11), 2257-2273.

[2] R. Baur, "Modeling reactive distillation dynamics". 2000, Clarkson University.

[3] R. Baur, A. Higler, A, R. Taylor & R. Krishna, "Comparison of equilibrium stage and nonequilibrium stage models for reactive distillation". Chemical Engineering Journal, 2000, 76(1), 33-47.

[4] M.N. Murat, A.R. Mohamed & S. Bhatia, "Modeling of a reactive distillation column: methyl tertiary butyl ether (MTBE) simulation studies". IIUM Engineering Journal, 2003, 4(2).

[5] A. Rehfinger & U. Hoffmann, "Kinetics of methyl tertiary butyl ether liquid phase synthesis catalyzed by ion exchange resin—I. Intrinsic rate expression in liquid phase activities". Chemical Engineering Science, 1990, 45(6), 1605-1617.

[6] S. Nijhuis, F. Kerkhof & A. Mak, "Multiple steady states during reactive distillation of methyl tert-butyl ether". Industrial & engineering chemistry research, 1993, 32(11), 2767-2774.