

SIMULATION AND INTELLIGENT CONTROL OF NATURAL GAS PURIFICATION

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ABSTRACT

Natural gas sweetening process is a controlled process that must be done at maximum efficiency and with the highest quality. In this work, the process of H₂S gas refining and separation with DEA in an absorption tower tray is simulated in MATLAB. Due to complexity and non-linearity of the process, intelligent fuzzy controller is used to control the process. The absorption rate and heat generation of reaction has been included in simulation also. To achieve the best sweetening rate three controllers are proposed to control the inlet gas rate, the inlet amine rate and the temperature of inlet amine. Simulation results show a novel improvement in process performance.

Keywords: Gas Sweetening, Intelligent, Fuzzy Controller, Absorption

INTRODUCTION

Hydrogen sulfide in sour natural gas causes corrosion in the equipment, reduces the economic value of gas, pollutes the environment and its smell brings poisoning and at relatively low concentration has lethal effect; therefore, it is necessary to separate it from the sour gas. If the concentration of H₂S in the natural gas is more than 4 PPM, it will be acid gas. To separate acid gas, such as hydrogen sulfide, from natural gas, towers tray and solutions such as amines are used in the gas industries. Hydrogen sulfide is absorbed along with chemical reaction and this reaction has two positive effects on the absorption intensity; first, the absorption rate is increased by reduce of the equilibrium partial pressure, and second, it increases the mass transfer coefficient, so arise the absorption intensity. Various models are presented for simulation of the absorption processes with chemical reaction. Smith and Jaswon presented the first dynamic model of gas absorption towers (Jaswon and Smith, 1954). Dynamic model which was obtained by Smith *et al.*, had complex mathematical equations, was analyzed and evaluated by Tommasi and Rice and they suggested a new solution for unknown boundary conditions of those complex equations (Tommasi and Rice, 1970). Macavy and Liapis presented a linearization method to overcome the slow convergence of Smith and Jaswon method (Liapis and Macavoy, 1981). Bardly and Andrew presented another dynamic model for gas absorption tower in which a particular method is used to calculate the amount of mass transfer (Bardley and Andre, 1972). Potter and Lakshmanan presented a dynamic model of time provided for systems with different directions currents, which was used for the modeling of the absorption packed columns (Lakshmanan and Potter, 1987). Rahimpour had studied simulation and optimization of the reverted gas into the methanol reactor (Rahimpour *et al.*, 1998). Ghader presented optimal values for feed temperatures (operating point) reactor feed to methanol reactor which was simulated (Ghader and Fathikalajahi, 2000). Løvik obtained optimal value for temperature of the cooling water and amount of reverted gas into reactor, in addition to simulated methanol synthesis reactor (Løvik, 2001). Doyle and Balasubramhanya (2000) presented a controller based nonlinear model for Ethyl Acetate Reactive distillation tower (Doyle and Balasubramhanya, 2000). Kumar and Daoutidis presented a framework of algebraic differential equations for control continuous reactive distillation tower (Kumar and Daoutidis, 1995). Fatima Barcelo Rico et al used a fuzzy control technique to control continuous distillation tower (Fatima *et al.*, 2011). Jeen and Ruey-Jing (2009) presented continuous fuzzy logic and neural network controller for systems with multiple inputs and outputs (Jeen and Ruey-Jing, 2009). Harun *et al.*, presented a fuzzy logic controller to improve the dynamic operation of catalytic cracking unit (Harun *et al.*, 2006). Although much works are done in the field of the topics of thermodynamics and synthetic

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reaction absorption towers, no other studies have been done except of the Mohamed. Mohamed (2011) investigated absorption tower without chemical reaction at constant pressure and temperature conditions, and he just controlled the concentration of exhaust gas from the tower (Mohamed, 2011). In this study, the process of H₂S gas refining and separation with DEA in an absorption tower tray is simulated by the use of MATLAB software. Due to complexity and non-linearity of the process, intelligent fuzzy controller is used to control the process. In this simulation the absorption rate and heat generation of reaction has been also included. To achieve the best sweetening rate three controllers are proposed to control the inlet gas rate, the inlet amine rate and the temperature of inlet amine. Simulation results show an appropriate improvement in process of performance.

Modeling of Reactive Absorption Tower

For mass and energy balance in the reactive absorption counter-current, following assumptions to simplify the model equations and equations of state have been considered.

Assumptions and Definitions

1- Gas and liquid flow are counter-current and perpendicular through the tower. Each stage of the process is an equilibrium stage, that is, the vapor leaving a stage is in thermodynamic equilibrium with the liquid on that stage.

2- Absorption process in the tower is adiabatic and its thermal capacity of tower is ignored.

3- The liquid molar holdup is constant.

We now introduce the following variable definitions:

L = moles inert liquid per time: = liquid molar flow rate.

V = moles inert vapor per time: = vapor molar flow rate

m = moles liquid per stage: = liquid molar holdup per stage

ω = moles vapor per stage: = vapor molar holdup per stage

x_j = moles solute (stage j) per mole inert liquid (stage j)

y_j = moles solute (stage j) per mole inert vapor (stage j)

$$\frac{\omega dy_{j-1}}{dt} + \frac{mdx_j}{dt} = Lx_{j-1} + Vy_j - Lx_j - Vy_{j-1} - mr_j(1 - 2)$$

Using the equilibrium assumption and relationship:

$$y_{j-1} = kx_j$$

The derivative of equation (2-2) will result:

$$k \frac{dx_j}{dt} = \frac{dy_{j-1}}{dt} (2 - 2)$$

Replacing equation (3-2) into side left of equation (1-2) result in equation (4-2):

$$\frac{\omega k dx_j}{dt} + \frac{mdx_j}{dt} = Lx_{j-1} + Vy_j - Lx_j - Vy_{j-1} - mr_j(4 - 2)$$

$$(m + k\omega) \frac{dx_j}{dt} = Lx_{j-1} + Vy_j - Lx_j - Vy_{j-1} - mr_j(5 - 2)$$

The total amount of solute on stage j is the sum of the solute in the liquid phase and the gas phase (that is, $m x_j + \omega y_j$). Thus the rate of change of the amount of solute is $d(m x_j + \omega y_j)/dt$ and the component material balance around stage j can be expressed as(4 - 2).

Where we assumed that in accumulation, liquid is much denser than vapor. Under assumption3), then (6 - 2)simplifies and equation (7-2) is obtained:

$$\frac{mdx_j}{dt} \cong Lx_{j-1} + Vy_j - Lx_j - Vy_{j-1} - mr_j(6 - 2)$$

$$\frac{dx_j}{dt} \cong \frac{L}{m} x_{j-1} + \frac{V}{m} y_j - \frac{L}{m} x_j - \frac{V}{m} y_{j-1} - r_j(7 - 2)$$

Replacing (2-2) into (7-2) will result:

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$$\frac{dx_j}{dt} = \frac{L}{m} x_{j-1} + \frac{V}{m} kx_{j+1} - \frac{L}{m} x_j - \frac{V}{m} hx_j - r_j (8-2)$$

Arrangement of (8-2) equation led to following term:

$$\frac{dx_j}{dt} = \frac{L}{m} x_{j-1} + \frac{V}{m} kx_{j+1} - \frac{(Vk + L)}{m} x_j - r_j (9-2)$$

Now, equation (9-2) is obtained for each of 6 trays as follow:

$$\frac{dx_1}{dt} = ax_0 + bx_1 + cx_2 - r_1 (10-2)$$

$$\frac{dx_2}{dt} = ax_1 + bx_2 + cx_3 - r_2 (11-2)$$

$$\frac{dx_3}{dt} = ax_2 + bx_3 + cx_4 - r_3 (12-2)$$

$$\frac{dx_4}{dt} = ax_3 + bx_4 + cx_5 - r_4 (13-2)$$

$$\frac{dx_5}{dt} = ax_4 + bx_5 + cx_6 - r_5 (14-2)$$

$$\frac{dx_6}{dt} = ax_5 + bx_6 + cy_6 - r_6 (15-2)$$

$$y(t) = Qx(t) \quad (17-2)$$

$$\frac{dx}{dt} = Ax + BC$$

$$x(t) = Ax(t) +$$

In last equation, A, B and Q is the system matrix, inlet matrix and outlet matrix, respectively, in the equations of state:

$$B = \begin{bmatrix} \frac{L}{m} & 0 \\ 0 & 0 \\ 0 & 0 \\ 0 & 0 \\ 0 & 0 \\ 0 & \frac{V}{m} \end{bmatrix}, A = \begin{bmatrix} b & c & 0 & 0 & 0 & 0 \\ a & b & c & 0 & 0 & 0 \\ 0 & a & b & c & 0 & 0 \\ 0 & 0 & a & b & c & 0 \\ 0 & 0 & 0 & a & b & c \\ 0 & 0 & 0 & 0 & a & b \end{bmatrix}, x = \begin{bmatrix} x_1 \\ x_2 \\ x_3 \\ x_4 \\ x_5 \\ x_6 \end{bmatrix}, Q = \begin{bmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{bmatrix}, C = \begin{bmatrix} x_0 \\ 0 \\ 0 \\ 0 \\ 0 \\ y_6/h \end{bmatrix}$$

Temperature Changes Equation of Trays

Then the energy balance for a tray Amine on the tray and with this assumption that Amine value is too low .Amine rate changes versus length of the tray irrespective and at one point is considered. And then be extended to the whole tray Thus, the energy equation is obtained as follows:

In this equation, the enthalpy is obtained of tables Maddox's book, HΔ heat of reaction The reaction rate R(mol/ft³.s), CP heat capacity amine, (T_{n+1}^L - T_n^L) amine temperature difference between inlet and outlet of each tray °F, L flow rate amine, (V (ft³ Size Tray, c concentration Amine is (mol/ft³).

$$\Delta H = 613btu$$

The amount of heat transferred by the gas mass ratios are obtained using stoichiometric relationships And with the assumption that methane gas is transmitted to the top of the tower (sweet gas) (Maddox) value of 0.19 is obtained By Multiplication 0.81ΔH considered in designs.

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Design of Fuzzy Controller

Fuzzy control is by far the most successful application of fuzzy sets and systems theory to practical problems. Numerous applications of fuzzy logic controllers to a variety of consumer products and industrial systems have been recorded (Løvik, 2001). Fuzzy systems are linguistic knowledge based system. The heart of a fuzzy system is what so-called fuzzy IF THEN rules. These rules are statements in which some words are described by continuous membership function. For example, IF vessel temperature High THEN fuel value opening Small. IF vessel temperature Low THEN fuel value opening wide.

Simulation Absorption Tower

Figure 1 Schematic of an absorption tower with three fuzzy controllers and show location each of these controllers with two inputs and one output. The reactive absorption tower containing six trays in Matlab environment using equations of state mass (5-10) to (5-15) and energy equation (5-19) is simulated (Figure 2).

In this tower flow rate amine inlet 80 mol/ min, gas rate inlet 100 mol/ minutes, and Molly liquid molar holdup 200 mol/min has been considered (Fatima *et al.*, 2011). Then tower simulation, three fuzzy controllers up and down the tower to help a number of fuzzy rules is designed (Figure 3).

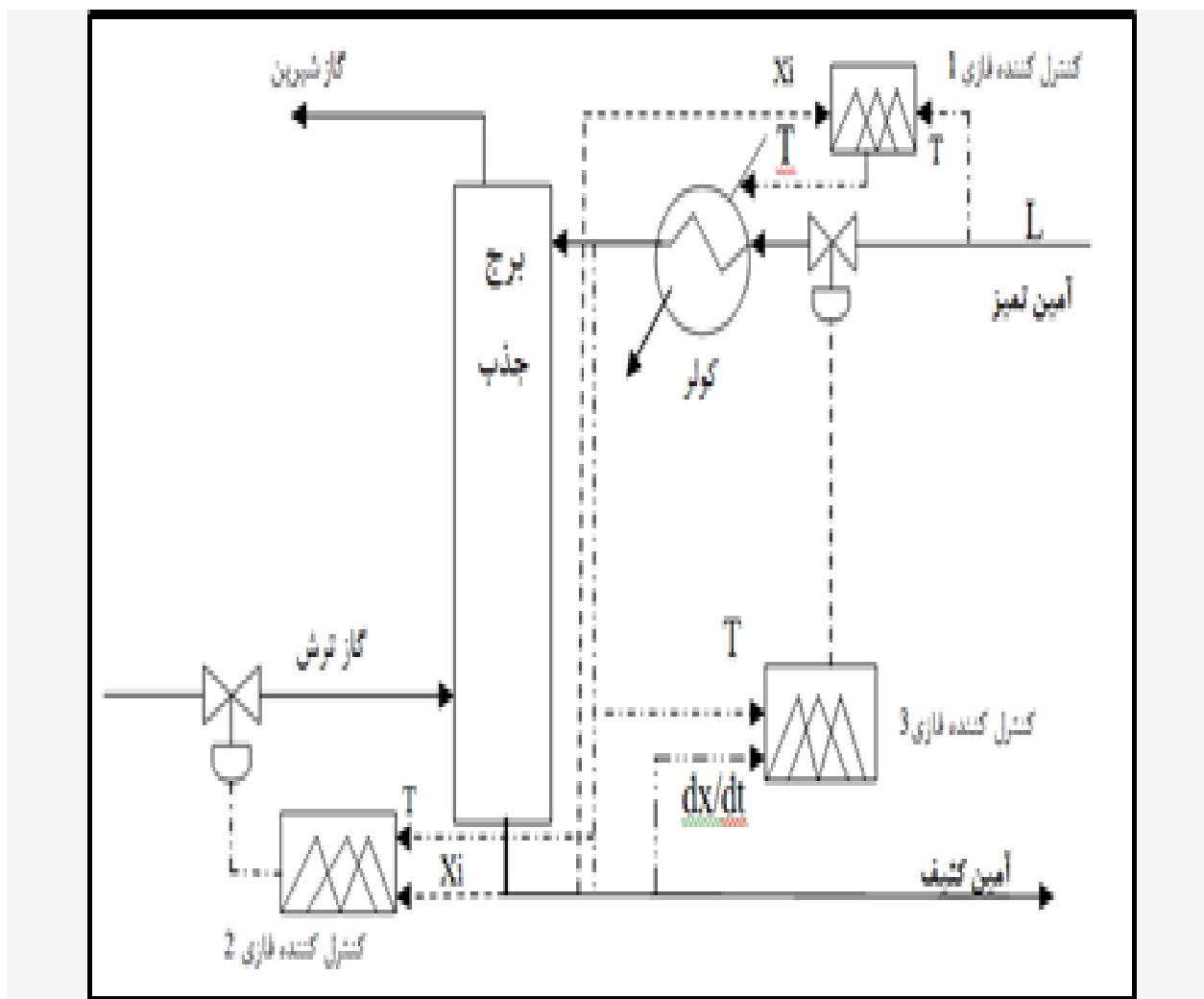


Figure 1: Absorption towers and installation of three fuzzy controllers

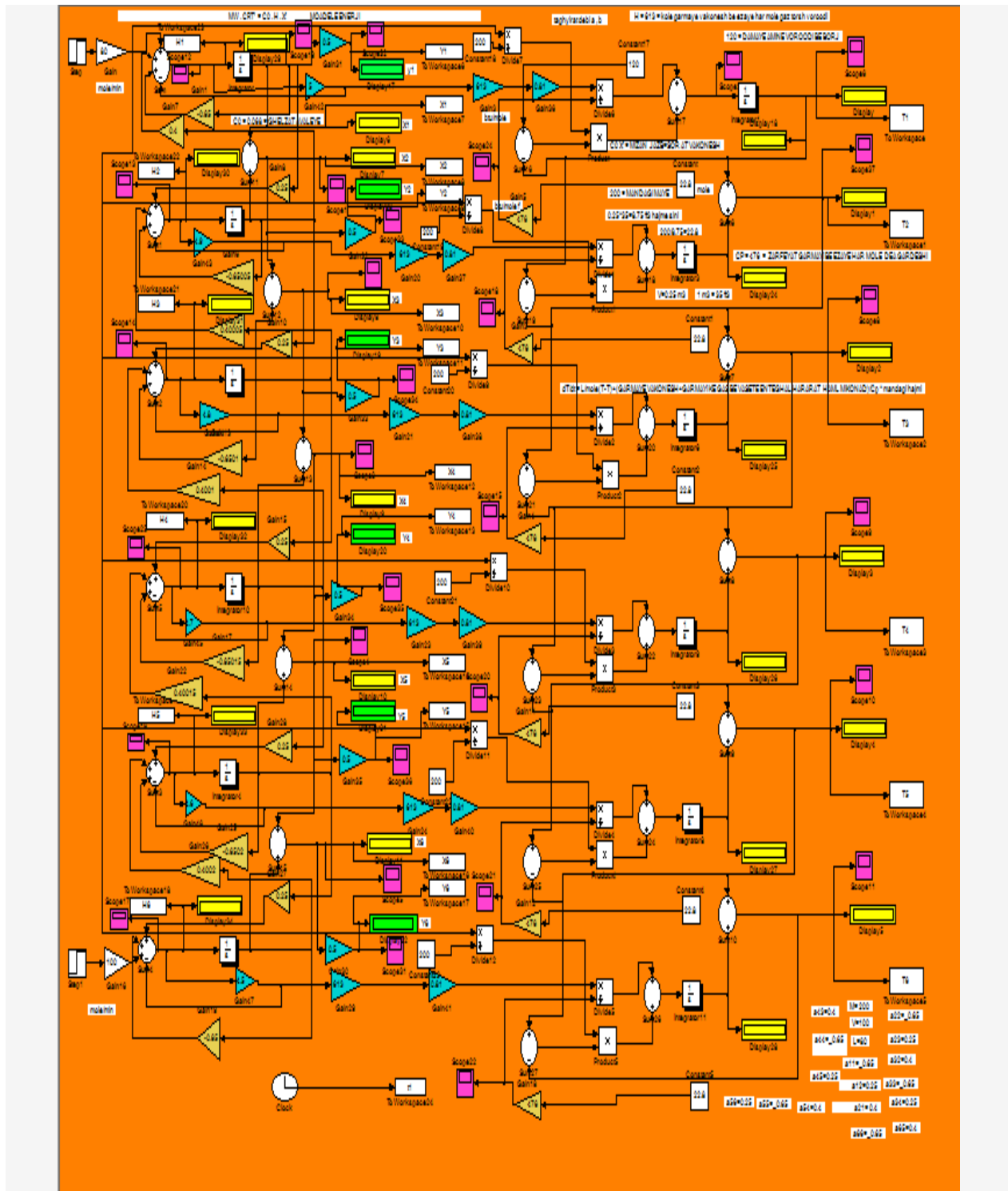


Figure 2: Simulated absorption tower with six trays

Control Laws in top of Tower 1 (Table 1)

The controller (1) truly one of the entrances to controller the tower inlet Amine temperature
 And other entries Amine concentration in the tower top and the controller output, at the entrance to the
 tower is the amines.

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Table 1: Control Rules (1)

T	VL	ML	L	H	MH	VH
VL	VH	MH	H	L	ML	VL
ML	H	H	H	L	ML	L
L	H	MH	H	L	ML	VL
MID	CON	CON	CON	CON	CON	CON
H	H	MH	H	L	ML	ML
MH	VH	H	H	L	L	VL
VH	VH	MH	H	L	ML	VL

Control Law (3) and Table (2)

One of the inputs to the controller Amine inlet temperature and other entries Rate Absorption Sour gas is at the bottom of the tower and the controller output, Rate Amine entrance of the tower.

Table 2: Control Rules (3)

T	VL	Lmid	L	mid	H	Hmid	VH
VL	VL	L	H	H		Lmid	VL
Lmid	L	H	H	H		L	Lmid
L	L	L	H	H	L		L
MID		H	H	CON	CON	CON	
H		CON	CON	CON	CON	CON	
Hmid		CON	CON	CON	CON		
VH		Lmid	L	L	L	L	

Table 3: Control Rules (2)

Xi	T	VL	Lmid	L	mid	H	Hmid	VH
VL		VL	L	H	H		Lmid	VL
Lmid		Lmid	Lmid	H	H		L	Lmid
L		L	H	H	H			L
MID			H	CON	CON	CON	CON	
H			CON	CON	CON	CON	CON	
Hmid			CON	CON	CON	CON	L	
VH			Lmid	L	CON	L		

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Control Rules (2) Table 3

Amine inlet temperature is one of the inputs to the tower controller and other entries gas Concentration in amine at the bottom of the tower Output is the amount of gas entering the tower.

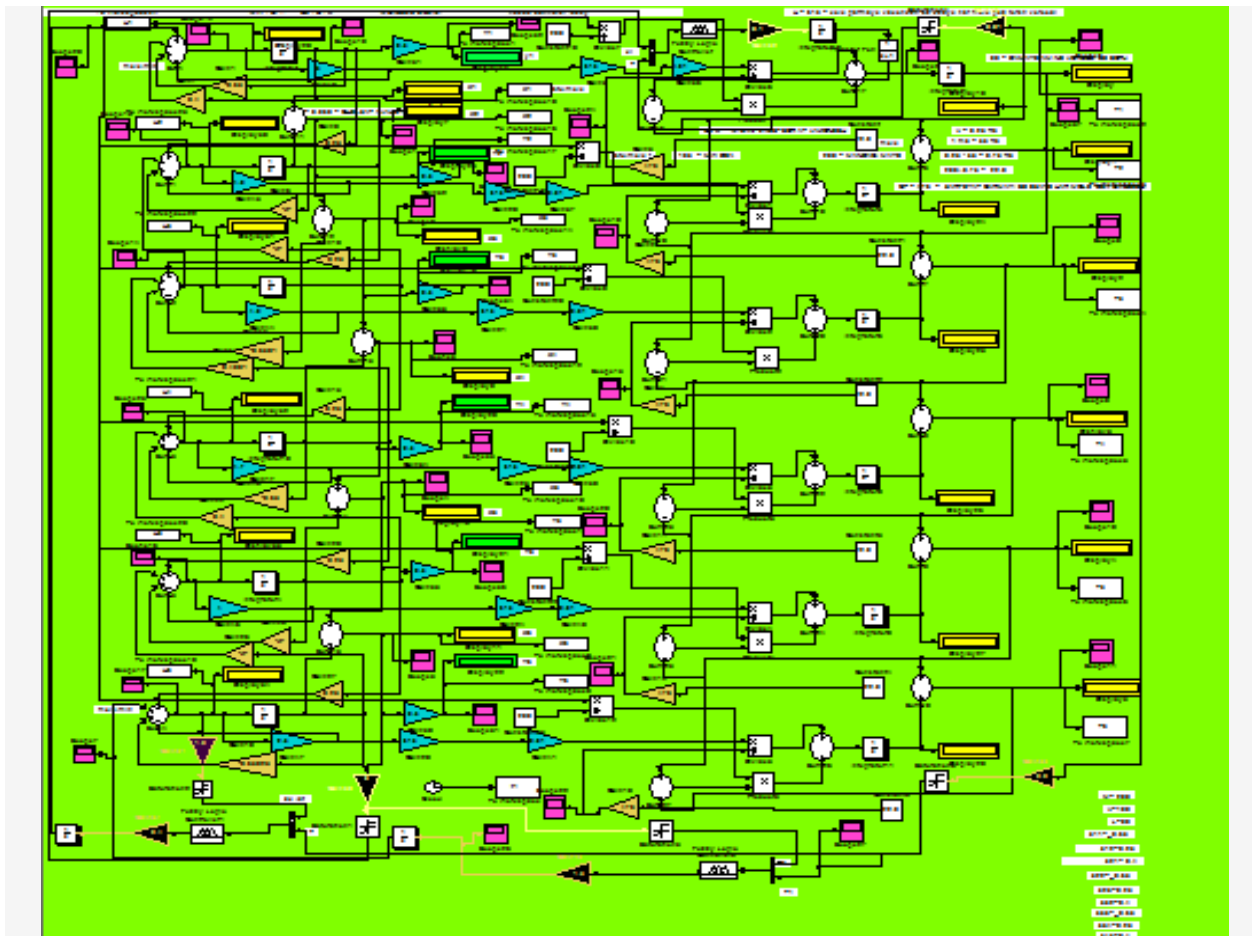


Figure 3: Simulated absorption tower with six trays and three fuzzy controllers

Analysis Somerules

(6-1) Icon of Fuzzy Rules

VL: very low, ML: Low-Medium, L: Low, MID: medium, H: high: MH High Medium: VH, veryhigh, CON: Fixed

If the concentration of gas in the amine VL and amine temperature VL then amine temperature entrance to the tower is VH.

(6-2)Analysis of the Law No.1: One reason for the lack of favorable absorption tower, foaming amine and created one of the reasons for this defect, low temperatures, Amine entrance to the tower is too much So Fuzzy controller to control and reduce energy consumption Coolers Do not let the temperature go down. Amine temperature is higher compared to the previous case.

6) If (xi), VL and amine temperature VH is the temperature of the incoming amine tower is VL.

(6-3) Analysis of the Law No.6: amine decomposition is the One reason for the lack of favorable absorption tower, The reason for this defect is due to high temperatures, amine input to tower So Fuzzy controller to control And increased energy consumption Coolers Do not let the temperature is too high and Amine temperature is Holds down compared to the previous case.

20) If (xi), MID and temperature of the amine is the L The temperature of the incoming amine tower is CON.

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(6-4) Analysis of Law No. 20: If the amount of sour gas in amine is not too high and not too low and in this situation, amine temperature is low. Such a situation is favorable. Thus, the controller keeps a constant temperature in the amine.

Analysis of Results

Simulation results include the operating parameters such as input temperature, amine hydrogen sulfide concentration in the gas phase, and amine phase and are rate phases. Figure 4 shows temperature changes in the tower when a controller is not installed, as it can be seen, input amine temperature when it enters the first tray is 120° F. The temperature of the top tray to the bottom tray tower is increasing by about 2 degrees Fahrenheit which is due to exothermic chemical reaction between the gas phase and amine. As can be seen, after increasing the temperature of each tray temperature increases due to lack of suitable design of a controller, during tray the temperature remains constant until the amine re-enters the next tray and after the temperature is raised by 2 and then is constant. Figure 5 shows changes in the concentration of hydrogen sulfide in amines without a controller. As it can be seen the concentration of Sulfide Hydrogen at the top tray of the tower is about 220ppm and at the bottom tray are 110, which show exhaust gas is sour.

Left-side diagram Figure 6 shows the temperature changes of the liquid phase input tray and the right diagram shows the outlet temperature of each tray. As it can be seen where concentration of gas in the amine due to the high temperature controller is low, the controller orders decreasing the temperature till it becomes about 25 to 30 ° C, or 65 to 75 F because of the foaming amine is controlled and the graph remains constant. In the right side of the graph the temperature is increased because the fuzzy rules are defined; for example, if the amine concentration is low and the temperature is low too, it causes the amine foam and reduces the absorption, so the controller orders the cooler that decrease the energy until the temperature raises.

The raised temperature is shown in Fig. But it does not allow the temperature to raise much because the increase in temperature not only causes decomposition of amines but also reduces absorption. Figure (7) changes in the concentration of hydrogen sulfide in the gas phase without controlling the show in this case, concentration of each tray. is high Concentration in the bottom tray of the tower and the Atop the tower ppm500 is almost ppm110 As shown in Figure (8) and (9) are shown The amount of dissolved hydrogen sulfide is absorbed by with time the tower rises from the top down Due to the absorption more of hydrogen sulfide in the bottom of the tower and The fourth and fifth sections of the trays, changes in at the bottom of the tower relative change in concentration the Atop the tower, first and second trays tower is more and The first tray of hydrogen sulfide gas in its standard range is approximately 3.6 ppm . Figure (10) changes in the inlet gas temperature for each tray at a different rate compared with each other and show increasing gas flow rate increases the temperature of the tray.

Figure (11) changes in concentrations of acid gas output tray at a different rate the gas inlet compared with each other. And shows With increasing inlet gas flow H₂S concentration in the output gas is further The increase in concentrations higher gas rates may be higher. Figure 12 Changes in absorption rate of H₂S gas in each tray at different inlet gas flow rate shows As can be seen Absorption maximum in the middle of the tray tower With increasing gas flow rate very low increases the absorption rate of each tray. Figure 13 compares the temperature variation in the liquid phase shows both the controller and the controller. As can be seen, due amine reaction temperature increased from top to bottom tower this increase in temperature without control is more. Most differences in temperature in the control are trays 4 and 5 because there is maximum absorption. Figure 14 compares the concentration of hydrogen sulfide in the gas phase over the tower without the controller and the controller shows Most of the hydrogen sulfide is absorbed in the first column and thus the concentration of hydrogen sulfide gas in the tray top of the tower is near the permitted limit. And when leaving the tower controlling the state reach to be below the standard value 4 ppm .whereas output concentration without controlling is 92.48 ppm. Figure (15) acids Gas absorption rates of different rates at the entrance amine is show As can be seen, with increasing amine input increases the absorption rate Also .The maximum absorption tower appears in the middle of the tray.

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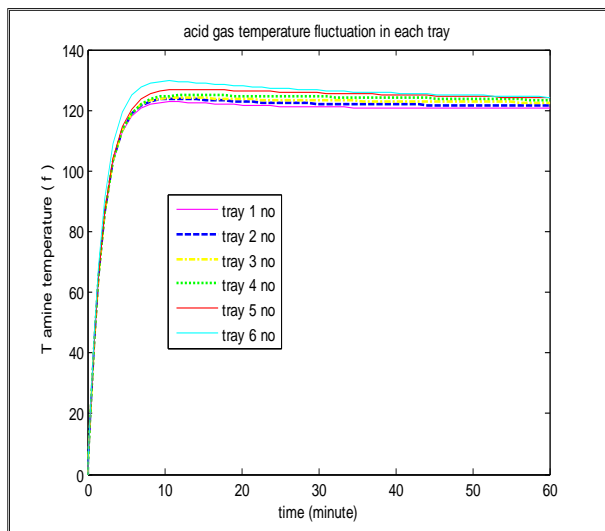


Figure 4: Changing the tray temperature without controller

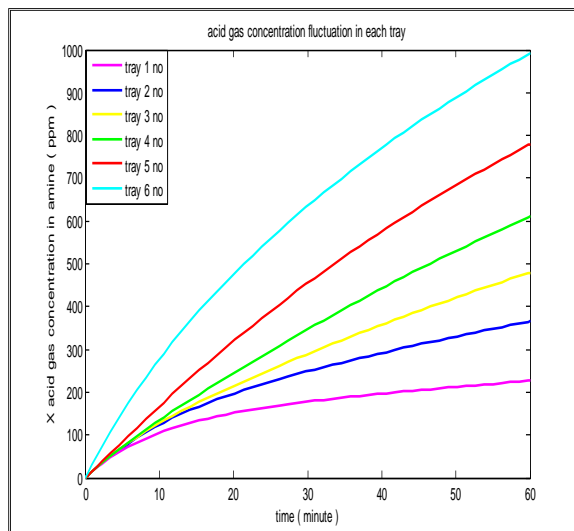


Figure 5: Changes in the concentration of hydrogen sulfide without controller

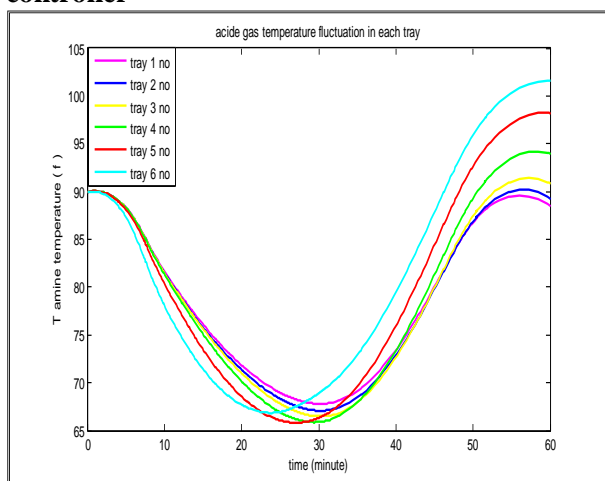


Figure 6: Liquid phase change temperature with controller

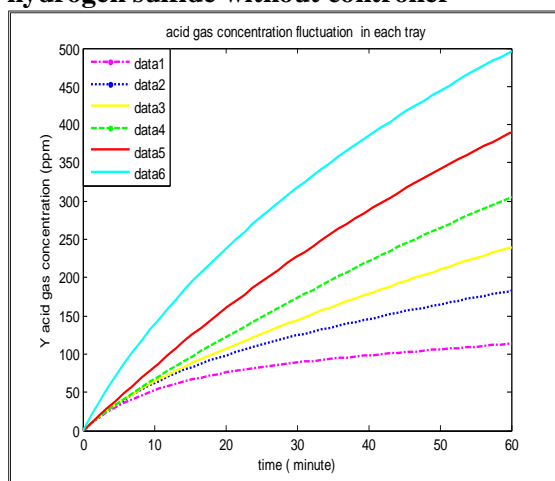


Figure 7: Changes in the concentration of hydrogen sulfide gas without controller

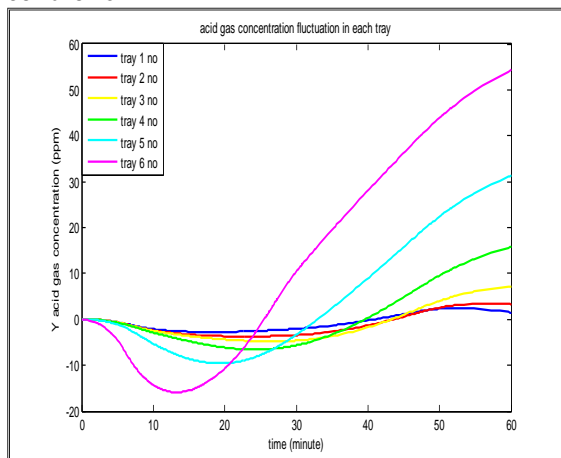


Figure 8: The concentration of hydrogen sulfide in the gas phase with controller

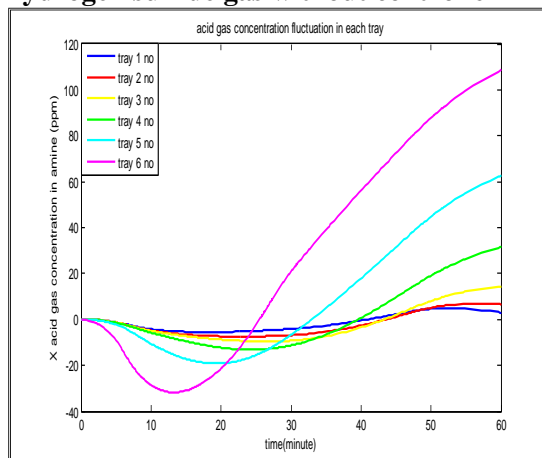


Figure 9: Hydrogen sulfide concentration in the liquid phase with controller

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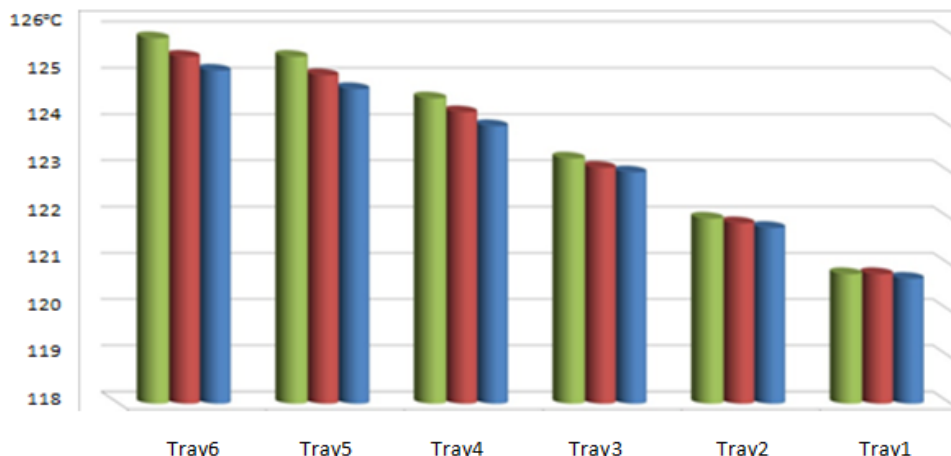


Figure 10: Changes temperature of each tray in different flow rate at inlet gas

L=80 (Mol/s) G=120 (Mol/s) T=120 (f) ■
 L=80 (Mol/s) G=110 (Mol/s) T=120 (f) ■
 L=80 (Mol/s) G=100 (Mol/s) T=120 (f) ■

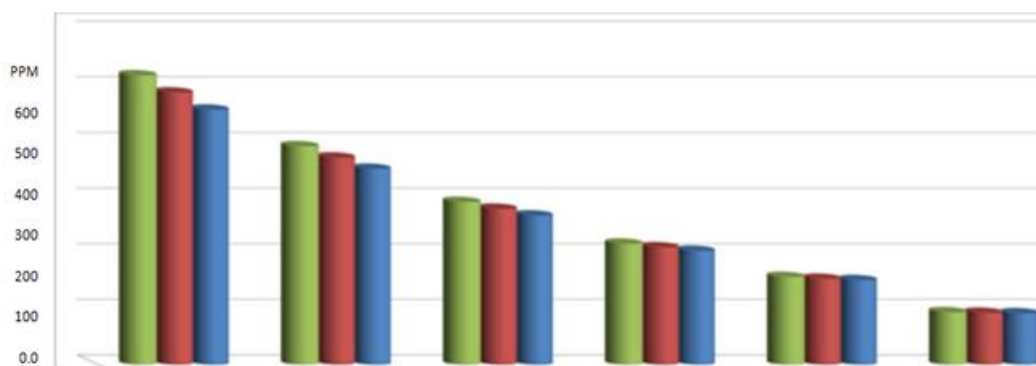


Figure 11: Changes in concentrations of acid gas output tray at different flow rate discharge gas inlet (ppm)

f) دما=80 (Mol/s) G=120 (Mol/s) T=120 (f) ■
 f) دما L=80 (Mol/s) G=110 (Mol/s) T=120 (f) ■
 f) دما L=80 (Mol/s) G=100 (Mol/s) T=120 (f) ■

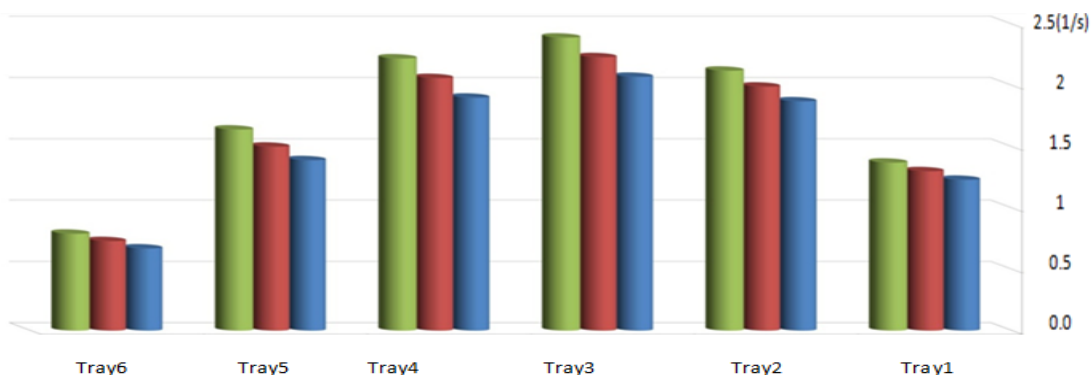


Figure 12: Changes in absorption rate of H2S gas in each tray at different inlet gas flow rate (1 / s)

L=80 (Mol/s) G=120 (Mol/s) T=120 (f) ■
 L=80 (Mol/s) G=110 (Mol/s) T=120 (f) ■
 L=80 (Mol/s) G=100 (Mol/s) T=120 (f) ■

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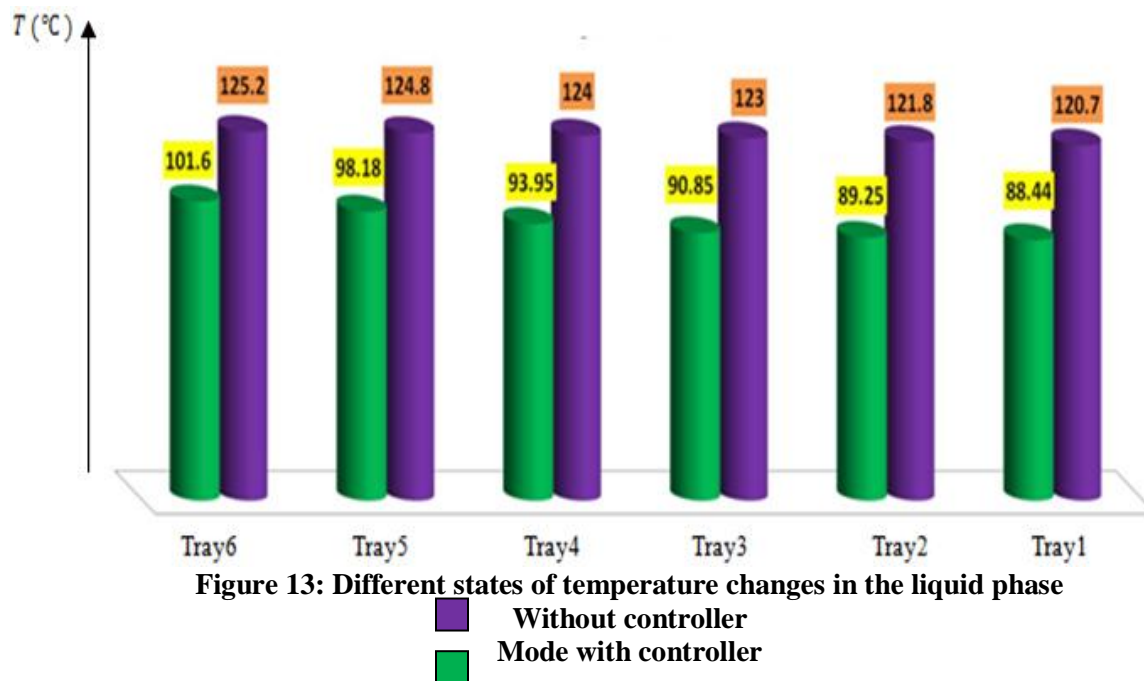


Figure 13: Different states of temperature changes in the liquid phase

■ Without controller
■ Mode with controller

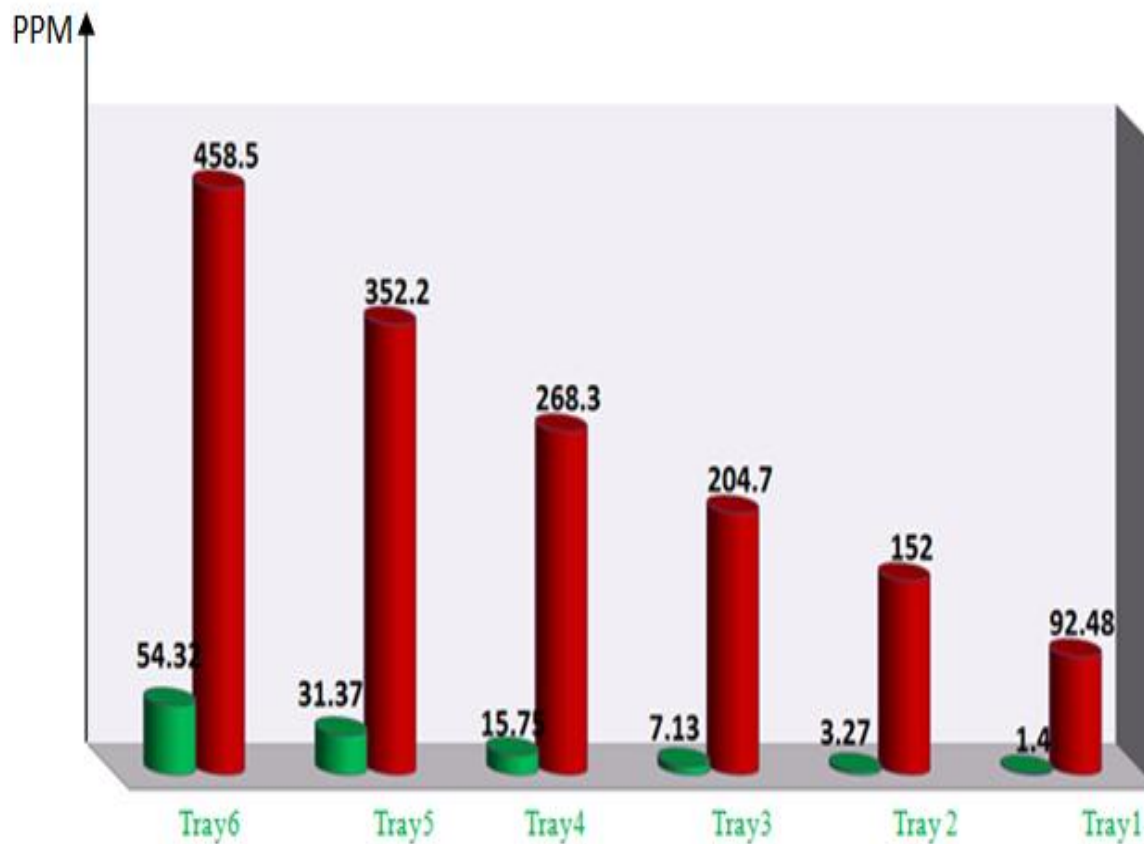


Figure 14: Changes in the concentration of acid gas (hydrogen sulfide) gas phase in different states

■ Without controller
■ Mode with controller

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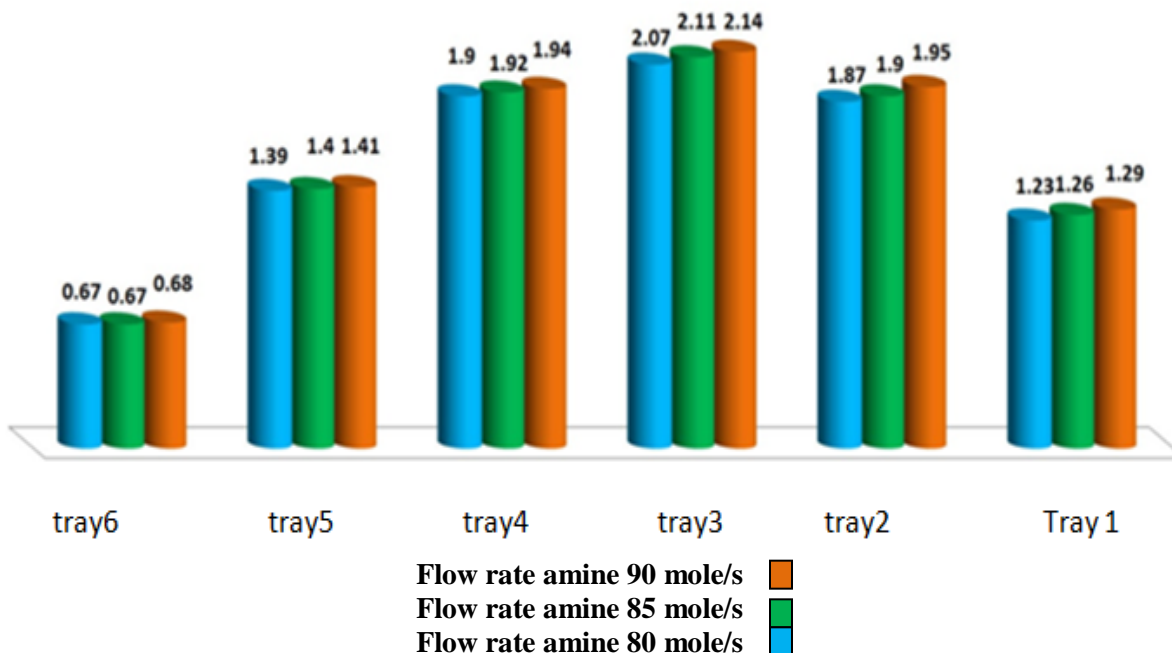


Figure 15: Acid Gas absorption rates of different rates at the entrance amine

Conclusion

- 1- In this work, equations of state for reactive absorption tower are obtained and reactive absorption tower is simulated with MATLAB for the first time. Comparison of result with experimental data shows the simulation well present the behavior of process in the given operating conditions at dynamic state.
- 2- In this work, intelligent control of reactive absorption tower is done by fuzzy controllers.
- 3- It was found that temperature of the inlet fresh amine is a function of inlet gas temperature, value of consumption energy in the coolers and ambient temperature. At lower temperature, the absorption reaction due to improved conditions can do better and absorption value will be greater.
- 4- The maximum temperature difference there are in the bottom and middle trays of tower and temperature of trays increase from top to bottom of tower, i.e., tray 1 to 6 because, at first, the inlet sour gas to the tower has more mass transfer value according to the acid gases concentration gradient and, consequently, the released heat value of reaction causes to increase the temperature. Of course, it is predictable that for a tower with 24 trays will be occurred at middle trays of tower.
- 5- Applying the fuzzy controllers caused the outlet acid gas concentration reach to 4 PPM, while, this value reached to 220 PPM at designing without fuzzy controllers.
- 6- The input amine temperature designed without controller is 120°F and from the first tray to the Sixth the temperature is rising. Temperature of each tray shows a two-degree temperature difference with the previous one. But in the case-control design and cooling system, the amine inlet temperature is reduced to 90°F.
- 7- In the case-control design the amount of dissolved hydrogen sulfide in a tower by passing the time rises from the top to the bottom and because the trays absorb more hydrogen sulfide in the bottom of tray which are the fifth and sixth column, the relative change in concentration of the top of the tower, which are the first and the second is greater. In the sixth tray (lower column) hydrogen sulfide of gas is approximately 105 ppm and the first tray (exhaust gas from the top of the tower), hydrogen sulfide gas in its standard range is approximately 3.6 ppm.
- 8- Once the controllers make foam in amine and reduce the absorption of H₂S, they control gas flow rate in inlet tower in order to control and optimize amine and prevent the loss of it. So, forbid industrial pollution and increases the sweetened gas amount.

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