

# Packed Amine Absorber Simulation Tracks Plant Performance<sup>1</sup>

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## ABSTRACT

The most serious practical difficulty with simulating packed columns in gas treating applications is probably the translation of theoretical HETPs into the physical depth of a specific packing. Translation is made extraordinarily difficult by the facts that (i) HETPs are quite different for CO<sub>2</sub> and H<sub>2</sub>S, (ii) they vary markedly with local gas composition, and (iii) they show extreme variation with changing conditions from point to point in the column. In gas treating applications HETP is impossible to correlate with process and local column conditions.

The engineer is invariably left with the question of how to translate simulated theoretical stages into practical packed bed depths. If experience is lacking (and it usually is), there really is no solid answer to the question. For new construction the result is frequently the installation of more packing than needed, but with the risk that the tower is under-staged. In selective H<sub>2</sub>S treating applications excessive total bed depth typically results in much poorer selectivity than could have been achieved. In some ways a revamp situation is even worse because the available column height may be insufficient for a reasonable choice of packing to meet process specifications at all, but the answer doesn't become known until the plant is started up and the column becomes a problem tower. The shortcoming lies with equilibrium stage simulation, even when an effort is made to account for reaction kinetics. No matter what adjectives are used to increase its appeal, an equilibrium stage simulation yields theoretical stages. They are simply incapable of simulating actual depths of packing, something that will remain forever beyond their scope. The only way to simulate a packed column is to apply a mass and heat transfer rate-based model to physical depths of real packing, and thereby avoid the translation issue altogether.

This paper briefly introduces the mass transfer rate basis for modeling, but it focuses primarily on a comparison between actual plant performance tests on a packed column and the results of mass transfer rate simulation of the column, using the actual packed depth and specific packing type and size. It is shown that because the mass and heat transfer rate approach refers only to equipment specifics and not to hypothetical stages, no translation is needed to go from HETPs to actual depths of packing. This allows us to look at performance sensitivity to a variety of packing-specific parameters and to select the most effective packing for the application. The solution to the dilemma of translating theoretical stages into actual packed bed depths is detailed mass and heat transfer rate-based simulation, a completely new approach to simulating complex chemically-reactive separations like amine treating.

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## 1. Introduction

Removal of the acid gases  $\text{CO}_2$  and  $\text{H}_2\text{S}$  from gas streams has been an important separation process for over half a century. Gas streams requiring treatment include natural gas, ammonia synthesis gas, and gases in petrochemical plants and oil refineries. Almost all acid gas removal processes currently employed involve the absorption of one or more acid components into chemically reactive solvents, mainly aqueous alkanolamines, with the acid gas rich solvents subsequently regenerated thermally, usually by stripping with steam.

In selective absorption process,  $\text{H}_2\text{S}$  must be removed almost completely with very minimal absorption of  $\text{CO}_2$ . In other instances, in addition to the total removal of  $\text{H}_2\text{S}$ , we require that various amounts of  $\text{CO}_2$  be left in the treated gas. A major consideration apart from the attainable purity of the treated gas is the energy requirement for solvent regeneration. This has brought about the use of new amines, the development of blended amine technology, and the use of special additives to enhance  $\text{H}_2\text{S}$  removal.

Despite the advances in amine treating technologies, absorbers and regenerators are often designed based on experience and rules of thumb. This inadequate design methodology, in most cases if not all, results in overdesign, consumes excessive energy and usually fails to reap fully all the benefits offered by advances in amine technology.

An understanding of how amine treating units work can most easily be reached by concentrating on the process fundamentals, namely, the physics and chemistry of the processes themselves, and the influence of equipment characteristics on their operation. This means taking a truly mechanistic approach in terms of mass and heat transfer rate fundamentals at the microscopic level.

This paper uses the mass and heat transfer rate approach to simulate contacting equipment used in amine-treating units. The next section provides an overview of the rate-based nonequilibrium model. This then allows us to investigate the operation of packed absorbers in Section 3.

## 2. The Mass and Heat Transfer Rate Model Explained

### 2.1. Equilibrium Model and Its Drawbacks

Equilibrium stage models treat amine units in much the same way as used for hydrocarbon absorbers and distillation columns, i.e., they use HETP (**H**eight **E**quivalent to a **T**heoretical **P**late) to translate from ideal stages to real heights of packing. These models operate under the assumption that vapor and liquid phases attain thermodynamic equilibrium and then resort to correction factors such as HETP to account for any departure from equilibrium. These correction factors still assume the existence of thermal equilibrium between vapor and liquid phases. In addition, equilibrium models fail to properly take into consideration the role played by chemical reactions—the effect of reactions on mass and heat transfer processes taking place in amine systems. The lumping of all uncertainties and deficiencies into one single parameter such as HETP makes the equilibrium approach quite unsuitable for amine systems. Moreover these models can never be predictive, and they certainly are incapable of predicting the performance for a new solvent for which plant data do not exist in a form specific to the particular type of packed column being built.

## 2.2. Rate-Based Nonequilibrium Model

The drawbacks of the equilibrium approach can be addressed by considering the degree of separation to depend on the *rates* of mass and heat transfer from one phase to another. In addition, the model should also be capable of describing the complex reaction and phase equilibria and their roles on the degree of separation. The mass and heat transfer rate approach thus results in a fundamental model based on the physics and chemistry of the amine process that can be applied to packed columns under all possible operating conditions.

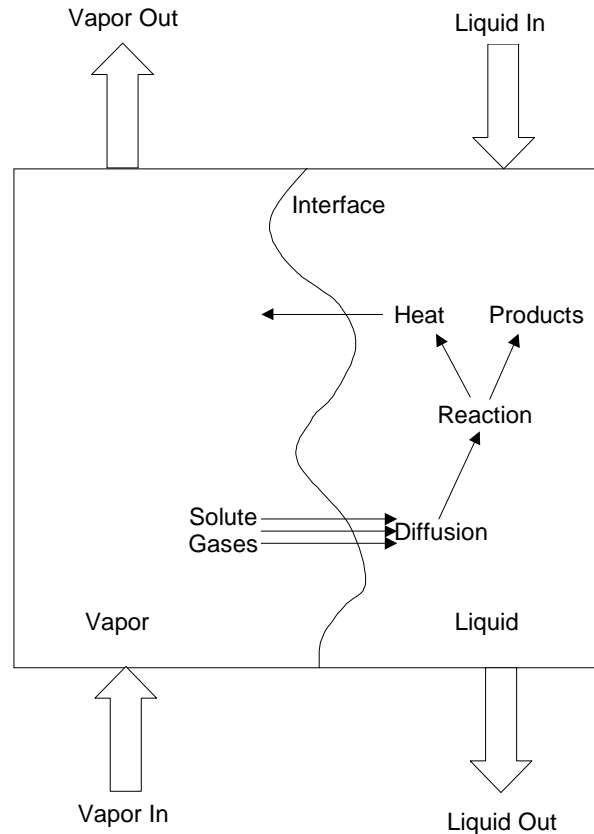
By the 1950's heat exchanger design had been put on a rational basis. Today, no one would contemplate designing a shell-and-tube exchanger, for example, without referring to the physical dimensions of the exchanger and its components (tube diameter, baffling arrangements, number of passes etc.), using the physical and transport properties of the fluids, and estimating the shell-side and tube-side heat transfer coefficients that contribute to the overall heat transfer coefficient used in the heat-transfer-rate equation. Compared with separations in columns, most heat exchangers are pretty simple because there is only a single transferring quantity, namely heat. Mass transfer is much more complicated, sometimes with a dozen or two species transferring. Until recently a theoretical (equilibrium) stage approach was the only avenue available to confront this problem. The advent of high-speed desktop computers, however, has offered the possibility of describing mass transfer operations such as distillation and absorption calculations on the same sound, detailed, mechanistic footing as heat transfer.

At the simplest level, mass and heat transfer rates depend on driving forces, transfer coefficients, and interfacial contact area:

$$Rate = (Coefficient) \times (Area) \times (DrivingForce)$$

In heat transfer, the rate is the heat transfer rate through the tube area that is driven by a (log mean) temperature difference between the shell-side and tube-side fluids. The overall heat transfer coefficient is derived from individual film coefficients on the shell- and tube-sides of the heat transfer surface, possibly including the resistance of the tube metal and scale deposits. Film coefficients depend on the fine details of the exchanger's construction, on fluid transport properties, and on flow rates. The phenomenon of mass transfer is completely analogous.

Figure 1 is a sketch of the vapor and liquid phases in contact in a small section of packing. The gas-liquid interface separates the phases. An absorbing gas dissolves into the liquid at the interface, then diffuses across a thin layer of liquid (called the diffusion layer). As it diffuses, the gas meets the reactive amine component in the solvent, reacts with it, and generates heat and reaction products such as carbamate and carbonate. Reaction products diffuse into the bulk liquid while the liberated heat of reaction heats the liquid and also transfers into the vapor. What makes the mass and heat transfer rate model such an attractive tool is the fact that the extent to which a packing element is effective in removing a species is actually calculated for the element as it really exists in the column. For example, the transfer rate of an acid gas component through the vapor towards the interface, in addition to its dependence on the diffusion coefficient of the acid gas and the driving force, depends on the state of flow of the vapor phase, i.e., the vapor hydraulics. Phase hydraulics are highly equipment specific. All the other physical and chemical steps in the overall transfer process depend on the specific component, as well as on the amine, reaction kinetics and, very importantly, the hydraulic state of the fluids being contacted. In principle all the pertinent parameters are just as calculable and determinate for

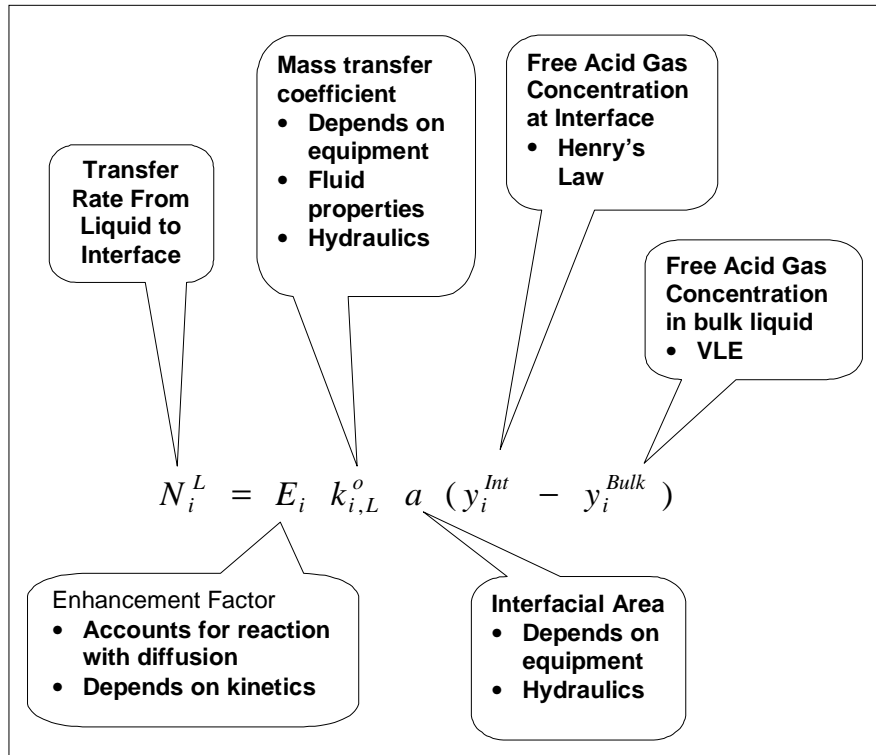


**Figure 1 Schematic of absorption accompanied by diffusion with reaction**

mass transfer as they are for the more familiar heat exchange process. This is illustrated further in the equation of Figure 2 which describes the physical processes acting on an acid gas component once it enters the liquid phase. The model equations apply to vapor and liquid phases in each segment of packing (essentially differential heights of packing) in a column.

There are some features of the mass transfer rate equation for the liquid phase that are worth further elaboration:

1. The model directly calculates the transfer rates of the species (and heat) from one phase to the other. The exiting liquid and vapor phases are not assumed to be in equilibrium.
2. Actual equipment design details and parameters are directly involved in determining the transfer rates, in other words, in determining the degree of separation. Equipment details are required in order to evaluate the mass transfer coefficients and interfacial contact area. These parameters depend on the hydraulics of the contacting device and on the physical and transport properties of the fluids (phase density, surface tension, viscosity, diffusion coefficients). Obviously, hydraulics depend very much on the type and details of the contacting device. For packing, the packing style (random vs. structured), type (specific rings, saddles, other shapes), size, and material will affect mass transfer performance.
3. Chemical reaction kinetics enhances the mass transfer rates. The reaction acts as a sink for dissolved acid gas, it steepens concentration gradients, and it speeds up diffusion of the acid gas through the liquid. The enhancement factor is a well-defined and readily calculated



**Figure 2 Absorption rate of component "i" into the liquid**

quantity; it is not a correction factor and it should neither be grouped nor be associated with HETPs.

4. Phase equilibrium is an important factor because it determines the relationship between the concentrations of the acid gases in the bulk vapor and liquid phases through the vapor-liquid equilibrium at the interface separating the two phases.
5. Although not explicitly shown in the figure, component balances must still be closed, but now around each phase rather than around the packed segment as a whole. Further modeling details can be found elsewhere (Sivasubramanian, 1985; Vickery et al., 1988; Weiland et al., 1985; Weiland and Dingman, 1995).

The availability of the results of an enormous amount of fundamental laboratory and large pilot-scale research on mass transfer provides a sound foundation under the mass and heat transfer rate model and allows it to be used quite effectively to predict column (and plant) performance without recourse to experience with similar plants. There are no adjustable parameters and there are no translations from theoretical stages to real tower internals. As shown in the next section, the mass and heat transfer rate-based model is a faithful mirror of the realities of processing in actual plant equipment—it maintains a one-to-one correspondence between simulation and the actual plant.

### 3. Simulation of Absorbers Using the Rate-Based Model ProTreat™

This section illustrates the comparison between simulation results and the plant data for two cases, namely selective H<sub>2</sub>S removal using MDEA and CO<sub>2</sub> removal using MEA.

### 3.1. Selective H<sub>2</sub>S Removal Using MDEA

In this example, selective treating means removing as *much* of the H<sub>2</sub>S from a gas as possible, while simultaneously removing as *little* of the CO<sub>2</sub> as possible using MDEA as the solvent. The absorber consists of 3 sections of stainless steel Hy-Pak random packing in a 3.2-ft diameter column. The operating conditions of the absorber are provided in Table 1.

**Table 1 Tail gas analysis and stream conditions**

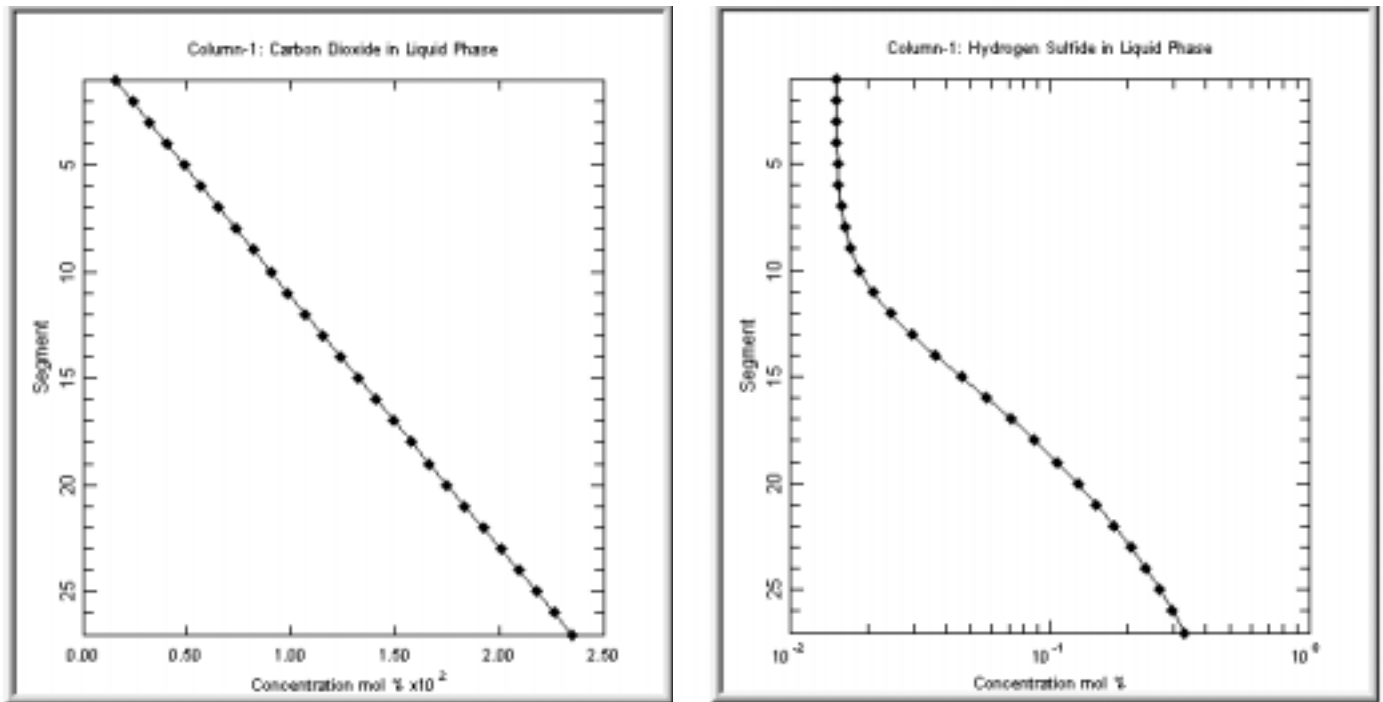
<b>Tail Gas</b>	
H <sub>2</sub> S (%)	1.5
CO <sub>2</sub> (%)	3.0
N <sub>2</sub> (%)	95.5
H <sub>2</sub> O saturation (%)	100
Temperature (°F)	100
Pressure (psig)	0.5
Flow (MMscfd)	5
<b>Solvent</b>	
Temperature (°F)	100
Pressure (psig)	5
Flow (USgpm)	120
MDEA total concentration (wt%)	35
H <sub>2</sub> S loading	0.002
CO <sub>2</sub> loading	0.0001

The sour gas stream contains 1.5% H<sub>2</sub>S and 3% CO<sub>2</sub> (H<sub>2</sub>S:CO<sub>2</sub> ratio of 1:2), a low enough H<sub>2</sub>S/CO<sub>2</sub> ratio that it can be treated selectively with MDEA, with a substantial part of the CO<sub>2</sub> slipping through the column. The results of the mass and heat transfer rate-based simulation of a column packed with 27 feet of Hy-Pak are provided in Table 2 and in Figures 3 and 4.

**Table 2 Comparison of simulation results with plant data**

<b>Treated Gas</b>	
H <sub>2</sub> S (ppmv)	10.4
CO <sub>2</sub> (%)	2.76
H <sub>2</sub> S absorption (%)	99.9
CO <sub>2</sub> slip (%)	96.4
<b>Rich Liquid</b>	
H <sub>2</sub> S loading	0.044
CO <sub>2</sub> loading	0.0031
<b>Plant Data</b>	
H <sub>2</sub> S in treated gas (ppmv)	10-15

The column is slipping about 96.4% of the CO<sub>2</sub> while achieving the target of 10-15 ppmv H<sub>2</sub>S. The plant data is about 10-15 ppmv H<sub>2</sub>S and the simulated value is 10.4 ppmv, in excellent agreement with the plant data.



**Figure 3 Mole % CO<sub>2</sub> (left) and H<sub>2</sub>S (right) in the liquid phase of a contactor using 35 wt% MDEA to treat gas containing 1.5% H<sub>2</sub>S and 3% CO<sub>2</sub>. The column is packed with 27 feet of 1.5 inch Hy-Pak and each 'segment' actually contains one foot of packing. Note the logarithmic mole fraction scale for H<sub>2</sub>S, but the linear scale for CO<sub>2</sub>.**

It is seen from Figure 3 that CO<sub>2</sub> in the liquid phase exhibits a linear profile, whereas, H<sub>2</sub>S changes exponentially in the bottom of the column. The absorption of CO<sub>2</sub> (whatever little) occurs throughout the column while most of H<sub>2</sub>S gets absorbed in the bottom of the column. The linear profile for CO<sub>2</sub> can be attributed to the slow reaction of CO<sub>2</sub> with MDEA, i.e., the CO<sub>2</sub> mass transfer rate is essentially unassisted by reaction. For H<sub>2</sub>S, the top of the column is used to reduce its content to ppm level.

The vapor temperature (Figure 4) exhibits a bulge at the bottom of the absorber. The bulge is due to heating of the cold sour gas entering the bottom of the absorber by the hot exiting rich amine. The temperature profiles also illustrate that vapor and liquid are not at thermal equilibrium.

All amines are capable of selective treating to some extent. This is because the finite-rate kinetics of reaction between CO<sub>2</sub> and the amine favors H<sub>2</sub>S absorption, while the thermodynamics always favors CO<sub>2</sub>. The less reactive the amine, the better the selectivity, a fact that is taken to its logical limit with MDEA which does not form reaction products with CO<sub>2</sub> at all. Table 3, for operating conditions specified in Table 1, shows the comparison between MEA, DEA and MDEA for the selective H<sub>2</sub>S removal operation.

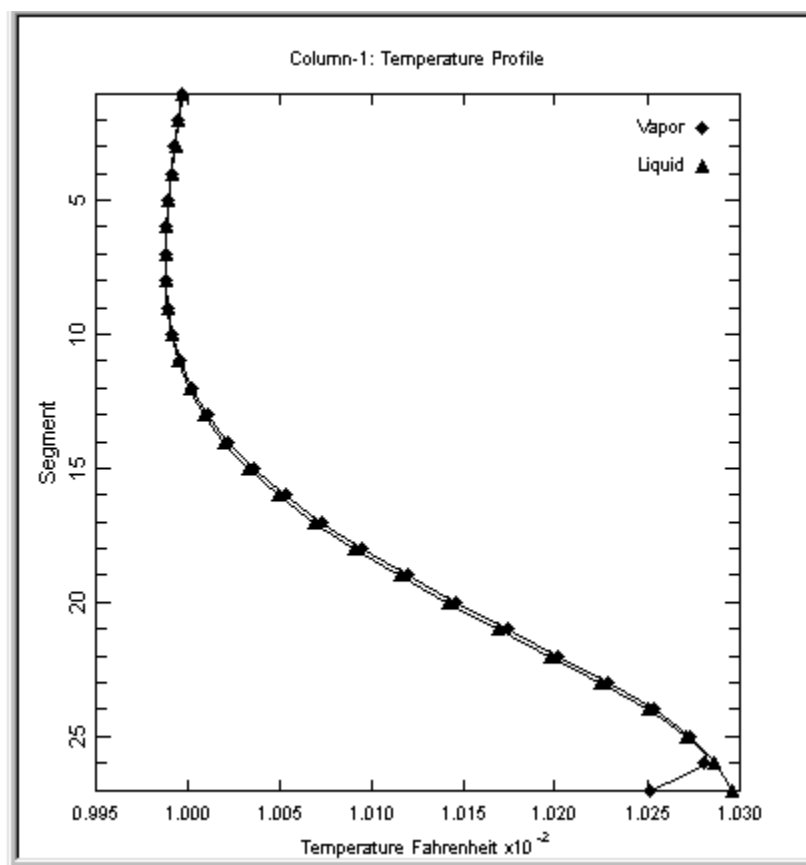


Figure 4 Simulated liquid (▲) and vapor (◆) temperature profiles.

Table 3 Comparison between MEA, DEA and MDEA

Solvent (35 wt%)	CO <sub>2</sub> slip (%)	H <sub>2</sub> S (ppmv)
MEA	4.3	1.5
DEA	34.8	5.7
MDEA	96.4	10.4

As seen from Table 3, for same CO<sub>2</sub> and H<sub>2</sub>S lean amine loadings, MEA is suitable for complete removal of CO<sub>2</sub> and H<sub>2</sub>S (low H<sub>2</sub>S selectivity) while MDEA is suitable for selective H<sub>2</sub>S removal (high H<sub>2</sub>S selectivity). DEA exhibits intermediate behavior and is commonly used with MDEA in formulated solvents to attain high H<sub>2</sub>S selectivity and *controlled* CO<sub>2</sub> slip.

The reaction between H<sub>2</sub>S and the solvent is one of simple ionic dissociation:



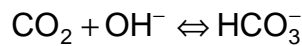
The extent of the reaction is influenced by the solution pH and, of course, this is controlled largely by the alkaline amine. In fact, the amine is a substantial sink for the H<sup>+</sup> ions formed by



H<sub>2</sub>S absorption and dissociation. The ionization reaction is instantaneous but readily reversed just by a shift in pH. When CO<sub>2</sub> reacts with an amine, on the other hand, it forms a carbamate, and the carbamate formation reaction is not nearly so easily reversed:



Carbamate formation is possible only with primary and secondary amines, i.e., with amines having labile hydrogen. The faster that dissolved, molecular<sup>2</sup> CO<sub>2</sub> reacts with the amine, the faster it is removed from solution, the steeper its concentration gradient *near* the interface, the faster its transfer rate *away from* the interface and, therefore, the faster it absorbs. MDEA does not form reaction products with CO<sub>2</sub>. However, CO<sub>2</sub> reacts with the hydroxyl ion formed by the dissociation of water, and MDEA is known to be a catalyst for this reaction:



MDEA is also a sink for the coproduced H<sup>+</sup> ions. Thus, as far as the chemistry is concerned, MDEA achieves its selectivity by being non-reactive with CO<sub>2</sub> and is, at best, a catalyst for the CO<sub>2</sub> hydrolysis reaction. It has a number of other desirable properties for amine treating, including a relatively lower enthalpy of reaction with CO<sub>2</sub>. But by being nonreactive and, therefore, allowing substantial amounts of CO<sub>2</sub> slip (i.e., CO<sub>2</sub> non-absorption), more amine is available for H<sub>2</sub>S pickup and solvent rates can be greatly reduced. With reduced solvent rates come lower reboiler heat loads, reduced solvent pumping requirements, and smaller columns (both height and diameter!). But choice of amine is not the only parameter one can use to obtain and control selectivity. The mass transfer rate model teaches us that equipment selection might be an important element, too. How can this be?

The mass transfer rate of an acid gas from the bulk vapor to the interface, and from the interface to the bulk liquid is given, respectively, by the following two expressions:

$$\begin{aligned} \text{Rate} &= k_{G,i} a (y_i^{\text{bulk}} - y_i^{\text{Int}}) \\ &= E_i k_{L,i}^o a (x_i^{\text{Int}} - x_i^{\text{bulk}}) \end{aligned}$$

(The rates must be equal because what leaves the vapor must enter the liquid.)

Kinetics affects liquid-phase mass transfer through the enhancement factor, *E*. The contacting equipment affects both the liquid and vapor phase mass transfer through the mass transfer coefficients *k<sub>G</sub>* and *k<sub>L</sub>* and the interfacial area, *a*. In amine treating, H<sub>2</sub>S absorption is gas-phase controlled while CO<sub>2</sub> absorption is controlled by liquid-phase resistance. It also happens that the liquid flow on trays is highly turbulent but the liquid flow over packings is relatively quiescent. Therefore, choice of tower-internals type and design can be expected to affect not only selectivity but also the ability of the column to treat the gas satisfactorily.

Table 4, for the operating conditions specified in Table 1, shows the comparison between different types and sizes of packings. All comparisons have been made for the same height of the packing in the column. As illustrated below, the mass and heat transfer rate-based model, in addition to properly accounting for the physics and chemistry of processes, includes the effect of column internals on the achieved degree of separation, in this case the amount of H<sub>2</sub>S in the treated gas and CO<sub>2</sub> slippage from the column.

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<sup>2</sup> This is the *only* form of CO<sub>2</sub> that exists in, and can therefore transfer from, the vapor phase. Therefore, it is the form pertinent to CO<sub>2</sub> mass transfer, in both absorption and stripping.

**Table 4 Comparison between different packings**

Packing	CO <sub>2</sub> slip (%)	H <sub>2</sub> S (ppmv)
1.5" Hy-Pak	96.4	10.4
3" Hy-Pak	98.0	69.6
#1.5 Mini-rings	94.9	10.4
1.5" Pall rings	95.9	10.7

The effect of column operating variables such as amine concentration and circulation rate on the efficiency of the absorber is provided in Table 5. As seen below, for a given circulation rate, as the MDEA concentration is increased from 30 to 40 wt%, the CO<sub>2</sub> slippage increases. The reason is that the liquid viscosity increases with increasing concentration of amine resulting in lower liquid-phase mass transfer coefficients, hence, lower CO<sub>2</sub> absorption. Similar behavior is exhibited by H<sub>2</sub>S. As expected, the increase in circulation rate produces increased CO<sub>2</sub> and H<sub>2</sub>S absorption.

**Table 5 Comparison between different amine concentrations and flow rates using MDEA as the solvent**

Amine (wt%)	Flow (USgpm)	CO <sub>2</sub> slip (%)	H <sub>2</sub> S (ppmv)
30	100	96.2	9.7
30	120	96.1	8.6
30	140	95.9	8.4
35	100	96.5	13.6
35	120	96.4	10.4
35	140	96.3	9.9
40	100	96.8	30.4
40	120	96.8	14.1
40	140	96.7	12.0

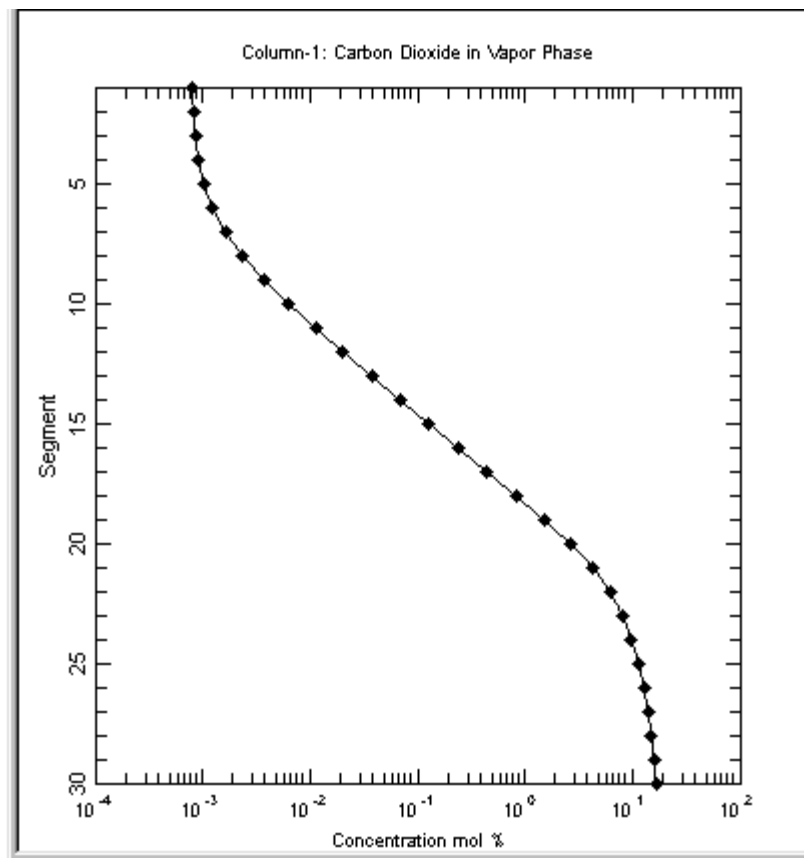
### 3.2. CO<sub>2</sub> Removal Using MEA

In this example, MEA is used to remove CO<sub>2</sub> from gases containing N<sub>2</sub>, H<sub>2</sub>, Ar, CH<sub>4</sub> etc. The absorber consists of 31.5 feet of # 2 Cascade Mini-Rings (CMR) in a 9.5-ft diameter column. The operating conditions (Zehnder et al., 1990) of the absorber are provided in Table 6.

**Table 6 Sour gas analysis and stream conditions**

<b>Sour Gas</b>		
CO (lbmol/hr)	18	0.15 %
CO <sub>2</sub> (lbmol/hr)	2,123	17.34 %
Hydrogen (lbmol/hr)	7,185	58.68 %
Nitrogen (lbmol/hr)	2,476	20.22 %
Argon (lbmol/hr)	29	0.24 %
Methane (lbmol/hr)	39	0.32 %
Water (lbmol/hr)	374	3.05 %
Temperature (°F)	196	
Pressure (psia)	371.7	
<b>Solvent</b>		
Temperature (°F)	125	
Pressure (psia)	369.3	
MEA (lbmol/hr)	7,130	31.6 wt%
H <sub>2</sub> O(lbmol/hr)	49,188	
CO <sub>2</sub> (lbmol/hr)	1,301	0.182 loading

The results of the mass and heat transfer rate-based simulation of the CMR packed column are provided in Figures 5, 6 and 7 and in Table 8.

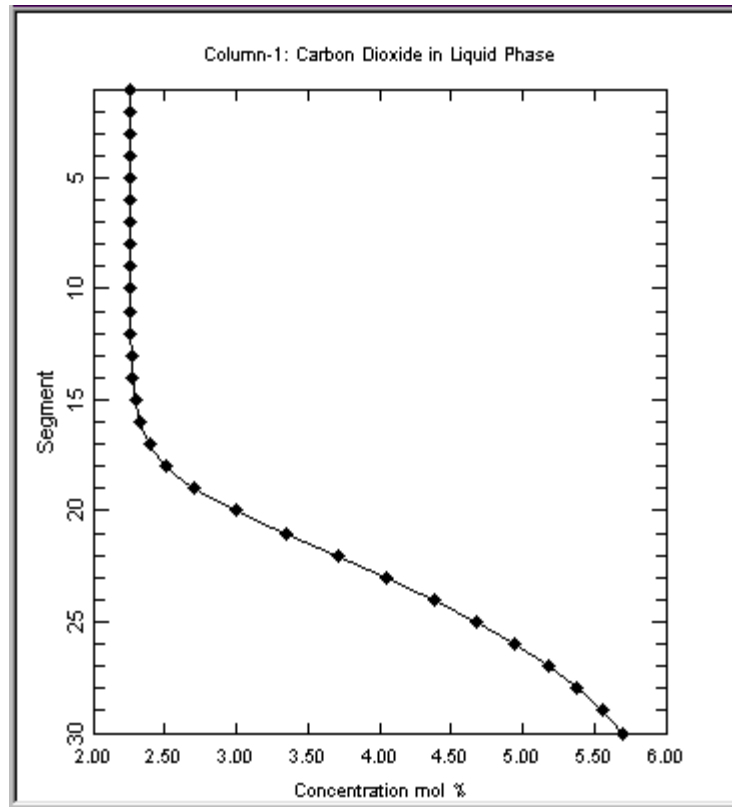


**Figure 5 Mole % CO<sub>2</sub> in the vapor phase of a contactor using 31.6 wt% MEA. The column is packed with 31.5 feet of #2 inch Cascade Mini-Rings.**

**Table 8 Comparison of simulation results with plant data**

<b>Treated Gas</b>	
CO <sub>2</sub> (ppmv)	8.1
<b>Rich Liquid</b>	
CO <sub>2</sub> loading	0.48
Temperature (°F)	207.8
<b>Plant Data</b>	
CO <sub>2</sub> in treated gas (ppmv)	5
Rich amine temperature (°F)	197

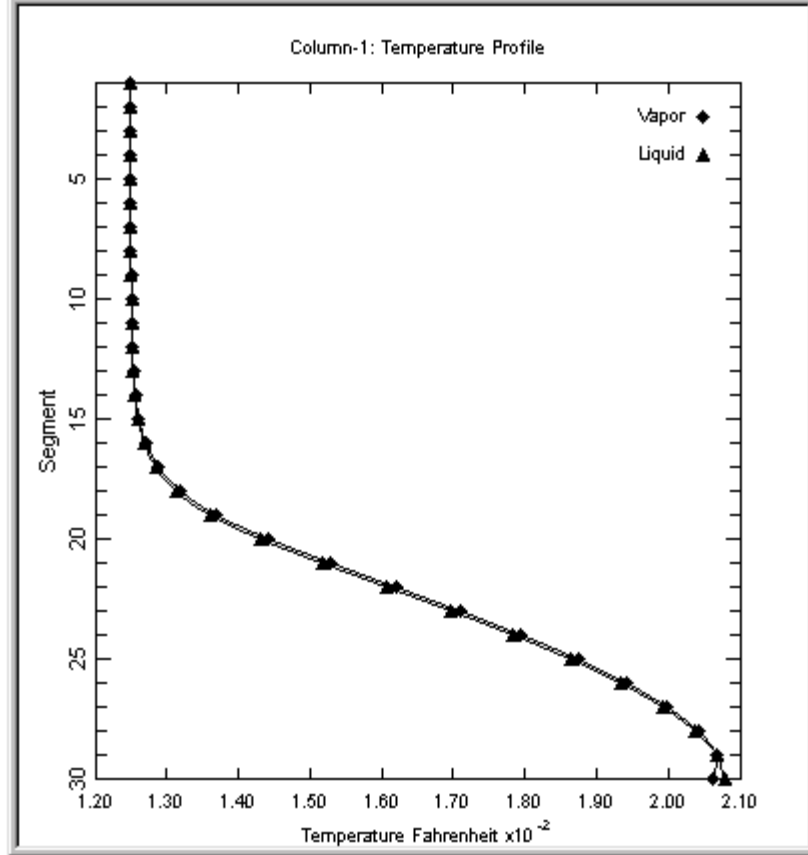
The plant data is about 5 ppmv CO<sub>2</sub> and the simulated value is 8.1 ppmv. As seen from Figure 6, CO<sub>2</sub> absorption is confined to the bottom of the absorber. The top of the absorber is used to reduce the acid gas content to ppm level. The vapor and liquid temperatures (Figure 7) are constant at the top of the column and increase at the bottom of the column, consistent with the absorption of CO<sub>2</sub> in the bottom section of the column. The concentration of CO<sub>2</sub> in the treated gas is directly controlled by the lean loading of the amine solution entering the absorber. The effect of lean loading on the treated gas is shown in Table 9.



**Figure 6 Mole % CO<sub>2</sub> in the liquid phase of a contactor using 31.6 wt% MEA. The column is packed with 31.5 feet of #2 inch Cascade Mini-Rings.**

**Table 9 Effect of lean loading on CO<sub>2</sub> content**

CO <sub>2</sub> loading (mol/mol)	CO <sub>2</sub> in treated gas (ppmv)
0.1	1.63
0.15	4.54
0.182	8.1
0.25	61.8



**Figure 7 Simulated liquid (▲) and vapor (◆) temperature profiles.**

#### 4. Summary

This paper has attempted to provide an overview of rate-based nonequilibrium modeling and its ability to handle amine treating systems. The following conclusions can be drawn:

- The detailed equipment design parameters of packed columns play just as important a part in setting absorber performance as do kinetics and thermodynamics.

- Comparison with observations of plant behavior show that a mass and heat transfer rate-based simulator mirrors field performance with uncanny accuracy.
- Rate-based models are capable of predicting packed column absorber performance without reliance on operating experience.

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