

# Rate-Based-Model Analytical Method for Sour Gas Absorption Simulation by Alkanolamine

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## ABSTRACT

In this research, a film-model-based for gas sweetening by alkanolamine is considered and three different approaches to the solution of the CO<sub>2</sub> mass transfer- reaction in the liquid film are studied. Also the results are compared with each other and pilot plant data. The first approach is based on numerical method for solving nonlinear boundary value problem. The second approach requires a simplification of differential transport equation by assuming uniform amine concentration in liquid film and bulk of liquid. The third approach is polynomial method and furthers these two later methods, by taking advantage of an analytical technique. Numerical method was solved by complex matrix solution. Advantage and disadvantage of these methods are analyzed and investigated. The three models have good agreement with pilot plant data.

## KEYWORDS

CO<sub>2</sub> absorption columns, Alkanolamine, MEA, DEA, Modeling, Analytical solution, concentration profiles, Polynomial solution

## 1. INTRODUCTION

Absorption of gases in liquids accompanied by chemical reaction (reactive absorption) is a fundamental operation in a broad spectrum of chemical process technology [1].

Alkanolamine solutions are frequently used for the removal of acid gases (carbon dioxide) from industrial and natural gas streams. Simultaneous absorption and the reactions between CO<sub>2</sub> and alkanolamine solutions have been studied extensively [2, 3, 4].

In reality, equilibrium is rarely attained at a stage since absorption is a rate controlled phenomenon [5]. This means that mass and heat transfer are kinetically limited processes driven by gradient of the chemical potential and temperature. Due to these facts, traditional equilibrium-base models and efficiency approaches are usually inadequate [6].

Resistances to mass and heat transfer can only be considered by rate-base models, in which gas and liquid phase are balanced separately under consideration of mass and heat fluxes across the interface [7]. In addition, rigorous predictive models have to regard the accelerating effect of chemical reactions on mass transfer [8]. Modeling of reactive absorption unit have been based on two film theory. It should be noted that mass transfer and reaction with amines take place simultaneously.

Concentration profiles of CO<sub>2</sub> and amine in liquid film are needed for determining mass fluxes at gas and liquid interface, liquid film and liquid bulk.

The governing equations for calculation of concentration profile, in detail, were presented in other works [9]. This equation is difficult to solve and requires tedious calculation.

Using numerical solution for reactive separation modeling and design may still cause convergence and stability problems. Kenig & Gorak modified analytical method is based on a linear approximation of the reaction term [10]. Delancy used an analytical solution for reactive separation model. Unfortunately, several errors were discovered in this solution. Kenig *et al.* solved Delancy system by using simple modification, and avoided wrong results [9]. Other researchers have introduced enhancement factor in order to solve the analytical solution [11, 12]. Enhancement factor is the ratio of absorption rate of solute gas in the presence of chemical reaction to that obtained with physical absorption in the liquid film.

Tomcej *et al.* [12] developed a model for estimating the enhancement factor for removal of CO<sub>2</sub> using alkanolamine by assuming a pseudo first order and irreversible chemical reaction. Onda *et al.* [1] derived an approximate solution for a very fast and reversible reaction. Leye *et al.* [1] derived an analytical solution by

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assuming CO<sub>2</sub> reaction with alkanolamine is irreversible and of finite rate.

In this paper a rate-based model have been considered for the simulation and design of CO<sub>2</sub> absorption by alkanolamine solution. The approach requires a simplification of differential transport equation by assuming constant concentration of alkanolamine in the liquid film in each tray, and further, takes advantage of an analytical solution. A comparison of simulation results (analytical and numerical solution) and pilot plant data are presented

## 2. MATHEMATICAL MODELING

In the model, the gas is assumed to be in plug flow, while the liquid on the plates is completely mixed [1, 9]. The molar flux and composition of the gas entering the trays are calculated [9]. By assuming the composition of transferred components in the liquid bulk and using mass balance equations, the amount of liquid leaving each tray and amine composition concentration is evaluated [9]. The composition of CO<sub>2</sub> in the liquid bulk is evaluated by using the concentration profile of CO<sub>2</sub> and amine in the liquid film [9].

Calculated quantities of composition obtained by these equations are compared with initial assumed values and iterative procedure is repeated until convergence criteria are satisfied.

## 3. ANALYTICAL METHODS FOR EVALUATION OF CONCENTRATION PROFILE

### A. Constant amine concentration in liquid film method

In this method, the concentration profile of CO<sub>2</sub> in liquid film derived by assuming the uniform concentration for alkanolamine in film and bulk of liquid and by using the second Fick's law. It can be calculated that:

$$D_A \frac{d^2 C_A}{dz^2} = k C_A C_B^2 \quad A = \text{CO}_2, B = \text{amine} \quad (1)$$

Boundary condition:

$$\text{At } z=0 \quad \frac{dC_A}{dz} = -\frac{K_g P(y_{Ab} - H'_A X_{A1})}{D_A} \quad \text{and} \quad \frac{dC_B}{dz} = 0 \quad (2)$$

$$\text{At } z=\delta \quad C_A = C_{Ab}, \quad C_B = C_{Bb}$$

By rearranging and simplification

$$\frac{d^2 X_A}{dz^2} = \xi^2 X_A \quad (3)$$

$$\text{where} \quad \xi^2 = \frac{k}{D_A} C^2 X_B^2$$

Integrating equation (3) by using the boundary condition (2) leads to the expression for CO<sub>2</sub> mole fraction which depend on z only:

$$X_A = \frac{1}{\cosh(\xi\delta)} [X_{Ab} \cosh(\xi z) - \Psi \sinh[\xi(z-\delta)]] \quad (4)$$

$$\text{where } \Psi = \frac{N_{Ayo}}{C^2 X_B \sqrt{D_A k}}$$

From equation, (4) the CO<sub>2</sub> mole fraction at gas film-liquid film interface  $z=0$  and the CO<sub>2</sub> mole fraction profile at  $z=\delta$  are

$$X_{A1} = \frac{X_{Ab}}{\cosh(\xi\delta)} + \Psi \tanh(\xi\delta) \quad \text{at } z=0 \quad (5)$$

$$\frac{dX_A}{dz} \Big|_{z=\delta} = \xi X_{Ab} \tanh(\xi\delta) - \frac{\Psi \xi}{\cosh(\xi\delta)} \quad \text{at } z=\delta \quad (6)$$

### B. Polynomial method

The concentration profile of CO<sub>2</sub> in liquid film is derived by polynomial approach.

By rearranging of equation [1] for CO<sub>2</sub> and amine and using boundary condition [2], the relation between concentration CO<sub>2</sub> and amine in liquid film is obtained:

$$x_B = x_{Bb} + 2 \frac{D_A}{D_B} (x_A - x_{A,b}) - \frac{2N_A \Big|_{z=0}}{D_B C} (\delta - z) \quad (7)$$

By replacing [7] in [1], rearranging and integration, using the boundary condition [2] the CO<sub>2</sub> concentration profile is derived as:

$$x_A = a_0 + a_1 z + a_2 z^2 + a_3 z^3 + a_4 z^4 + a_5 z^5 + a_6 z^6 + a_7 z^7 \quad (8)$$

$$a_1 = \frac{-N_A \Big|_{z=0}}{D_A C}$$

$$a_2 = \left( \frac{a_0}{2} \right) \left( \frac{kC^2}{D_A} \right) \left( 2 \frac{D_A}{D_B} a_0 + \Psi \right)^2$$

$$a_3 = \frac{1}{6} [(\beta_1 + 3\beta_2 a_0^2 + 2\beta_4 a_0) a_1 + \beta_5 a_0^2 + \beta_6 a_0]$$

$$a_4 = \frac{1}{12} [(\beta_1 + 3\beta_2 a_0^2 + 2\beta_4 a_0) a_2 + (3\beta_2 a_0 + \beta_4) a_1^2 + (\beta_6 + 2\beta_3 a_0) a_1 + \beta_3 a_0]$$

$$a_5 = \frac{1}{20} [(\beta_1 + 3\beta_2 a_0^2 + 2\beta_4 a_0) a_3 + (\beta_6 + 2\beta_4 a_1 + 2\beta_3 a_0) a_2 + \beta_2 a_1^2 + \beta_3 a_1^2 + \beta_3 a_1]$$

$$a_6 = \frac{1}{30} [(\beta_1 + 3\beta_2 a_0^2 + 2\beta_4 a_0) a_4 + (\beta_6 + 2\beta_4 a_1 + 2\beta_3 a_0) a_3 + (3\beta_2 a_0 + \beta_4) a_2^2 + (\beta_3 + 3\beta_2 a_1^2 + 2\beta_3 a_1) a_2]$$

$$a_7 = \frac{1}{42} [(\beta_1 + 3\beta_2 a_0^2 + 2\beta_4 a_0) a_5 + (\beta_6 + 2\beta_4 a_1 + 2\beta_3 a_0) a_4 + (\beta_3 + 3\beta_2 a_0^2 + 2\beta_4 a_2 + 2\beta_3 a_1) a_3 + (3\beta_2 a_1 + \beta_3) a_2^2]$$

$$a_0 = x_{A,b} - [a_1 \delta + a_2 \delta^2 + a_3 \delta^3 + a_4 \delta^4 + a_5 \delta^5 + a_6 \delta^6 + a_7 \delta^7]$$

where:

$$\beta_1 = \frac{kC^2}{D_A} \Psi^2$$

$$\beta_2 = \frac{4kC^2 D_A}{D_B^2}$$

$$\beta_3 = \frac{4k}{D_A} \left( \frac{N_A \Big|_{z=0}}{D_B} \right)^2$$

$$\beta_4 = \frac{4kC^2}{D_B} \Psi$$

$$\beta_5 = \frac{8kC}{D_B^2} N_A \Big|_{z=0}$$

$$\beta_6 = \frac{4kC\psi}{D_A D_B} N_A \Big|_{z=0}$$

$$\psi = x_{B,b} - \frac{2D_A}{D_B} x_{A,b} - \frac{2N_A \Big|_{z=0}}{D_B C} \delta$$

#### 4. PHYSICO-CHEMICAL PROPERTIES AND MODEL PARAMETERS

In gas phase calculations, the compressibility factor of non ideal gas ( $Z$ ) is evaluated from Dranchuck *et al.* method [13]. Gas diffusivity coefficients and gas viscosity are calculated from modified Hirschfelder-Bird-Spotz and Underling methods, respectively [14, 15].

Mounik method is applied for the evaluation of the interfacial area per unit froth volume on the sieve plate [16].

The physical mass transfer coefficient in the liquid and gas is determined by Grester *et al.* [16].

Liquid viscosity and density are determined by Pohorecki *et al.* and Meisen *et al.* methods, respectively [16]. Henry and specific heat coefficients are evaluated by Kent *et al.* and Prausnitz *et al.* methods, respectively [17,13]. Diffusivity of CO<sub>2</sub> in liquid phase is calculated from Barret and Danckwerts method [16].

Film thickness is evaluated by modified correlation, using heat and mass transfer analogy as follows [18]:

$$\delta = \frac{0.024h_f}{(n_1^{0.8} + n_2^a)} \left( \frac{Z_1 h_w \mu_1}{V_1 \rho_1} \right)^{0.50} \left( \frac{\rho_1 D_{1,CO_2}}{\mu_1} \right)^{0.33} \quad (9)$$

where,  $a$  is 1.3 for very fast reaction, 1 for moderately fast reaction and 0.8 for very slow reaction. In the above equation, the number of the components which enter the liquid film or leave it and take place in the reaction is " $n_2$ " but without reaction is " $n_1$ ".

#### 5. EXPERIMENTAL SETUP

Absorption experiments were conducted using a pilot with seven tray column, where the amine solution was exposed to a flowing gas stream, consisting of knowing composition as shown in Table 1.

In this operation state, a natural gas stream is cleaned at 420.4 kPa by an amine solution with rate and temperature of 50 kg mol/h, 60 °C, respectively. The weight percent of amine in the relevant feed stream is 28.6% for DEA (Di Ethanol Amine), 19.9% for MEA (Mono Ethanol Amine) and 30.1% for MDEA-MEA (Methyl Di Ethanol Amine-Mono Ethanol Amine).

The CO<sub>2</sub> load of gas feed is about 6.5%. Tables 1,2 and Fig.1 give more detail on feed condition, and pilot plant flow diagram, respectively. The gas and liquid composition and temperature are measured at each tray.

Fig. 1 shows a simplified flow diagram of an amines gas treating pilot. The raw gas stream is fed to the bottom of absorption tower with sieve tray and is sweetened by

the counter-currently flowing amine solvent. The dissolved sour gases are removed from the solvent in the subsequent stripping column. The regenerated amine solution flow to surge drum and then is pumped to top of the absorber for further removal of acid impurities. The absorber column detail is shown in Table 2. CO<sub>2</sub> concentration at each tray was monitored by gas chromatography (Varian 3400).

#### 6. RESULTS AND DISCUSSION

Simulation of pilot plan data is performed to check whether a successful model is possible.

According to pilot plan data, the agreement between the experimental and simulated (Numerical and two Analytical methods) for the gas phase (Fig. 2), liquid phase (Figs. 3, 4) and tray temperature (Fig. 5) is very good.

In the first method, from the modeling point of view, the use of uniform concentration for alkanolamine in film and bulk of liquid in each tray means that we perhaps suffer loss in accuracy on the stage of model formulation, yet we benefit during the solution. The simplification is accomplished with a reasonable exactness. Therefore, the results seem to be good enough for getting adequate solution, and we can thus avoid calculation trouble. In the second method, the model was formulated without any simplification and must have good accuracy and agreement to plant data, as shown in Figs. 2-5.

We apply numerical solution techniques [9] and compare its result with two analytical findings. The results are shown in the Figs. 2-5. The numerical and two analytical results have good agreement, thus, the analytical methods provide reliable results which can be applied to the gas sweetening with reasonable accuracy.

The analysis of curves in Fig. 2 shows that the CO<sub>2</sub> mole fraction difference between each analytical methods and numerical method with pilot plant data is minor, in gas feed tray (Tray-7). The relative error grows as gas flow to upper trays (as shown in Tray 1), because the error of each tray is added to the next tray, therefore deviation became wider. But, the final error is now quite satisfactory.

To see whether these two solution approaches agree for all alkanolamine, we compared the CO<sub>2</sub> concentration profile for MEA (Fig.2-a), DEA (Fig 2-b) and MEA-MDEA (Fig.2-c) systems. They have similar results.

Although the relative error between the two analytical, and also the numerical results with pilot plant data is negligible but the error between three simulations model is smaller than that of each simulation model with pilot plant data, separately, because of

1. Non-uniform gas distribution in each tray.
2. Uncompleted mixing in liquid phase in each tray.
3. Non-uniform temperature on each tray.
4. Using the same procedure for determining the physical and chemical property of two

simulation models. Therefore, the results of these two simulation models are approximately the same.

5. The fact that the deviation between simulation results and pilot plant data is due to physico-chemical property calculation by semi-empirical correlations.

The analysis of the curves in Fig. 3, shows that the simulation results (two analytical and numerical models) and pilot plant data have good agreement with reasonable accuracy.

In amine feed tray (Tray 1) and three next trays (2-4) the simulation results and pilot plant data are approximately the same. The accuracy in these trays is due to:

- 1-High amine concentration in liquid phase
- 2- Low CO<sub>2</sub> concentration in gas phase.

As the amine flow to lower trays, the deviation between simulation results (three method) and pilot plant data increases, because the error in upper trays is added to each other (as mentioned for CO<sub>2</sub> mole fraction in gas phase).

Figs. 4, 5 illustrate amine concentration in liquid phase and temperature profiles, respectively.

Fig. 5 shows that the temperature increases rapidly in Tray -7 (gas feed tray) and Tray -6, also the maximum temperature exists in Tray -5. It is concluded that the most of the reaction between CO<sub>2</sub> and amine takes place at this three trays. This increasing in temperature causes vaporization of water from liquid phase, therefore the amine concentration in liquid phase increases (Tray -7) as shown in Fig. 4. From Tray -5 to Tray -1 temperature decreases, therefore water vapor condenses and decreases alkanolamine mole fraction in liquid phase, which is because of counter current flow.

The comparison of the two different analytical solution methods and numerical method done in this work is a very useful tool in providing important information on the peculiarities. Comparing the three methods suggested in this work, we can advocate that they are almost equivalent with regard to application, provided that care is taken of being within a proper range of process parameters and variables. This means that one can obtain almost identical concentration profile, by three methods (numerical and two analytical). If the weight percent of MEA, DEA and MEA-MDEA is not lower than 16, 20, 25, respectively, therefore the first analytical solution (constant amine concentration in liquid film method) can be recommended as a good alternative simulating large scale gas sweetening system.

The relative error between first analytical solution and plant data for CO<sub>2</sub> in sweet gas (Tray 7) and rich MEA (Tray 1) is 3.2% and 6.7%. These relative errors for DEA solution are 5.2% and 7%, respectively. Also, for MEA-MDEA the error are 3.9% and 7.6%, respectively.

In the first analytical solution, the loss in accuracy on the model can be compensated if the proper assumption is

applied. The first analytical solution either can be employed by itself providing quite good results, or it can help getting convergence in combination with the numerical solution.

The relative error between second analytical solution method and numerical solution is negligible, and in some cases is equal to zero. This method can be applied for all range of MEA, DEA and MEA-MDEA concentrations. The relative error of second analytical method and numerical solution with plant data for CO<sub>2</sub> in sweet gas (Tray 7) and rich MEA (Tray 1) are 2.3% and 4.2%. These errors for DEA solution are 3.8% and 4.5%, respectively. Also, for MEA-MDEA they are 2.1% and 5.3%, respectively.

Polynomial Solution has good agreement with numerical solution and plant data with eight terms series.

Accuracy of the polynomial method is lost if we use lower than four terms in series.

## 7. CONCLUSION

In this work, analytical and numerical solution for sour gas absorption by aqueous alkanolamine is compared with pilot plant data. Good agreement between experimental data and simulation results is obtained for gas sweetening operation. Analytical model enables accurate design of complete column. The analytical method converges faster than numerical method. The analytical solution can be employed as initial guess in order to numerical solution converge rapidly.

## 8. NOMENCLATURE

### A. Notation

- N* Molar flux (kmol m<sup>-2</sup>h<sup>-1</sup>)  
*D* Molecular diffusivity (m<sup>2</sup>h<sup>-1</sup>)  
*C* Molar concentration (kmol m<sup>-3</sup>)  
*y* Mole fraction in the gas stream  
*k* Reaction rate coefficient (m<sup>3</sup>kmol<sup>-1</sup>h<sup>-1</sup>)  
*K<sub>g</sub>* Gas mass transfer coefficient for absorbed component (kmol m<sup>-2</sup>h<sup>-1</sup> bar<sup>-1</sup>)  
*P* Pressure (bar)  
*H* Henry's coefficient (bar m<sup>3</sup>kmol<sup>-1</sup>)  
*z* Axial coordinate (m)  
*x* Mole fraction in the liquid stream  
 $\delta$  Liquid film thickness (m)  
*hf* Froth height on the plate (m)  
*V* Volume flow rate (m<sup>3</sup>h<sup>-1</sup>)  
 $\rho$  Density (kg/m<sup>3</sup>)  
 $\mu$  Viscosity (mPa.s)  
*hw* Weir height (m)  
 $N_{A,y^0}$  Molar flux of component A at interfacial area (kmol m<sup>-2</sup>h<sup>-1</sup>)  
*Z<sub>t</sub>* Average width of liquid flow on the plate (m)  
**Subscripts**  
*I* Gas-liquid interface  
*b* Bulk

- i* Component
- g* Gas
- l* Liquid
- A* CO<sub>2</sub>
- B* Amine

TABLE-1: FEED GAS CONDITION

Flow rate(kg mole/h)	13.6	
Pressure(kpa)	420.4	
Temperature(°C)	40	
Molecular weight	22.44	
Composition mole fraction	H <sub>2</sub> O	2 x10 <sup>-6</sup>
	CH <sub>4</sub>	0.696073
	C <sub>2</sub> H <sub>6</sub>	0.183028
	C <sub>3</sub> H <sub>8</sub>	0.02727
	n-C <sub>4</sub> H <sub>10</sub>	0.016362
	i-C <sub>4</sub> H <sub>10</sub>	0.010908
	n-C <sub>5</sub> H <sub>12</sub>	0.000873
	i-C <sub>5</sub> H <sub>12</sub>	0.000764
	n-C <sub>6</sub>	0.000185
	n-C <sub>7</sub>	0.000131
CO <sub>2</sub>	0.064405	

TABLE-2: ABSORBER COLUMN DETAIL

No. of tray	7
Tray space(cm)	25
Column inner diameter (cm)	19.3
Type of tray	sieve
Tray orifice diameter (mm)	4
Weir height (cm)	2.2
Weir length(cm)	14.3
Down comer cross-section area (cm <sup>2</sup> )	31.3695
Tray area(cm <sup>2</sup> )	292.5529
Tray active area(cm <sup>2</sup> )	261.1834

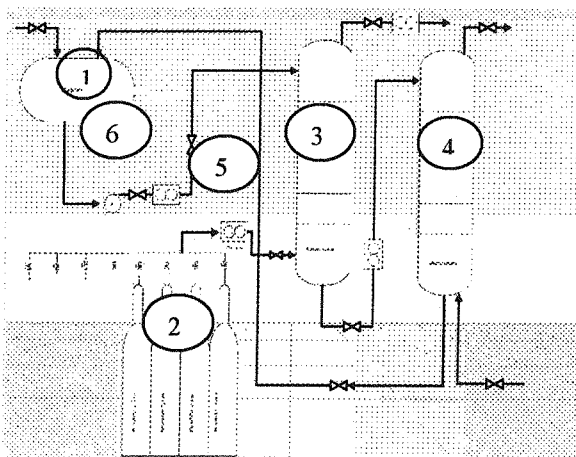
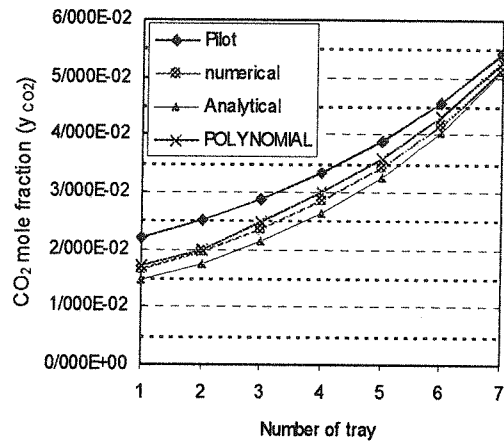
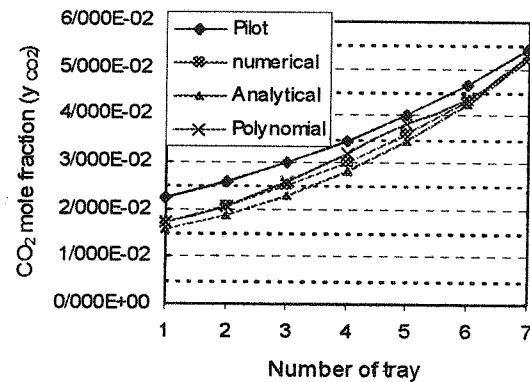


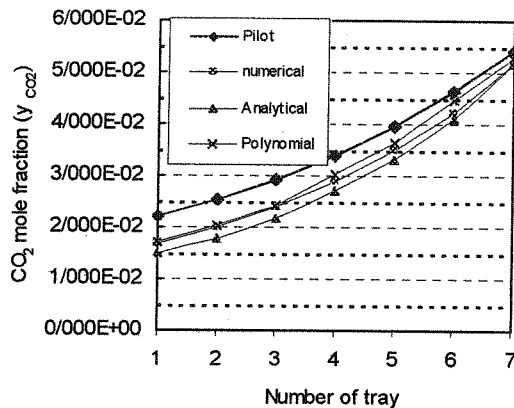
Figure-1: Schematic flow diagram of gas sweetening pilot  
 1) surge drum 2) gas cylinder 3) absorption column 4) stripper column 5) flow meter 6) pump



(a)

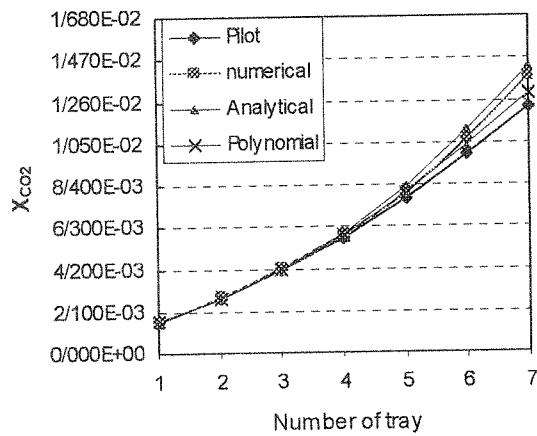


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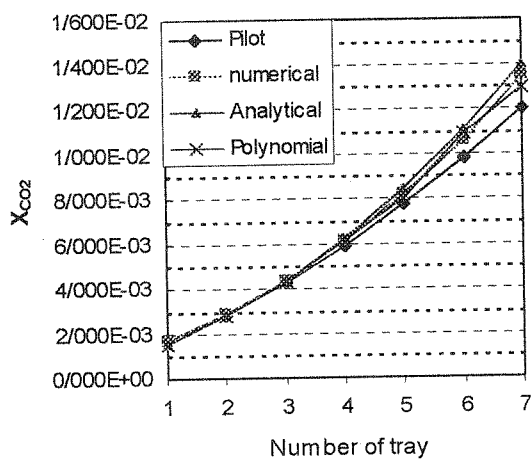


(c)

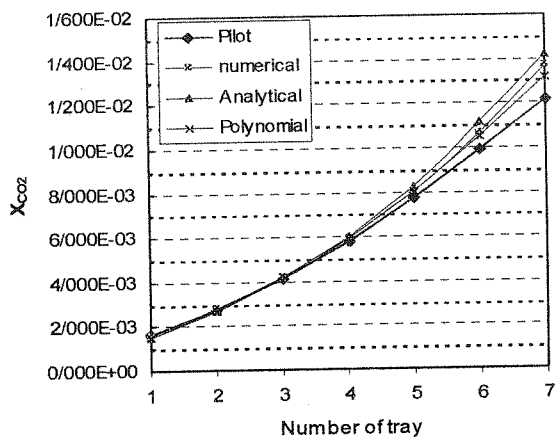
Figure 2- CO<sub>2</sub> mole fraction profile in gas Phase with: a) MEA b) DEA c) MDEA-MEA absorption



(a)

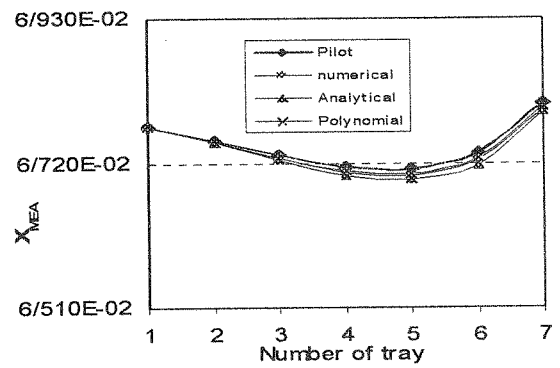


(b)

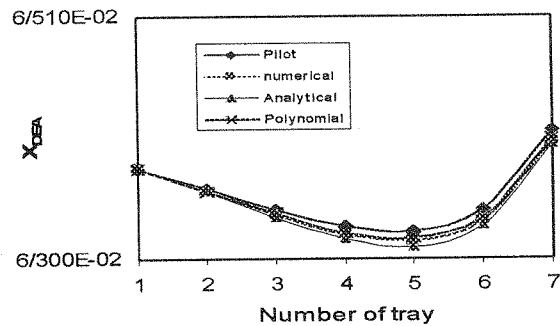


(c)

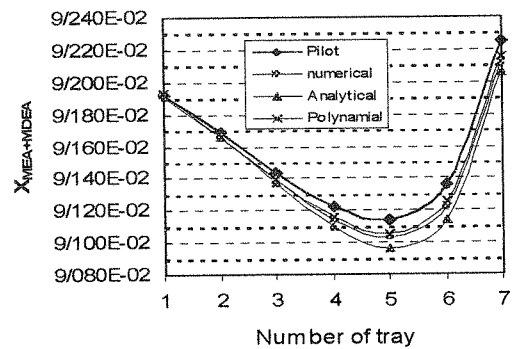
Figure 3- CO<sub>2</sub> mole fraction profile in liquid Phase with: a) MEA b) DEA c) MDEA-MEA absorption



(a)

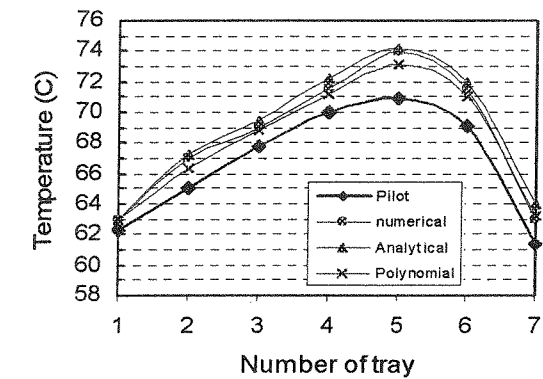


(b)

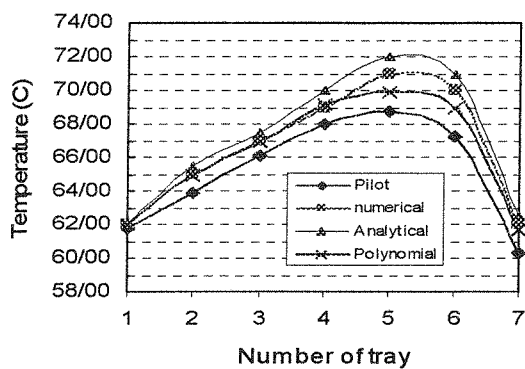


(c)

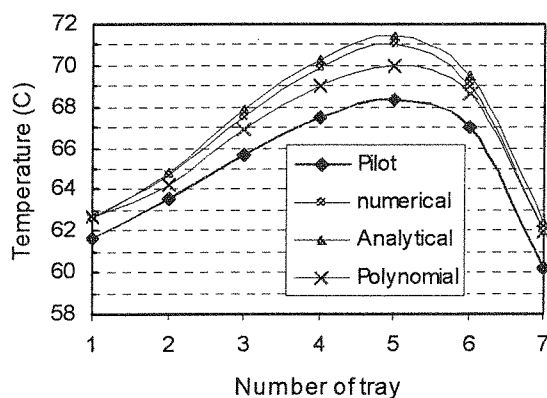
Figure 4- Alkanolamine mole fraction profile with: a) MEA b) DEA c) MDEA-MEA absorption



(a)



(b)



(c)

Figure 5- Temperature profile: a) MEA b) DEA c) MDEA-MEA absorption

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