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MODELLING, SIMULATION AND OPTIMIZATION OF A REACTIVE DISTILLATION PROCESS USING MINITAB AND MATRIX LABORATORY

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ABSTRACT

Reactive distillation is a novel process that combines both chemical reaction and separation in a single piece of equipment. It is normally accomplished inside a column. Actually, the process has a lot of benefits, especially for those reactions occurring at temperatures and pressures suitable for the distillation of the resulting components. However, the combination of both reaction and separation in a single unit has made the modelling of the process a bit challenging. It has been deemed necessary to employ a mathematical method, with the aid of Minitab, to handle the modelling of this process in an effective manner. Therefore, in this research work, the modelling knowledge of mathematics has been employed to develop equations for the different phenomena occurring at some specific sections of a reactive distillation column. The developed models were simulated and, further, optimized using Matrix Laboratory in order to obtain the values of the model parameters required to give the desired mole fractions of the product components of the process. The results obtained revealed that the developed models were good representatives of the top and the bottom sections of the column used because there were good correlations between the measured and the simulated mole fractions as the R-squared values of the top and the bottom section models were estimated to be 99.32% and 99.03% respectively. Furthermore, the optimization carried out revealed that multiobjective problem formulation was the best way of handling this type of a system because that was the one that gave the desired optimum values of the two products from their respective sections of the column.

Keywords: reactive distillation, modelling, simulation, optimization, Minitab, matrix laboratory.

1. INTRODUCTION

In recent years, reactive separation processes have attracted considerable attentions in academic research and industrial applications, (Völker *et al.*, 2007; Giwa and Karacan, 2012b). One of these processes, which is known as "reactive distillation", is potentially attractive whenever conversion is limited by reaction equilibrium (Balasubramhanya and Doyle III, 2000; Giwa and Karacan, 2012b).

Reactive distillation is defined as a process that has the capability of combining both separation and chemical reaction in one equipment unit (Giwa, 2012; Giwa, 2013; Giwa *et al.*, 2013a; Giwa *et al.*, 2013b; Giwa, 2016). A lot of advantages are associated with it, especially, for those reactions occurring at temperatures and pressures suitable for the distillation of the components involved in the reaction (Sneesby *et al.*, 1997; Giwa and Karacan, 2012b; Giwa and Karacan, 2012d; Giwa and Karacan, 2012e; Giwa and Giwa, 2013a; Giwa *et al.*, 2013c; Giwa and Giwa, 2013b; Giwa, 2014; Giwa *et al.*, 2014; Giwa *et al.*, 2015a; Giwa *et al.*, 2015b; Giwa and Giwa, 2016). Furthermore, this process combines the benefits of equilibrium reaction with a unit operation (known as distillation) to achieve a substantial progress in promoting reaction conversion as a result of constant recycling of unconverted materials and removal of products as well as reducing capital and operating costs because of the reduction of number of equipment units of the plant (Giwa and Karacan, 2012a; Giwa and Giwa, 2013c; Giwa *et al.*, 2014; Giwa and Giwa, 2016). Basically, the combination of reaction and distillation has several advantages which include increase of reaction conversion by simultaneous reaction and separation of products, suppression of side reaction(s) and utilization of heat evolved from an exothermic reaction for mass transfer operation. These synergistic effects of this process result in significant economic benefits (lower capital investment, lower energy cost and higher product yields) of reactive distillation compared to a conventional design having a reactor followed by a number of distillation columns (Moritz and Hasse, 1999; Giwa and Karacan, 2012c; Giwa and Giwa, 2016). Owing to the combination of reaction and separation in a single unit, effective handing of this process, for instance, in the areas of modelling and optimization, is still a challenge to process engineers because many complexities are involved in its operation.

Optimization can be defined as a way of obtaining some specified sets of input parameters that give a maximum or a minimum value of an objective functions, which may be subject to some constraints. The most common objective functions are minimizing cost of production and maximizing throughput. In carrying out optimization of a process, the goal(s) can be set to maximizing one or more of the process specifications, while keeping all others within some constraints.

According to the information gathered from the literature, some researchers have carried out some investigations on optimization of reactive distillation processes. For instance, Cardoso *et al.* (2000) applied a simulated annealing-based algorithm suitable for the optimization of mixed integer non-linear programming problems to the synthesis of a non-equilibrium reactive distillation column. A simulation model based on an extension of conventional distillation was proposed for the simulation step of the optimization problem. In the case of ideal vapour-liquid equilibrium, the simulation results were found similar to those obtained by Ciric and Gu (1994) using GAMS environment and to those obtained with Aspen Plus modular simulator. The optimization results were also obtained to be similar to those obtained using an adaptive random search algorithm (MSGA). The optimizations of the work were also performed with non-ideal vapour-liquid equilibrium, considering distributed feed and reaction trays and the results showed that the optimized objective function values were very similar and mostly independent of the number of trays and of the reaction distribution. It was also discovered that the proposed simulation/optimization equation-oriented environments were capable of providing optimized solutions which were close to the global optimum, and that revealed the adequacy of the algorithm for the optimization of reactive distillation problems encountered in chemical engineering practice. Phuenduang *et al.* (2011) carried out the optimization of biodiesel production from jatropha oil using reactive distillation process by employing response surface methodology (RSM) based on central composite design (CCD) in which methanol flowrate and reflux ratio were the independent variables while the yield of biodiesel obtained from the process was the dependent variable. They were able to obtain very high yield of biodiesel as the optimum value of the output parameter. Giwa and Giwa (2012) used Design Expert and Excel Solver to find the optimum values that could be used to obtain high mole fractions of n-butyl acetate and methanol at the bottom segment and the top segment of a reactive distillation column respectively using Aspen HYSYS model data generated for the process. The achievements of the approximate optimum values of the objective functions given by the optimization carried out using the Excel Solver when the optimum values of reflux ratio and reboiler duty were used to run the experimental simulations showed that the obtained optimum values were valid ones. Giwa and Giwa (2013a) applied response surface methodology in conjunction with Matrix Laboratory (MATLAB) to optimize the reactive distillation esterification process used for the production of isopropyl myristate through the reaction between myristic acid and isopropanol by taking the mole fraction of isopropyl myristate obtained from the column as the objective function and reflux ratio, feed ratio and reboiler duty as the input variables. The optimization results obtained showed that the theoretical optimum values obtained with the aid of *fsolve* command of MATLAB were valid because the experimental simulation with these values gave the bottom isopropyl myristate mole fraction that compared very well with the theoretical simulation value of bottom isopropyl myristate mole fraction. Sakhre *et al.* (2014) used gravitational search algorithm (GSA), which is a heuristic optimization technique to obtain the optimum values for the operation of a reactive distillation process used for producing methyl *tert*-butyl ether. They applied feed flow rates as a test function along with universal bench mark unimodel test function to obtain optimized process. The GSA code was generated in MATLAB. It was found from the work that the performance of GSA was able to achieve good results regarding the quality and success rate in finding optimal solution. Edreder *et al.* (2015) used the model equations in terms of mass and energy balances and thermodynamic properties within gPROMS modelling software to optimize the operation of a batch reactive distillation process involving an esterification reaction between acetic acid with methanol to produce methyl acetate and water. Two case studies with varying amount of the reactants were considered in the work. The reflux ratio was selected as the manipulated variable of the optimization with different but fixed batch time ranging from 5 to 15 h in order to maximise the conversion of methanol subject to methyl acetate purity obtained as the product. The dynamic optimisation problem of the work was converted to a nonlinear programming problem by control vector parameterization (CVP) technique and solved using efficient sequential quadratic programming (SQP) method. From the literature review that was carried out, it was noticed that the work of Giwa and Giwa (2012) optimized the mole fractions of methanol and butyl acetate separately as two single-input multi-output objective functions to obtain the optimum values of the process using the *fsolve* command of MATLAB. However, it was felt that this optimization could be carried out better by considering the two objective functions simultaneously. Therefore, this work has been carried out to optimize the process (a reactive distillation process used for the production of methanol and butyl acetate from the transesterification reaction between methyl acetate and butanol) by considering the two objective functions of the process at the same time with the aid of *fsolve* and *fminimax* commands of MATLAB.

2. PROCEDURE

2.1 Prototype plant development

The prototype of the process developed with the aid of Aspen HYSYS (Aspen, 2012) from which the data used to develop the models of the reactive distillation process that were optimized was as shown in Figure-1. It (the prototype) was developed with UNIversal QUAsi Chemical (UNIQUAC) model as the fluid package using Distillation Column Sub-Flow sheet having two feed streams (upper and lower). The heavy feed of the process, which was 1-butanol (see Table-1 showing the basic properties of the components), was passed through the upper feed stream while the light one (methyl acetate) was fed into the column from the lower feed stream. The two feeds were passed into the column at the same temperature

and pressure of 25 °C and 1 atm respectively. The column of the prototype plant was divided into different sections, namely condenser section, rectifying section, reaction section, stripping section and reboiler section. The reaction section was between stages 7th and 14th inclusive and the rectifying and the stripping sections were above and below it, respectively. In this work, the pressures of the condenser and the reboiler sections were both made to be 1 atm.

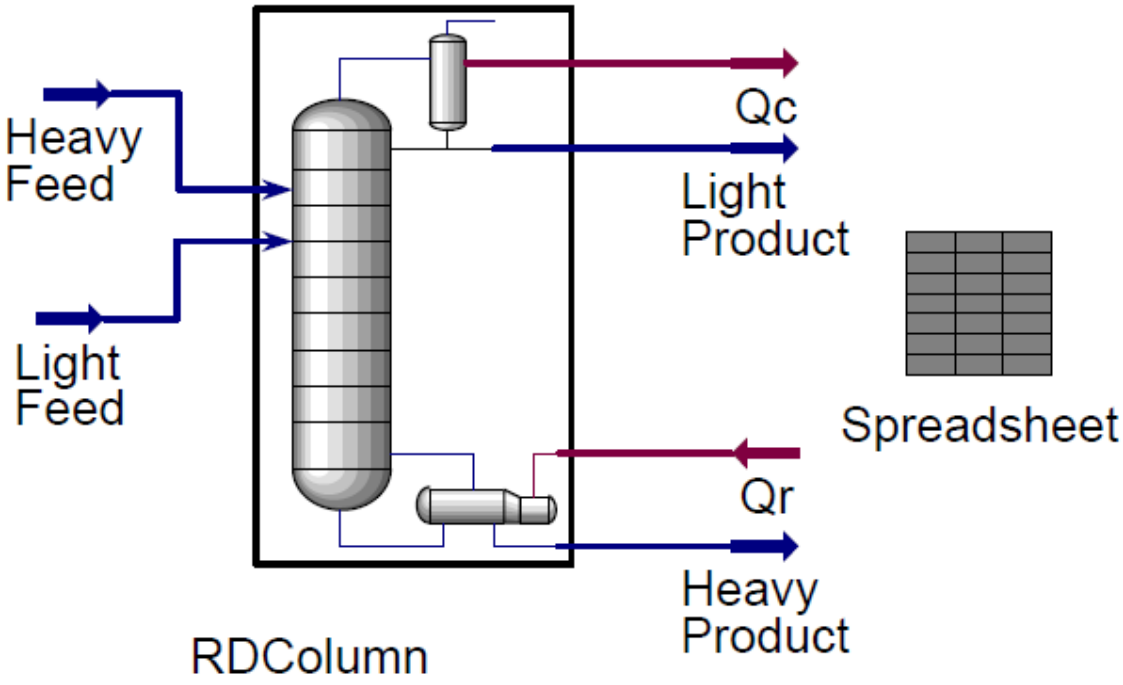

Figure-1. Developed Aspen HYSYS prototype plant of the reactive distillation process.

The reaction of the process is a transesterification type occurring, between butanol and methyl acetate to produce methanol and butyl acetate in the reaction section of the column. The stoichiometry of the reversible reaction is given as shown in Equation 1.

Table-1. Basic properties of the components.

Component	**Molecular weight** (kg/kgmol)	**Boiling point** (°C)	**Density** (kg/m^3)
Butanol	74.12	117.75	813.87
Methyl acetate	74.08	57.25	939.33
Methanol	32.04	64.65	795.72
Butyl acetate	116.16	126.15	885.84

Source: Aspen, 2012

$$\underset{(1)}{C_4H_9OH} + \underset{(2)}{CH_3COOCH_3} \xleftrightarrow{K_{eq}} \underset{(3)}{CH_3OH} + \underset{(4)}{CH_3COOC_4H_9} \quad (1)$$

Furthermore, using the numbering notation below the components, the reaction rate equation of the process is given as shown in Equation (2) (Wang *et al.*, 2008).

$$r = 2.018\times10^8\ exp(71960/RT)C_1C_2 - 2.839\times10^8\ exp(72670/RT)C_3C_4 \quad (2)$$

2.2 Experimental design

After the development of the prototype plant of the process, Minitab17 (Minitab, 2013) was used to design a set of custom experiments (given in Table-2) based on the four factors (independent variables) considered in this work. Those independent variables were reflux ratio (x_1), butanol volumetric flow rate (x_2), methyl acetate volumetric flow rate (x_3) and reboiler duty (x_4).

Table-2. Designed experiments for the reactive distillation process.

Run	x_1	x_2	x_3	x_4
1	3	25	35	0.95
2	5	25	25	0.95
3	3	25	25	0.25
4	3	35	25	0.95
5	3	25	25	0.95
6	3	25	25	0.95
7	3	25	15	0.95
8	4	30	20	0.6
9	2	30	30	1.3
10	4	30	30	0.6
11	4	20	30	1.3
12	4	30	20	1.3
13	4	20	30	0.6
14	2	20	30	0.6
15	4	20	20	0.6
16	2	20	20	0.6
17	2	30	30	0.6
18	3	25	25	0.95
19	3	25	25	0.95
20	3	25	25	0.95
21	2	30	20	0.6
22	4	30	30	1.3
23	3	25	25	0.95

In order to run the developed Aspen HYSYS plant using the values of the independent variables given in Table-2, Sparse Continuation Solver was employed because it was the one found suitable for a process of this nature.

2.3 Model development and optimization

After running the prototype plant using the experimental operating values generated with the aid of Minitab, the responses obtained were entered into the program (Minitab) and the experimental results were analysed to obtain model equations as well as carry out the analyses of variance for the phenomena occurring at the top and the bottom sections of the reactive distillation column in form of quadratic equations. Furthermore, the process was optimized using the developed model equations with the aid of Matrix Laboratory (MATLAB) (Mathworks, 2015) using *fsolve* (with Levenberg-Marquardt) algorithm and *fminimax*.

3. RESULTS AND DISCUSSIONS

The outputs obtained from the running of the prototype plant of the reactive distillation process using the input data generated with the aid of Minitab are given together with the input data in Table-3. As can be seen from the table, the mole fractions of the desired products of the process, which were methanol and butyl acetate obtained from the top and the bottom sections of the column respectively, were changing with changes in the values of the different input variables considered. The changes observed were indications that the input variables were actually having effects on the process.

Table-3. Experimental results obtained from the prototype plant of the reactive distillation process.

Run	x_1	x_2	x_3	x_4	x_{MeOH}	x_{BtAc}
1	3	25	35	0.95	0.39	0.50
2	5	25	25	0.95	0.36	0.42
3	3	25	25	0.25	0.87	0.30
4	3	35	25	0.95	0.85	0.72
5	3	25	25	0.95	0.78	0.99
6	3	25	25	0.95	0.77	0.99
7	3	25	15	0.95	0.53	0.84
8	4	30	20	0.6	0.56	0.37
9	2	30	30	1.3	0.33	1.00
10	4	30	30	0.6	0.54	0.34
11	4	20	30	1.3	0.51	1.00
12	4	30	20	1.3	0.61	0.81
13	4	20	30	0.6	0.38	0.33
14	2	20	30	0.6	0.52	0.55
15	4	20	20	0.6	0.38	0.40
16	2	20	20	0.6	0.77	0.99
17	2	30	30	0.6	0.68	0.48
18	3	25	25	0.95	0.78	0.99
19	3	25	25	0.95	0.78	0.99
20	3	25	25	0.95	0.78	0.99
21	2	30	20	0.6	0.88	0.68
22	4	30	30	1.3	0.82	0.96
23	3	25	25	0.95	0.78	0.99

Using the input and output data presented in Table-3, Equation 3 which relates the input variables with top methanol mole fraction and Equation 4 that gives mathematical relationship between the second response, bottom butyl acetate mole fraction and input variable of the process, were developed. Given in Tables 4 and 5 are the results of the analyses of variance carried out on the developed models.

$$x_{MeOH} = -0.439 + 0.0270x_1 + 0.0693x_2 + 0.0957x_3 - 2.095x_4 - 0.12150x_1^2 - 0.001532x_2^2 + \cdots$$
$$\cdots - 0.003326x_3^2 - 0.2035x_4^2 + 0.00139x_1x_2 + 0.01164x_1x_3 + 0.4225x_1x_4 + \cdots$$
$$\cdots + 0.000357x_2x_3 + 0.01540x_2x_4 + 0.02296x_3x_4 \quad (3)$$

$$x_{BtAc} = 0.890 - 0.377x_1 - 0.0227x_2 + 0.0436x_3 + 1.355x_4 - 0.0631x_1^2 - 0.000887x_2^2 + \cdots$$
$$\cdots - 0.003092x_3^2 - 0.1828x_4^2 + 0.00903x_1x_2 + 0.01457x_1x_3 + 0.0115x_1x_4 + \cdots$$
$$\cdots + 0.001648x_2x_3 - 0.01921x_2x_4 + 0.01103x_3x_4 \quad (4)$$

As shown in Table-4, considering 95% confidence level, all the terms of x_{MeOH} model were found to be significant, because the probability value of each of them was less than 5% except those of x_1x_2 and x_2x_3 which were greater than 0.05. Based on this, the model was found necessary for modification so that the terms having probability levels greater than 5% could be removed from it.

Table-4. Analysis of variance of the developed top section model.

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Model	15	0.745274	0.049685	87.51	0.000
Blocks	1	0.001603	0.001603	2.82	0.137
Linear	4	0.095911	0.023978	42.23	0.000
x_1	1	0.003298	0.003298	5.81	0.047
x_2	1	0.054982	0.054982	96.84	0.000
x_3	1	0.007729	0.007729	13.61	0.008
x_4	1	0.029638	0.029638	52.20	0.000
Square	4	0.333876	0.083469	147.01	0.000
x_1^2	1	0.150189	0.150189	264.52	0.000
x_2^2	1	0.014930	0.014930	26.30	0.001
x_3^2	1	0.172109	0.172109	303.13	0.000
x_4^2	1	0.005258	0.005258	9.26	0.019
2-Way Interaction	6	0.253770	0.042295	74.49	0.000
x_1x_2	1	0.000389	0.000389	0.68	0.435
x_1x_3	1	0.029796	0.029796	52.48	0.000
x_1x_4	1	0.113724	0.113724	200.30	0.000
x_2x_3	1	0.000699	0.000699	1.23	0.304
x_2x_4	1	0.003776	0.003776	6.65	0.037
x_3x_4	1	0.010933	0.010933	19.26	0.003
Error	7	0.003974	0.000568		
Lack-of-Fit	3	0.003974	0.001325	18107.81	0.000
Pure Error	4	0.000000	0.000000		
Total	22	0.749248			

Table-5. Analysis of variance of the developed bottom section model.

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Model	15	1.66728	0.111152	108.58	0.000
Blocks	1	0.00078	0.000779	0.76	0.412
Linear	4	0.34879	0.087196	85.18	0.000
x_1	1	0.12590	0.125903	122.99	0.000
x_2	1	0.03828	0.038276	37.39	0.000
x_3	1	0.07601	0.076008	74.25	0.000
x_4	1	0.31937	0.319366	311.99	0.000
Square	4	0.19319	0.048297	47.18	0.000
x_1^2	1	0.04054	0.040536	39.60	0.000
x_2^2	1	0.00501	0.005008	4.89	0.063
x_3^2	1	0.14868	0.148684	145.25	0.000
x_4^2	1	0.00424	0.004244	4.15	0.081
2-Way Interaction	6	0.12580	0.020966	20.48	0.000
x_1x_2	1	0.01643	0.016431	16.05	0.005
x_1x_3	1	0.04670	0.046695	45.62	0.000
x_1x_4	1	0.00008	0.000085	0.08	0.782
x_2x_3	1	0.01494	0.014939	14.59	0.007
x_2x_4	1	0.00588	0.005877	5.74	0.048
x_3x_4	1	0.00252	0.002523	2.46	0.160
Error	7	0.00717	0.001024		
Lack-of-Fit	3	0.00717	0.002389	432063.84	0.000
Pure Error	4	0.00000	0.000000		
Total	2	1.67445			

Given in Table-5 are the outputs obtained from the analysis of variance done for the model equation developed for the bottom section of the column; that is, the model equation relating the bottom butyl acetate to the input variables of the process. From the information obtained from the table, it was discovered that, apart from x_2^2, x_2^2, x_1x_4 and x_3x_4 that had their probability values to be greater than 5%, all other terms of the model were found to be significant because the probability value of each of them was less than 0.05. This was found to be an indication that this model too needed to be modified so that all its terms could have probability values that would be less than 0.05.

Further shown in Equation (5) is the modified version of the top section model after the terms having probability values greater than 0.05 have been removed. The modified model was also analysed to be sure that all the terms would have probability values less than 5%. From the analysis of variance results obtained and given in Table-6, it was discovered that the model had been modified well because each of the terms present can now be seen to have a probability value less than 5%.

$$x_{MeOH} = -0.738 + 0.0580x_1 + 0.0864x_2 + 0.1025x_3 - 2.229x_4 - 0.12223x_1^2 - 0.001660x_2^2 + \cdots \\ \cdots - 0.003316x_3^2 - 0.2240x_4^2 + 0.01182x_1x_3 + 0.4285x_1x_4 + 0.01940x_2x_4 + 0.02449x_3x_4 \quad (5)$$

Table-6. Analysis of variance of the modified top section model.

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Model	13	0.744185	0.057245	101.76	0.000
Blocks	1	0.001434	0.001434	2.55	0.145
Linear	4	0.129026	0.032256	57.34	0.000
x_1	1	0.004656	0.004656	8.28	0.018
x_2	1	0.083935	0.083935	149.21	0.000
x_3	1	0.007060	0.007060	12.55	0.006
x_4	1	0.038796	0.038796	68.97	0.000
Square	4	0.344671	0.086168	153.18	0.000
x_1^2	1	0.161315	0.161315	286.77	0.000
x_2^2	1	0.019406	0.019406	34.50	0.000
x_3^2	1	0.171763	0.171763	305.34	0.000
x_4^2	1	0.006762	0.006762	12.02	0.007
2-Way Interaction	4	0.252682	0.063170	112.30	0.000
x_1x_3	1	0.031026	0.031026	55.16	0.000
x_1x_4	1	0.141355	0.141355	251.29	0.000
x_2x_4	1	0.008036	0.008036	14.29	0.004
x_3x_4	1	0.013362	0.013362	23.75	0.001
Error	9	0.005063	0.000563		
Lack-of-Fit	5	0.005062	0.001012	13839.87	0.000
Pure Error	4	0.000000	0.000000		
Total	22	0.749248			

$$x_{BtAc} = 1.481 - 0.400x_1 - 0.0739x_2 + 0.0414x_3 + 1.681x_4 - 0.0616x_1^2 - 0.003078x_3^2 + \cdots \\ \cdots + 0.00987x_1x_2 + 0.01445x_1x_3 + 0.002063x_2x_3 - 0.03058x_2x_4 \quad (6)$$

Given in Equation (6) and Table-7 are the modified model equation for the bottom section and the results of its analysis of variance, respectively. According to results of analysis of variance given in the table, all the terms in the model equation are now having probability values that are less than 5%.

Table-7. Analysis of variance of the modified bottom section model.

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Model	11	1.65828	0.15075	102.59	0.000
Blocks	1	0.00017	0.00017	0.12	0.738
Linear	4	1.20922	0.30230	205.72	0.000
x_1	1	0.26004	0.26004	176.96	0.000
x_2	1	0.11792	0.11792	80.25	0.000
x_3	1	0.14042	0.14042	95.56	0.000
x_4	1	1.11934	1.11934	761.74	0.000
Square	2	0.19195	0.09597	65.31	0.000
x_1^2	1	0.04793	0.04793	32.62	0.000
x_3^2	1	0.14782	0.14782	100.59	0.000
2-Way Interaction	4	0.13633	0.03408	23.19	0.000
x_1x_2	1	0.02483	0.02483	16.90	0.002
x_1x_3	1	0.05469	0.05469	37.22	0.000
x_2x_3	1	0.02773	0.02773	18.87	0.001
x_2x_4	1	0.02205	0.02205	15.01	0.003
Error	11	0.01616	0.00147		
Lack-of-Fit	7	0.01616	0.00231	417708.19	0.000
Pure Error	4	0.00000	0.00000		
Total	22	1.67445			

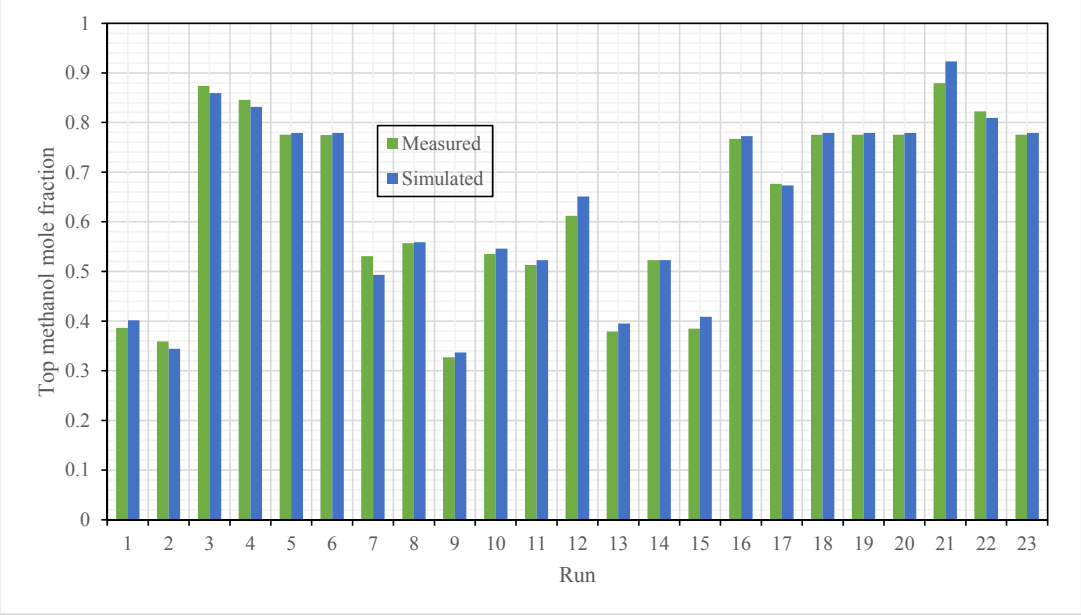

Figure-2. Measured and simulated mole fractions of methanol obtained from the top section of the column.

After obtaining the reliable models for the top and the bottom sections of the column, they were simulated and the results obtained from the simulations were as given in Figures 2 and 3 as plots of the measured and the simulated mole fractions of the products versus the runs.

Figure-2 shows the measured and the simulated mole fractions of methanol obtained from the top section

of the reactive distillation column. As can be seen from the figure, good correlations were found to exist between the results that were plotted side by side in a clustered column chart. To actually ascertain that there were good correlations between the measured and the simulated ones, the R-squared value of the relationship was estimated and obtained to be 99.32%.

Shown in Figure-3 are the results obtained when the simulation of the model equation developed for the bottom section of the column was executed. In this case also, good correlations were found to exist between the simulated mole fractions and the ones that were measured from the prototype of the process as the R-squared value was estimated in this case to be 99.03%.

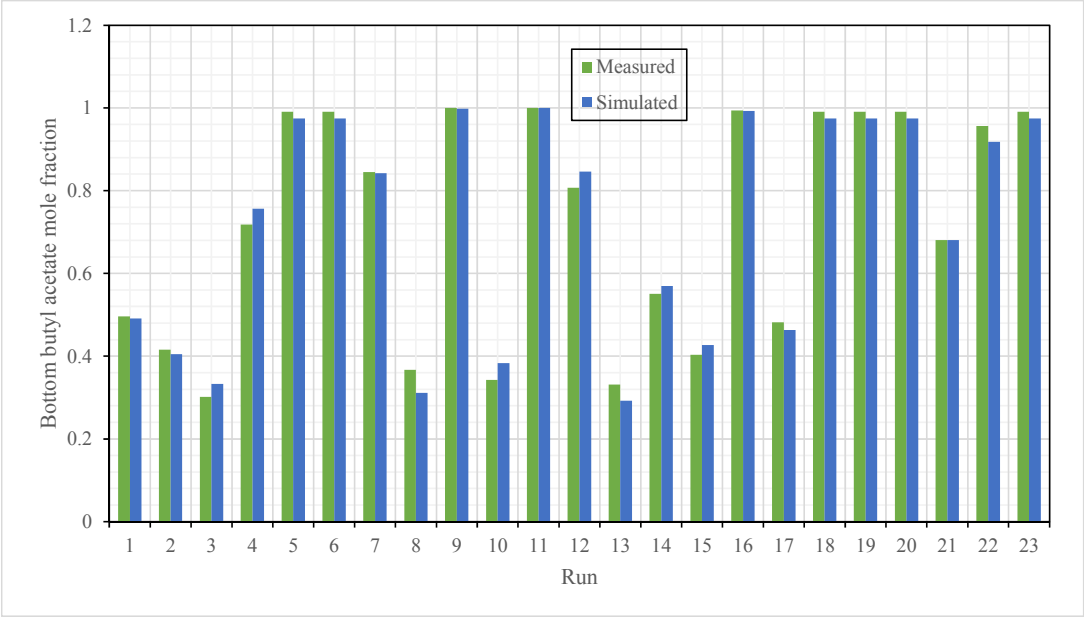

Figure-3. Measured and simulated mole fractions of butyl acetate obtained from the bottom section of the column.

After obtaining and ascertaining that the developed modified model equations for the top and the bottom sections of the reactive distillation column could represent the process very well, it was deemed necessary to optimize it (the process) because it was desired to collect at least 85% purity of each of the products from the top and bottom sections of the column accordingly. Based on that, the optimization of the process was carried out by taking the initial values of the reflux ratio, butanol flow rate, methyl acetate flow rate and reboiler duty to be 1, 20 mL/min, 20 mL/min and 0.5 kJ/s, respectively via codes written in MATLAB environment and the results obtained were as given in Tables 8-10.

The results given in Table-8 were the ones obtained when the objective function of the optimization was taken as the maximization of the mole fraction of methanol given from the top section of the column. According to the table, the objective of the optimization was achieved when *fsolve* was used because a maximum methanol mole fraction of 1.00 but with butyl acetate mole fraction of 0.70 was attained. From the same table, it was noticed that the values of the mole fraction of methanol obtained from the top section of the column when *fminimax* was used as the optimization command in the MATLAB was greater than the maximum value expected which was 1.00. Besides, the value of the other product was very low. The results given in this table revealed that the objectives of the optimization, which were the achievement of at least 85% (0.85) purity of each of the products had not been satisfied.

Table-8. Optimum values obtained when the maximization of top methanol mole fraction was the objective function.

Parameter	Initial value	Optimization using *fsolve*	Optimization using *fminimax*
Reflux ratio	1	1.06	1.29
Butanol flow rate (mL/min)	20	20.01	26.61
Methyl acetate flow rate (mL/min)	20	20.00	18.12
Reboiler duty (kJ/s)	0.5	0.24	0.10
Methanol mole fraction		1.00	1.23
Butyl acetate mole fraction		0.70	0.39

As such, the objective function of the optimization was changed to the maximization of the mole fraction of butyl acetate given from the process through the bottom section of the column, and the results of this

optimization are given in Table-9. From the results shown in Table-9, the optimization carried out using *fsolve* was found to be better than that done with *fminimax* command because when *fsolve* command was used, at least, the objective function was satisfied because a mole fraction of 1.00 was obtained for the butyl acetate product given by the system, even though that of the other product was not up to 0.85 as desired, but that of *fminimax* command was found not to be good at all.

Table-9. Optimum values obtained when the maximization of bottom butyl acetate mole fraction was the objective function.

Parameter	Initial value	Optimization using *fsolve*	Optimization using *fminimax*
Reflux ratio	1	1.00	5814.68
Butanol flow rate (mL/min)	20	20.00	0.10
Methyl acetate flow rate (mL/min)	20	20.00	0.10
Reboiler duty (kJ/s)	0.5	0.51	2392867.49
Methanol mole fraction		0.69	-1276629929194.66
Butyl acetate mole fraction		1.00	1930056.46

Furthermore, the optimization of the process was carried out as a multiobjective type in which the objective functions were the maximization of the mole fractions of methanol and butyl acetate leaving the top and the bottom sections of the column respectively. From the results obtained through the optimization, it was discovered that the two optimization commands used in this case were able to perform very well because both of them could give a mole fraction that was greater than 0.85, which was the target value for each of the products. According to the table, the mole fractions obtained for methanol and butyl acetate when *fsolve* command was used were 0.88 and 0.89 respectively while the values of the mole fraction obtained for the two products when *fminimax* command was used was 0.89.

Table-10. Optimum values obtained when the maximization of top methanol and bottom butyl acetate mole fractions were the objective functions.

Parameter	Initial value	Optimization using *fsolve*	Optimization using *fminimax*
Reflux ratio	1	1.28	0.60
Butanol flow rate (mL/min)	20	18.83	13.85
Methyl acetate flow rate (mL/min)	20	17.75	14.87
Reboiler duty (kJ/s)	0.5	0.35	0.10
Methanol mole fraction		0.88	0.89
Butyl acetate mole fraction		0.89	0.89

Furthermore, the values of the input variables in this case of multiobjective optimization were found to change significantly for the two MATLAB commands (*fsolve* and *fminimax*) employed when compared to the other two optimizations carried out before in this work. This was found to be an indication that the process really responded to the input variables in the course of this multiobjective optimization.

4. CONCLUSIONS

The results obtained from the modelling and simulation of the reactive distillation process used for the production of methanol and butyl acetate from the transesterification reaction between butanol and methyl acetate revealed that the developed models were good representatives of the top and the bottom sections of the column used, because there were good correlations between the measured and the simulated mole fractions to the extent of having R-squared values of 99.32% and 99.03% respectively for the top and the bottom section models. Furthermore, it was observed from the optimization carried out that handling this type of a system in a multiobjective way was better because that was the one that gave the desired optimum values of the two products from their respective sections of the column considered.

Nomenclature

Adj	Adjusted
Q_c	Condenser heat duty (kJ/s)
Q_r	Reboiler heat duty (kJ/s)
x_1	Reflux ratio
x_2	Butanol volumetric flow rate (mL/min)
x_3	Methyl acetate volumetric flow rate (mL/min)
x_4	Reboiler duty (kJ/s)

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