Mass Transfer Characteristics in Structured Packing for CO₂ Emission Reduction Processes

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Acid gas treating and CO₂ capture from flue gas by absorption have gained wide importance over the past few decades. With the implementation of more stringent environmental regulations and the awareness of the greenhouse effect, the need for efficient removal of acid gases such as CO₂ (carbon dioxide) has increased significantly. Therefore, additional effort for research in this field is inevitable. For flue gas processes the ratio of absorption solvent to gas throughput is very different compared to acid gas treating processes owing to the atmospheric pressures and the dilution effect of combustion air. Moreover, in flue gas applications pressure drop is a very important process parameter. Packing types are required that allow for low pressure drop in combination with high interfacial areas at low liquid loading per square meter. The determination of interfacial areas in gas−liquid contactors by means of the chemical method (Danckwerts, P. V. *Gas-liquid reactions*; McGraw-Hill: London, 1970) has been very frequently applied. Unfortunately, many of the model systems proposed in the literature are reversible and therefore this condition possibly is not met. Versteeg et al. (Versteeg, G. F.; Kuipers, J. A. M.; Beckum, F. P. H.; van Swaaij, W. P. M. *Chem. Eng. Sci.* **1989,** *44,* 2292) have demonstrated that for reversible reactions the conditions for the determination of the interfacial area by means of the chemical method are much more severe. In a study by Raynal et al. (Raynal, L.; Ballaguet, J. P.; Berrere-Tricca, C. *Chem. Eng. Sci.* **2004,** *59,* 5395), it has been shown that there is a dependency of the interfacial area on the packing height. Unfortunately, most model systems used, e.g., CO₂−caustic soda (as used by Raynal et al.), are much more complex and consist of (a set of) reversible reaction(s). The natures of these systems make the conditions at which the interfacial area can be determined much more severe and put more limitations on the process conditions and experimental equipment than a priori can be expected. Therefore, an extended absorption model is required to determine the conditions at which the interfacial area can be measured without detailed knowledge of the values of the liquid-side mass transfer coefficient, k_l, beforehand.

1. Introduction

Acid gas treating has gained a wide importance over the past few decades. With the implementation of more stringent environmental regulations, the need for efficient removal of acid gases such as CO₂ (carbon dioxide) and H₂S (hydrogen sulfide) has increased significantly. More specifically, the capture of CO₂ has gained substantial interest as a result of the Kyoto Protocol in order to realize stabilization of the CO₂ emission to the environment. Postcombustion removal of CO₂ from flue gas produced by power plants with, e.g., amine solutions is believed to be a very promising technology to reduce CO₂ emissions. However, at this time the costs of the available processes are about 0.50 euro per metric ton of CO₂ removed. To be economically feasible these, costs must be reduced to values below about 0.20 euro per metric ton of CO₂ removed. Therefore, additional effort for research in this field is inevitable.

In flue gas applications the ratio of absorption solvent to gas throughput differs from that of acid gas treating applications due to the atmospheric pressure and the dilution effect of air used as combustion agent. Moreover, in flue gas applications the pressure drop is a very important process parameter. It can be concluded that packing types are required that allow for low

pressure drop in combination with high interfacial areas at low liquid loading per square meter. Basically two types of packing are available: dumped and structured packing, respectively. In the present study attention is focused on structured packing as this usually has a lower pressure drop compared to dumped packing.

In a previous paper by Raynal et al.[3] results are presented on mass transfer characteristics, for example, interfacial area, for structured packing. Surprisingly, the observed interfacial areas turned out to be dependent on the position in the packed column. Differences up to 50% are reported. The measuring technique used was the well-known "absorption in the chemically enhanced region". In the present study this technique is critically evaluated.

2. Theory

In Raynal et al.[3] the absorption rate was determined in the chemically enhanced region, as characterized by

$$2 < Ha < E_{inf} \tag{1}$$

For an irreversible second-order gas−liquid reaction the following definitions apply:

$$Ha = \frac{\sqrt{k_{1,1}C_B D_{A,l}}}{k_l} \tag{1a}$$

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$$E_{\text{inf}} = 1 + \frac{D_{\text{B,l}} C_{\text{B,l}}}{D_{\text{A,l}} C_{\text{A,l}}^{i}} \tag{1b}$$

Strictly spoken, this boundary condition is only applicable for the combination of absorption with irreversible chemical reaction(s). In the open literature there are two model systems frequently used for the determination of interfacial areas in gas–liquid contactors: (a) the oxidation of sulfite to sulfate with air; (b) the absorption of (diluted) CO_2 in aqueous caustic soda solutions.

One of the most important and major drawbacks of this so-called chemical method is that a priori information is required on the value of the liquid-side mass transfer coefficient, k_l; otherwise, the lower limit $Ha > 2$ cannot be estimated. A more suitable technique would be simultaneous measurements of $k_l a$ and a as proposed by Cents et al.[4] For more details on both systems the interested reader is referred to Danckwerts[1] and Deckwer.[5]

Raynal et al.[3] have selected the caustic soda system to determine the interfacial area of structured packing. Basically, two reactions occur in the aqueous caustic soda system:

$$CO_2 + OH^- \leftrightarrow HCO_3^- \tag{2}$$

$$HCO_3^- + OH^- \leftrightarrow CO_3^{2-} + H_2O \tag{3}$$
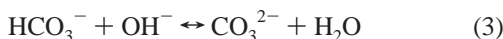
Besides these two main reactions a third, instantaneously fast, reaction occurs:

$$H_2O + H_2O \leftrightarrow H_3O^+ + OH^- \tag{4}$$
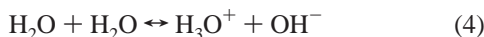
This latter reaction can be regarded to be at equilibrium compared to the reaction rate of the two aforementioned reactions and the mass transfer rate, respectively. Moreover, eqs 2–4 are reversible reactions and therefore the use of eq 1 is not completely justified beforehand. Versteeg et al.[2] demonstrated that reversibility has a substantial effect on the value of E_{inf}.

Therefore, the results of Raynal et al.[3] are evaluated by means of an absorption model in which all occurring reversible, parallel reactions are taken into account in the governing diffusion equations.[6]

2.1. Absorption Model. The problem considered is mass transfer accompanied by reversible chemical reactions of general order with respect to both reactants and products. The description of the absorption model and the numerical treatment is illustrated for a single reversible reaction:

$$A(g) + \gamma_b B(l) \leftrightarrows \gamma_c C(l) + \gamma_d D(l) \tag{5}$$
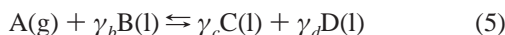
with the following overall reaction rate equation:

$$R_a = k_{\text{R},m,n,p,q}[A]^m[B]^n[C]^p[D]^q - k_{\text{R},r,s,t,v}[A]^r[B]^s[C]^t[D]^v \tag{6}$$

It is clear that for systems with parallel and/or consecutive reactions the number of reactions will increase.

Usually for gas–liquid systems most reactions can be expressed with sufficient accuracy with a reaction rate expression similar to (6), and therefore this expression was used in this model.

Moreover, for the caustic soda system Haubrock et al.[7] recently demonstrated that the use of concentrations in the kinetic expression may lead to erroneous results and it is preferred to use activities and activity coefficients instead. The latter ones could be estimated by, e.g., the Pitzer approach as proposed by Haubrock et al.[7] However, in the present study the traditional approach, concentration-based equations, will be used in combination with the physical data used by Raynal et al.[3]

The mass transfer in the gas phase is described with the stagnant film model, while for the liquid phase the penetration model is used, as the penetration model is expected to be the most realistic one in describing gas–liquid absorption.

For the penetration model the balances for each species for the phenomenon of mass transfer followed by a chemical reaction yield the following set of equations:

$$\frac{\partial[A]}{\partial t} = D_a \frac{\partial^2[A]}{\partial x^2} - R_a \tag{7}$$

$$\frac{\partial[B]}{\partial t} = D_b \frac{\partial^2[B]}{\partial x^2} - \gamma_b R_a \tag{8}$$

$$\frac{\partial[C]}{\partial t} = D_c \frac{\partial^2[C]}{\partial x^2} + \gamma_c R_a \tag{9}$$

$$\frac{\partial[D]}{\partial t} = D_d \frac{\partial^2[D]}{\partial x^2} + \gamma_d R_a \tag{10}$$

These four coupled nonlinear partial differential equations must be solved numerically, because an analytical solution method is not available. To be solved uniquely, the four nonlinear partial differential equations (7)–(10) require one initial and two boundary conditions, respectively. The initial condition is given by

$t = 0$ and $x \geq 0$,
$$[A] = [A]_0, [B] = [B]_0, [C] = [C]_0, [D] = [D]_0 \tag{11}$$

where the concentrations $[i]_0$ satisfy $R_a = 0$ in eq 6. The boundary condition for $x = \infty$ can also be derived with the assumption of chemical equilibrium for the bulk of the liquid for a given solute loading:

$t > 0$ and $x = \infty$,
$$[A] = [A]_0, [B] = [B]_0, [C] = [C]_0, [D] = [D]_0 \tag{12}$$

For detailed information on the calculation of the equilibrium composition according to eq 12, one is referred to Blauwhoff et al.[8]

The second associated boundary condition is obtained by assuming that the species B, C, and D are nonvolatile and that the flux of component A from the gas phase is equal to the flux of component A to the liquid phase. The use of the latter assumption instead of assuming that $[A] = [A]_i$ at $x = 0$ is convenient in view of the applicability of the model for cases where a part of the resistance against mass transfer is situated in the gas phase. This is the case in the asymptotic situation of instantaneously fast reactions:

$$-D_a \left(\frac{\partial[A]}{\partial x} \right)_{x=0} = k_g([A]_{g,\text{bulk}} - [A]_{g,i})$$

$$\left(\frac{\partial[B]}{\partial x} \right)_{x=0} = \left(\frac{\partial[C]}{\partial x} \right)_{x=0} = \left(\frac{\partial[D]}{\partial x} \right)_{x=0} = 0 \tag{13}$$

For detailed information on the applied numerical method used, the reader is referred to Versteeg et al.[2]

2.2. Absorber Model. The experiments of Raynal et al.[3] were carried out in a packed bed contactor with structured packing, and the operation mode was cocurrent with respect to both the gas phase and liquid phase. Gas and liquid were fed from the top of the absorber.

The absorber is simulated by a tray-to-tray procedure.[8] The absorption column is considered as a series of ideally mixed (with respect to both the gas phase and liquid phase) reactors, each corresponding to an actual tray. If the size of a "tray" is chosen small enough, the same approach can be applied for packing where each "tray" in the model represents a certain length of packing. It must be noted that this tray-to-tray approach is applicable for both counter- and cocurrent operation of the column.

For the present application process conditions and gas and liquid compositions are assumed to be uniform in each tray or segment. The mass transfer parameters (k_l, k_g, a, ρ) are considered to be equal for all trays of the absorber. Physical parameters such as viscosity, diffusivity, equilibrium constants, and partition coefficients are taken as a function of the temperature on each tray.

It is also assumed that experiments were carried out at isothermal conditions, as also stated by Raynal et al.[3] Based on this assumption the liquid and gas temperatures are known, and liquid-phase equilibrium constants, Henry coefficients, and gas- and liquid-phase diffusivities can be calculated. The liquid-phase composition, more specifically the concentrations of unreacted CO_2 and OH^-, is obtained by means of the equilibrium model.

The calculations start by taking the experimentally determined gas and liquid flows and the gas and liquid concentrations at the top of the column. These detailed data were provided by IFP, and the data were the basic information for the determination of the interfacial area presented by Raynal et al.[3] CO_2 gas-phase concentrations at tray 1, i.e., in the gas leaving tray 1, are estimated. Subsequently, gas-phase concentrations are calculated, so the absorption driving forces are now known. The molar flux (J_{CO_2}) is calculated using the absorption model (see section 2.1).

After the first estimation of the gas-phase concentration and the subsequent molar flux calculations, tray iteration proceeds using a direct substitution method for the first iteration and a Regula–Falsi procedure for the remaining iteration steps, until the following convergence criterion is satisfied within the desired accuracy (usually <0.01% error):

$$J_{CO_2}aV_{tray} = \Phi_g^{n+1}[CO_2]_g^{n+1} - \Phi_g^n[CO_2]_g^n \qquad (14)$$

The tray-to-tray procedure continues until the CO_2 specification experimentally determined by Raynal et al.[3] is reached. An additional iterative procedure using the interval halving method is used to converge the interfacial area until the desired CO_2 specification is reached exactly at tray N. The main result from the calculation is the value of the interfacial area a (m²/m³).

As the experiments of Raynal et al.[3] were carried out in a cocurrent packed bed contactor with structured packing, it was required to determine the minimum amount of trays (N) needed to ensure that the simulations describe ideal plug flow. Therefore, a number of simulations have been carried out to investigate the effect of the number (or height) of the trays on the outcome of the simulations. In Figure 1 the results of these simulations are presented.

Each section of the absorber has a height of 0.44 m. From Figure 1 it can be concluded that about 200 trays per section are required to ensure that the model outcome becomes
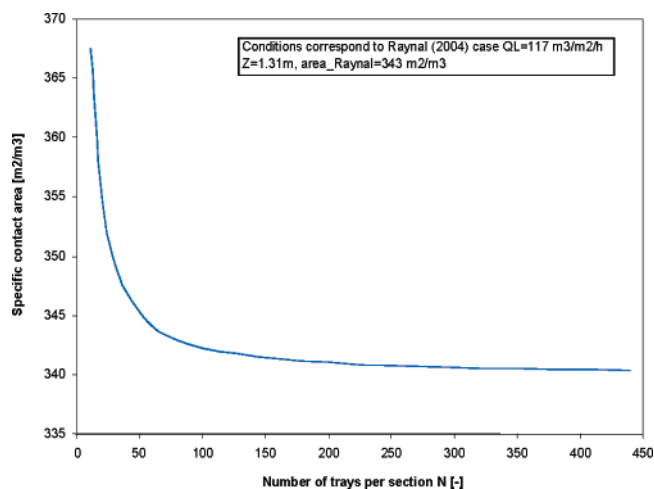

Figure 1. Minimum number of trays per section to be used in the tray-to-tray calculations.

independent of the tray height. Consequently, a maximum tray height of 0.0022 m would be optimal. In the reevaluation of the data of Raynal et al.[3] as presented in the current work, a tray height of 0.005 m (88 trays) was used as a compromise between accuracy and required calculation time. From Figure 1 it can be seen that results are still accurate within 1% when $N = 88$ is used.

3. Results

As mentioned above, one of the major drawbacks of the chemical method to determine interfacial areas in gas−liquid contactors is that the value of k_l must be know within reliable boundaries beforehand in order to fulfill the conditions stated in eq 1. Moreover, an additional issue is the pronounced effect of reversibility on the value that can be realized for E_{inf}. Versteeg et al.[2] demonstrated that the value for E_{inf} reduces substantially with increasing values of the equilibrium constant. Therefore, preliminary simulations with the present model have been carried out for a liquid flow rate of $Q_l = 117$ m³·m⁻²·h⁻¹. These simulations are carried out to check whether "an operation window" exists for the determination of the interfacial area in a packed bed reactor filled with structured packing. Therefore, in these simulations the value of k_l has been varied between 10^{-2} and 10^{-5} m·s⁻¹ (the range in which the value of k_l for this type of contactors can be expected). By means of these simulations the Ha number is varied over three decades and most likely conditions where $Ha > 2$ are encountered. Moreover, by performing simulations at different packed bed heights, effectively meaning varying the CO_2 concentration in the gas phase, also a variation in E_{inf} is realized. When the conditions of eq 1 are fully met, a range for the value of k_l must be observed in which the calculated interfacial area (e.g., based on the data provided by Raynal et al.[3]) is totally independent of k_l and a horizontal plateau must be encountered.

In Figure 2 the results of these simulations are presented. It is obvious that no k_l interval can be observed where a kind of horizontal plateau occurs and the value of the interfacial area is constant. This means that basically at this stage it must be concluded that it is not possible to fulfill the conditions stated by eq 1. Moreover, the simulations show that the derived interfacial area is also dependent on the partial pressure (i.e., in this situation the height of the packing), which must be regarded as an additional validation of the conclusion that the conditions given by eq 1 are not met. In Figure 2 it can be seen that for values $10^{-4} < k_l < 10^{-3}$ m·s⁻¹ a kind of plateau can
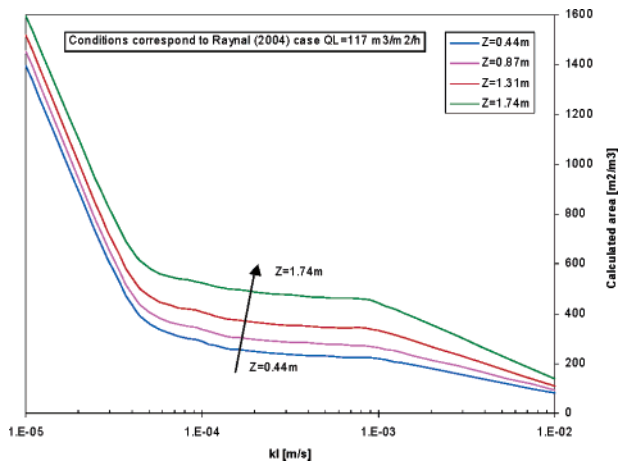
Figure 2. Relation between k_l (m/s) and a (m^2/m^3) from Raynal et al.;[3] Q_l = 117 m$^3 \cdot$m$^{-2} \cdot$h^{-1}).

be observed, indicating that this could coincide with the region $Ha > 2$; however, the calculated interfacial area is still dependent, although slightly, on the value of k_l. It must be noted, however, that for, e.g., the simulations for $Z = 0.44$ m the interfacial area increases about 35% if the value of k_l is reduced from 10^{-3} to 10^{-4} m$\cdot$s^{-1}. Also, it must be noted that the difference at identical k_l values between, e.g., $Z = 0.44$ m and $Z = 1.74$ m in the calculated area is about a factor of 2. Beforehand it is not likely to expect that this can be explained via hydrodynamic phenomena that could encountered in packed columns operated in countercurrent or cocurrent mode, respectively. In cocurrent downflow operated columns specific regions, e.g., pulse, spray, or dispersed bubble flow, are encountered which can lead to an influence of the absorber height on the observed effective interfacial area. However, it seems that this observation must be regarded as an additional motivation for the findings that conditions of eq 1 are not fulfilled. The decrease in CO$_2$ concentration, i.e., increasing the packing height, leads to higher values of E_{inf}, and therefore to higher values of the enhancement factor.

Overall, it must be concluded that it seems that, for the process conditions studied, the CO$_2$−caustic soda system cannot be applied to determine interfacial areas in packed bed reactors.

In the van Krevelen−Hoftijzer[9] plot the enhancement factor, Ea, for a second-order, irreversible gas−liquid reaction is derived as function of the Ha number and E_{inf}. From this figure the conditions as represented by eq 1 can be derived to determine the interfacial area. However, Versteeg et al.[2] demonstrated that for more complex, reversible reaction systems the actual observed enhancement factor for the complex systems could substantially deviate from the enhancement factor calculated according the relation proposed by van Krevelen and Hoftijzer.[9] Once again it must be noted that van Krevelen's relation is only applicable to irreversible reactions and that the CO$_2$−caustic soda system consists of three consecutive/parallel, reversible reactions (eqs 2−4).

Unfortunately, it is not straightforward to investigate the effect of the reversibility and the complex reaction scheme on the actual observed enhancement factor for the prevailing process conditions of the CO$_2$−caustic soda system and the presently studied packed contactor. Therefore, this so-called van Krevelen−Hoftijzer plot has been derived using the absorption model given in section 2.1; see Figure 3. In this plot the enhancement factor, Ea, for the CO$_2$−soda system is calculated as a function of the Ha number. When the conditions of eq 1 are fulfilled, the enhancement factor, Ea, should equal the value of the Ha number. In Figure 3 the line $Ea = Ha$ is also plotted.
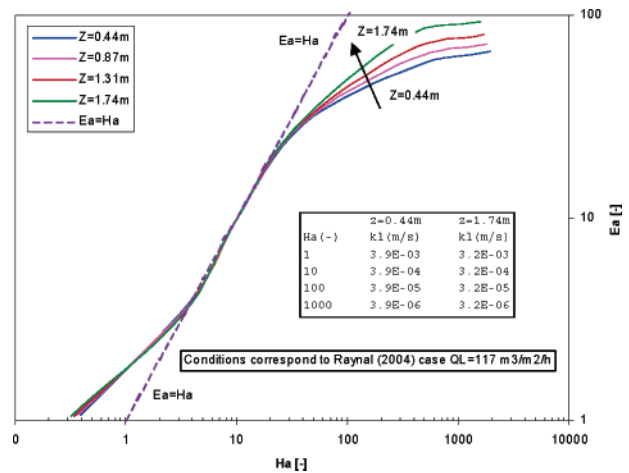

Figure 3. van Krevelen−Hoftijzer plot for the CO$_2$−caustic soda system.
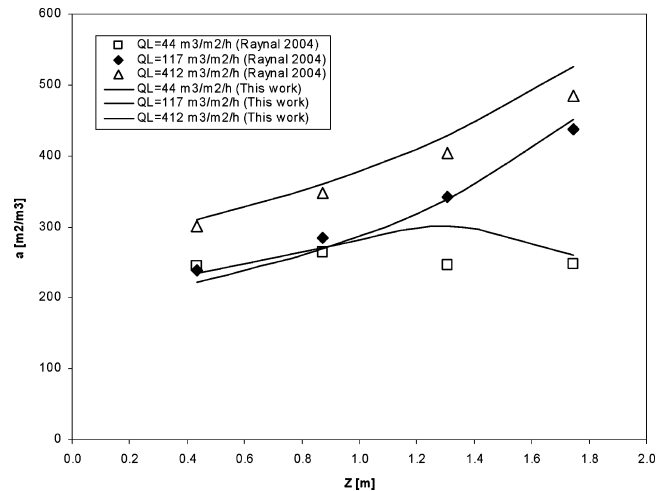

Figure 4. Comparison of the experimental and calculated relationship between the height of the packed bed z (m) and the interfacial area a (m^2/m^3).

As was shown by Versteeg et al.,[2] the value of the enhancement factor for complex, reversible systems, like the evaluated CO$_2$−soda system, is not only dependent on the Ha number and E_{inf}, irreversible, but also on the reversibility, degree of (liquid) conversion, and partial pressure of the reactive gas. All these effects are included in the results shown in Figure 3.

From this figure it can be concluded that only for the interval $4 < Ha < 20$ does the enhancement factor calculated with the used extensive absorption model coincide with the line $Ea = Ha$.

In their Figure 9, Raynal et al.[3] presented an estimation of the liquid-side mass transfer coefficient, k_l, derived from an adapted Higbie model. The k_l values for the experimental conditions studied vary between 6×10^{-4} and 7×10^{-4} m$\cdot$s^{-1} as calculated via this estimation method. This also means that the values of the corresponding Ha number are about 5−7 and therefore it should be possible to measure the interfacial areas.

These values for k_l are used in the sophisticated absorption model to calculate the interfacial area from the experimental data given. In Figure 4 the results are given and compared to the reported values in Raynal et al.[3] From Figure 4 it can be concluded that both tools used to derive the interfacial area from the experimental data given result in nearly identical values. With the restriction that the values of k_l are estimated correctly, one must conclude that the experimentally determined interfacial areas are increasing with bed height. Moreover, the value of the interfacial area becomes almost identical to that of the
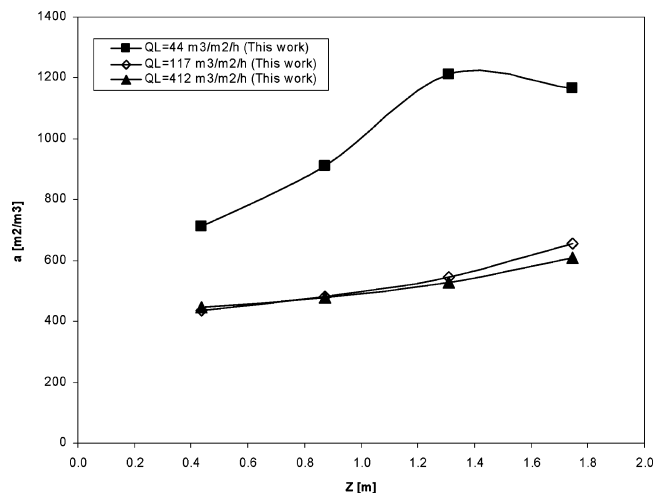
Figure 5. Calculated relationship between the height of the packed bed z (m) and the interfacial area a (m²/m³) for $k_l = 7 \times 10^{-5}$, 4×10^{-5}, and 3×10^{-5} m·s⁻¹, for flow rates 412, 117, and 44 m³·m⁻²·h⁻¹, respectively.

geometric surface of the packing, about 410 m²·m⁻³, indicating complete wetting.

However, the estimated values of the liquid-side mass transfer coefficient are relatively high. Moreover, the values of k_l seem to be almost independent of the liquid flow rate, although it is more likely to expect an increase of k_l with the flow rate. Unfortunately, no detailed information can be found for these values. Therefore, a sensitivity study has been carried out for the effect of the value of k_l on the estimated interfacial area. Simulations have been carried out with the following three values for k_l: 7×10^{-5} m·s⁻¹ (corresponds to the flow rate of 412 m³·m⁻²·h⁻¹), 4×10^{-5} m·s⁻¹ (=flow rate 412 m³·m⁻²·h⁻¹), and 3×10^{-5} m·s⁻¹ (=flow rate 412 m³·m⁻²·h⁻¹). From these results (see Figure 5) it can be concluded that the influence of the bed height is less pronounced, but also it is shown that the estimated value of the interfacial area is indeed higher than the geometric surface. This phenomenon has been observed more frequently in the open literature for cocurrent downflow operation in packed beds with dumped packing. Especially this is so if the conditions applicable to experiments are such that these are carried out in the pulse flow or dispersed bubble flow regime, respectively (Charpentier[10] and Versteeg et al.[11]). Moreover, the latter authors reported that the transition of the flow regime in cocurrently operated packed columns is dependent on the length of the packing height, at least for dumped packing.

Generally it must be concluded that, although the use of the chemical method for the determination of interfacial areas in gas−liquid contactors seems simple and straightforward as long as the conditions of eq 1 are obeyed, the actual applicability is severely restricted.

As long as the model system consists of a simple first or second reaction gas−liquid reaction, the chemical method can be applied. Unfortunately, most model systems used, e.g., the CO_2−caustic soda system, are much more complex and consist of (a set of) reversible reaction(s). The nature of these systems makes the conditions at which the interfacial area can be determined much more severe and puts more limitations on the process conditions and experimental equipment than a priori can be expected. Therefore an extended absorption model, as derived in section 2, is required to determine the conditions at which the interfacial area can be measured without detailed knowledge of the values of the liquid-side mass transfer coefficient, k_l, beforehand.

For the experimental data evaluated it must be concluded that, based on the outcome of the simulations (see Figures 2 and 3), it might be possible to reliably derive interfacial areas *for controlled flow regimes*.

4. Conclusions

The determination of interfacial areas in gas−liquid contactors by means of the chemical method (Danckwerts[1]) has been very frequently applied. However, this technique is only applicable for process conditions and absorption systems that obey the condition $2 < Ha < E_{inf}$. This condition has been derived for systems consisting of an irreversible second-order reaction. Unfortunately, many of the model systems used are reversible, and therefore the condition $2 < Ha < E_{inf}$ possibly is not met.

A major outcome of the present study is that it is concluded that the use of the chemical method for the determination of the interfacial area in gas−liquid contactors is a good technique, but the boundary conditions of its applicability are much more limited than generally is assumed. The effect of reversibility of the occurring reactions is a very important issue and adds severe constraints to the reaction system, experimental conditions, and the gas−liquid contactor, respectively.

In the study of Raynal et al.[3] the system used was the CO_2−caustic soda system, in which the reactions cannot be regarded as irreversible. Therefore, a system check has been carried out and it is concluded that, for the conditions and experimental equipment of this study, the applicability of the CO_2−caustic soda system is limited and effectively turns out to be suited *for controlled flow regimes*.

Notation

a = specific gas−liquid interface, m² m⁻³
C = concentration, mol m⁻³
D = diffusion coefficient, m² s⁻¹
E = enhancement factor
J = mass transfer flux, mol m⁻² s⁻¹
k_g = gas-phase mass transfer coefficient, m s⁻¹
k_l = liquid-phase mass transfer coefficient, m s⁻¹
k = reaction rate constant
m = partition coefficient
n = tray (or segment) number
N = total number of trays per column section
R = reaction rate, mol m⁻³ s⁻¹
R = gas constant, (8.314) J mol⁻¹ K⁻¹
t = time, s
T = temperature, K
x = molar fraction liquid phase
x = position in penetration model, m
y = molar fraction gas phase
V_{tray} = volume of liquid on a tray or segment, m³

Greek Symbols

α = liquid loading, mol mol⁻¹
γ = stoichiometry number
ρ = density, kg m⁻³
Φ = gas flow, mol (N) m⁻³

Subscripts

am = amine
back = backward reaction
eq = equilibrium
fwd = forward reaction
g = gas phase

l = liquid phase
i = component i
j = timeline index
tot = total, reacted as well as unreacted

Superscripts

n = tray number
i = interface

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