

*Dedicated to Professor Luminița Silaghi-Dumitrescu
on the occasion of her 65th anniversary*

CALCULATION OF THE EFFECTIVE MASS TRANSFER AREA IN TURBULENT CONTACT ABSORBER

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ABSTRACT. One of the most important efficiency parameters for separation columns is the effective interfacial mass transfer area. In this study the effective mass transfer area in gas-solid-liquid turbulent contact absorber was determined using the chemical absorption of carbon dioxide into sodium hydroxide aqueous solution. The effective mass transfer area determined in this study was found to be from three to six times higher than the geometric area of the solid packing.

Keywords: *Three phase fluidized bed, gas-solid-liquid turbulent contact absorber, effective mass transfer area, chemical methods*

INTRODUCTION

In order to obtain maximum absorption efficiency it is necessary to use proper equipments to maximize gas-liquid contact, such as new columns with three phase fluidized beds [1,2]. Gas-solid-liquid three phase fluidized bed absorber is mass transfer equipment in which the bed of low density packing is fluidized by the counter current flow of gas continuous phase and liquid as a dispersed phase.

The efficiency of the mass transfer depends on many factors such as the effective interfacial mass transfer area, mass transfer coefficients and liquid holdup.

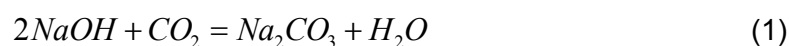
The effective mass transfer area (a_e) is not identical either to the geometric area (a) or to the wetted area of the packing (a_w). The measurement of the specific geometric area of packing is straightforward but is not a design parameter. The wetted area is that part of geometric area over which there is a liquid film.

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The most popular absorption system employed for determining interfacial mass transfer area with the use of the chemical method is the reactive absorption of CO₂ into aqueous solutions of sodium hydroxide [3-6]. The liquid phase reaction is considered to fall in the fast reaction regime with the mechanism given by Tsai [7, 8].

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The effective mass transfer area can be calculated using a reactive absorption system such as air-CO₂-NaOH. A pseudo-first order reaction has been chosen as model reaction in this study, due to the fact that the kinetics of the system is well-known and the chemicals are easy to handle. When the NaOH is in excess, the reaction between the dissolved CO₂ and the NaOH in solution can be expressed as:



The concentration profiles of the two reactants in the liquid film are described by the next differential equations:

$$D_{CO_2} \frac{\partial^2 C_{CO_2}}{\partial x^2} + k_2 \cdot C_{CO_2} \cdot C_{HO^-} = 0 \quad (2)$$

$$D_{HO^-} \frac{\partial^2 C_{HO^-}}{\partial x^2} + 2k_2 \cdot C_{CO_2} \cdot C_{HO^-} = 0 \quad (3)$$

with these boundary conditions:

$$at \ x = 0; \ C_{CO_2} = C_{CO_2}^i \ and \ \frac{dC_{HO^-}}{dx} = 0 \quad (4)$$

$$at \ x = \delta; \ C_{CO_2} = C_{CO_2}^0 \ and \ C_{HO^-} = C_{HO^-}^0$$

The analytical solution of the system (2)-(4) leads to the enhancement factor concept, E. This solution is available only if the reaction (1) is considered to be pseudo-first-order, i.e., the concentration of NaOH is not depleted in the liquid film ($C_{HO^-} = C_{HO^-}^0$). According to Danckwerts mass-transfer model the reaction rate can be expressed as [8]:

$$v_{abs} = a_e C_{CO_2}^i \left(k_1 D_{CO_2} + k_L^2 \right)^{1/2} \quad (5)$$

The mass-transfer process of the absorption of CO₂ is controlled by the liquid film, according to Henry's law:

$$C_{CO_2}^i = H \cdot P_{CO_2} \quad (6)$$

In the fast reaction regime, defined by $Ha > 5$, the enhancement factor equals the Hatta number and rapid chemical reaction is not sensitive to liquid side mass transfer coefficient k_L , the equation of the reaction rate becomes:

$$v_{abs} = a_e \cdot H \cdot P_{CO_2} \left(k_1 D_{CO_2} \right)^{1/2} \quad (7)$$

The solubility coefficient of CO₂ in the electrolyte solution can be estimated as follows [9]:

$$\lg \frac{H}{H_0} = -K_s \cdot I \quad (8)$$

$$K_s = i_+ + i_- + i_g = i_{Na^+} + i_{HO^-} + i_{CO_3^{2-}} + i_{CO_2} \quad (9)$$

For the system CO₂-NaOH-H₂O, ion coefficients are [3]:

$$\begin{aligned} i_{Na^+} &= 0.091; \\ i_{HO^-} &= 0.066; \\ i_{CO_3^{2-}} &= 0.021 \\ i_{CO_2} &= -0.019 \end{aligned} \quad (10)$$

So, $K_s = 0,159$ and equation (8) become:

$$\lg \frac{H}{H_0} = -0.159 \cdot I \quad (11)$$

The solubility coefficient of CO₂ in pure water, H_0 , depends only on temperature and can be calculated with relation (12) [9]:

$$\lg H_0 = \frac{1140}{T} - 5.30 \quad (12)$$

The rate constant of the pseudo-first-order reaction is k_1 and its relationship with temperature and ionic concentration is given by equation (13) [10]:

$$k_1 = k_2 \cdot C_{HO^-} \quad (13)$$

where k_2 is the pseudo-second-order rate constant:

$$\lg\left(\frac{k_2}{k_2^\infty}\right) = 0.1987I - 0.012I^2 \quad (14)$$

$$\lg k_2^\infty = 11.895 - \frac{2382}{T} \quad (15)$$

The formula for the ionic strength is as follows:

$$I = \frac{1}{2} \sum C_i \cdot Z_i^2 = C_{NaOH}^0 \quad (16)$$

Using the equation system (17) it can be determine the concentration of CO_2 at interface $C_{CO_2}^i$.

$$C_{CO_2}^i = H \cdot P_{CO_2}$$

$$H = H_0 \cdot 10^{-0.159C_{NaOH}^0} \quad (17)$$

$$\lg H_0 = \frac{1140}{T} - 5.30$$

In the fast reaction regime, the enhancement factor $E = Ha$ and the equation of the absorption rate becomes:

$$v_{abs} = a_e \cdot C_{CO_2}^i \left(k_2 \cdot C_{HO^-}^0 \cdot D_{CO_2} \right)^{1/2} \quad (18)$$

The effective mass transfer area can be determined from the slope of the straight line, passing through the origin, obtained by the representation of equation (18).

RESULTS AND DISCUSSION

The carbon dioxide concentration at the inlet and outlet of system was determined. With these values and height of the fluidized bed, the absorption rate for the two concentrations of NaOH solution and two gas rates were determined:

$$v_{ab} = \frac{273}{293} \cdot \frac{w_g}{22.4} \cdot \frac{1}{H_{sf}} \left(Y_{CO_2}^i - Y_{CO_2}^f \right) \quad (19)$$

The reaction constant k_2 was calculated by the relations of:

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Barett equation [11]:

$$\lg k_2 = 11.8609 - \frac{2338.28}{T} + 0.1317 \cdot C_{NaOH}^0 \quad (20)$$

Astarita equation [12]:

$$\lg k_2 = 13.635 - \frac{2895}{T} + 0.160 \cdot C_{NaOH}^0 \quad (21)$$

Pohorecki equation [13]:

$$\lg k_2 = 11.895 - \frac{2382}{T} + 0.221 \cdot C_{NaOH}^0 - 0.016 \left(C_{NaOH}^0 \right)^2 \quad (22)$$

The obtained values are presented in the table 1.

Table 1. Values of reaction constant k_2 [l / mol s] at T=293 K

Equation→ C_{NaOH}^0 ↓	(20)	(21)	(22)
0.0 N	7593.154	5681.381	5825.151
0.5 N	8836.335	6830.522	7444.014
1.0 N	10283.06	8212.094	9339.146

The values of k_2 obtained with Pohorecki equation are approximated the mean value of values obtained with equations (20) and (21), therefore these values are used.

The CO₂ diffusivity in pure water depends on the temperature as follows:

$$\lg D_{CO_2}^0 = -8.1764 + \frac{712.5}{T} - \frac{2.591 \cdot 10^5}{T^2} \quad (23)$$

While the diffusivity of the CO₂ in the NaOH aqueous solution was calculated by the equation (24):

$$D_{CO_2} = D_{CO_2}^0 \cdot \left(\frac{\eta_{H_2O}}{\eta_{NaOH}} \right)^{0.637} \quad (24)$$

Using equations (23), (24) and the viscosity values of NaOH aqueous solutions has been calculated the diffusion coefficient D_{CO_2} .

Henry constant of CO₂ in NaOH aqueous solutions has been calculated. These values are presented in table 2.

Table 2. Diffusion coefficient D_{CO_2} in NaOH aqueous solutions and Henry constant H at $T=293$ K

C_{NaOH}^0 [mol/l]	0	0.5	1
$D_{CO_2} \cdot 10^9$ [m ² / s]	1.76	1.74	1.62
$H \cdot 10^2$ [atm · l/mol]	3.897	3.245	2.7

In the tables 3 to 6 the concentrations of carbon dioxide at interfaces, the absorption rates and X coordinate from equation (18) are presented.

Table 3. Experimental results for the absorption in three phase fluidized bed of CO₂ into 0.5 N NaOH aqueous solution at $v_g = 1.1$ [m/s].

C_{CO_2}	$C_{CO_2}^i$	$C_{CO_2}^i \sqrt{k_2 \cdot C_{HO^-} \cdot D_{CO_2}}$	v_{ab} [Kmol/m ³ s]	Liquid flow [l/h]
0	0	0	0	L=100
5%	1.6200E-03	4.1940E-06	0.006221753	
8%	2.6000E-03	6.7300E-06	0.010433075	
10%	3.2400E-03	8.3900E-06	0.012784895	
0	0	0	0	L=200
5%	1.6200E-03	4.1940E-06	0.006815129	
8%	2.6000E-03	6.7300E-06	0.011182052	
10%	3.2400E-03	8.3900E-06	0.013601283	

Table 4. Experimental results for the absorption in three phase fluidized bed of CO₂ into 1.0 N NaOH aqueous solution at $v_g = 1.1$ [m/s].

C_{CO_2}	$C_{CO_2}^i$	$C_{CO_2}^i \sqrt{k_2 \cdot C_{HO^-} \cdot D_{CO_2}}$	v_{ab} [Kmol/m ³ s]	Liquid Flow [l/h]
0	0	0	0	L=100
5%	1.3500E-03	5.2300E-06	0.006636536	
8%	2.1600E-03	8.3700E-06	0.011128613	
10%	2.7000E-03	1.0500E-05	0.013637221	
0	0	0	0	L=200
5%	1.3500E-03	5.2300E-06	0.007520142	
8%	2.1600E-03	8.3700E-06	0.012338816	
10%	2.7000E-03	1.0500E-05	0.014539302	

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Table 5. Experimental results for the absorption in three phase fluidized bed of CO₂ into 0.5 N NaOH aqueous solution at v_g = 2.11 [m/s].

C _{CO2}	C ⁱ _{CO2}	$C_{CO_2}^i \sqrt{k_2 \cdot C_{HO^-} \cdot D_{CO_2}}$	v _{ab} [Kmol/m ³ s]	Liquid Flow [l/h]
0	0	0	0	L=100
5%	1.6200E-03	4.1940E-06	0.011137704	
8%	2.6000E-03	6.7300E-06	0.017339647	
10%	3.2400E-03	8.3900E-06	0.019845364	
0	0	0	0	L=200
5%	1.6200E-03	4.1940E-06	0.013427058	
8%	2.6000E-03	6.7300E-06	0.019669209	
10%	3.2400E-03	8.3900E-06	0.024171189	

Table 6. Experimental results for the absorption in three phase fluidized bed of CO₂ into 1.0 N NaOH aqueous solution at v_g = 2.11 [m/s].

C _{CO2}	C ⁱ _{CO2}	$C_{CO_2}^i \sqrt{k_2 \cdot C_{HO^-} \cdot D_{CO_2}}$	v _{ab} [Kmol/m ³ s]	Liquid Flow [l/h]
0	0	0	0	L=100
5%	1.3500E-03	5.2300E-06	0.012622731	
8%	2.1600E-03	8.3700E-06	0.020329241	
10%	2.7000E-03	1.0500E-05	0.024097942	
0	0	0	0	L=200
5%	1.3500E-03	5.2300E-06	0.015217332	
8%	2.1600E-03	8.3700E-06	0.022947411	
10%	2.7000E-03	1.0500E-05	0.029005427	

The effective mass transfer area was determined as a slope of the straight line from representation of equation (18):

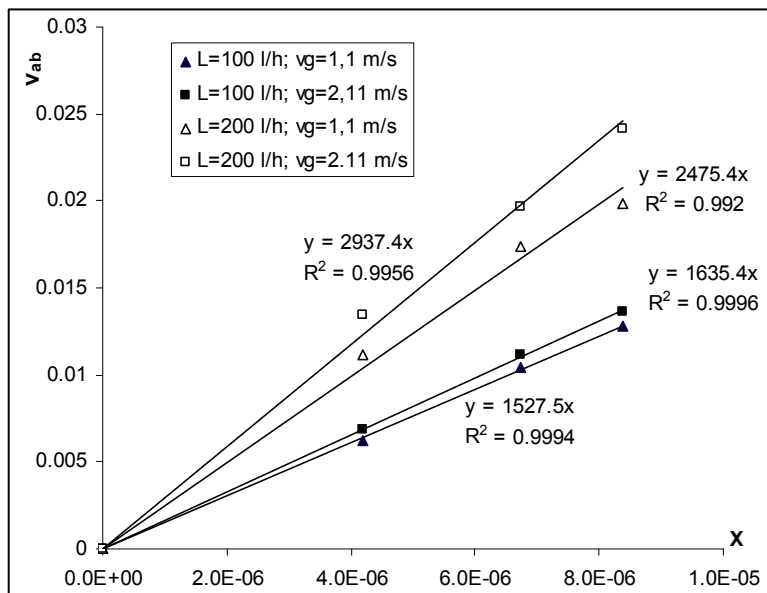


Figure 1. Absorption rate versus X at $C^0_{HO^-}=0.5 \text{ mol/l}$, $T=293\text{K}$

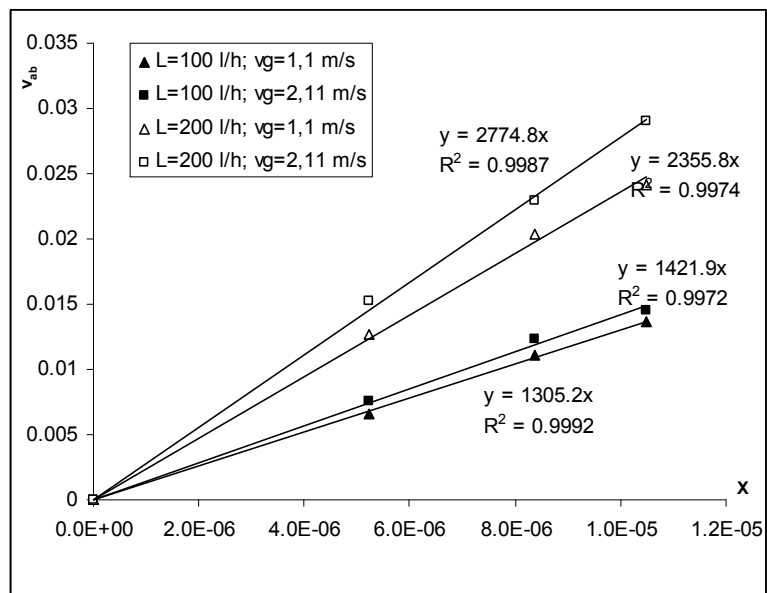


Figure 2. Absorption rate versus X at $C^0_{HO^-}=1 \text{ mol/l}$, $T=293\text{K}$

The numerical values of the determined effective mass transfer area from figures 1-2 are centralized in table 7.

Table 7. Effective mass transfer area in three phase fluidized bed ($a_e, [m^2/m^3]$)

$C_{NaOH}^0 / L [l/h]$	100	200	
0.5 [mol/l]	1527.5	1635.4	$V_g=1.10 [m/s]$
1.0 [mol/l]	1305.2	1421.9	
<i>Average</i>	<i>1416.3</i>	<i>1528.6</i>	
0.5 [mol/l]	2475.4	2937.4	$V_g=2.11 [m/s]$
1.0 [mol/l]	2355.8	2774.8	
<i>Average</i>	<i>2415.6</i>	<i>2856.1</i>	

At a constant concentration of NaOH solution and constant liquid flow rate, effective mass transfer area increases more with gas velocity, from $1527.5 \text{ m}^2/\text{m}^3$ ($C_{NaOH} = 0.5 \text{ mol/l}$, $v_g = 1.10 \text{ m/s}$ and $L = 100 \text{ l/h}$) at $2475.4 \text{ m}^2/\text{m}^3$ ($C_{NaOH} = 0.5 \text{ mol/l}$, $v_g = 2.11 \text{ m/s}$ and $L = 100 \text{ l/h}$).

At a constant concentration of NaOH solution and constant gas velocity, the effective mass transfer area increases low with the increase of the liquid flow, from $2475.4 \text{ m}^2/\text{m}^3$ ($C_{NaOH} = 0.5 \text{ mol/l}$, $v_g = 2.11 \text{ m/s}$ and $L = 100 \text{ l/h}$) at $2937.4 \text{ m}^2/\text{m}^3$ ($C_{NaOH}=0.5 \text{ mol/l}$, $v_g=2.11 \text{ m/s}$ and $L=200 \text{ l/h}$). The increase of liquid flow determines the increase of packing buoyancy and a better fluidization is obtained.

At a constant liquid flow rate and constant gas velocity, effective mass transfer area decreases with the increase of NaOH concentration, from $1527.5 \text{ m}^2/\text{m}^3$ ($C_{NaOH} = 0.5 \text{ mol/l}$, $v_g = 1.1 \text{ m/s}$ and $L = 100 \text{ l/h}$) at $1305.2 \text{ m}^2/\text{m}^3$ ($C_{NaOH} = 1 \text{ mol/l}$, $v_g = 1.1 \text{ m/s}$ and $L = 100 \text{ l/h}$). This phenomenon can be explained using the two-film theory. The reaction of CO_2 and NaOH takes place in liquid boundary layer, so CO_2 has to diffuse through gas film and enter in liquid film to reach the NaOH solution. The increase of NaOH concentration causes the viscosity liquid increase, thus CO_2 diffusion in liquid phase is slower and mass transfer area is decrease.

CONCLUSION

The effective mass transfer area in turbulent contact absorber with low density solid particles has been determined. The absorption of carbon dioxide from the mixture of air- CO_2 into 0.5 N and 1.0 N aqueous NaOH solutions was employed as model reaction of known kinetics.

The effective mass transfer area increases when gas velocity and liquid flow increase and decreases when the concentration of the NaOH solution increases.

The effective mass transfer area in turbulent contact absorber with three phase fluidized bed is from three to six times higher than the geometric area of the solid packing.

EXPERIMENTAL

In order to determine the effective mass transfer area in turbulent contact absorber with low density inert solid packing, the rate absorption of carbon dioxide from air-CO₂ mixture into the NaOH solution was measured. The flow diagram of the experimental equipment used for the absorption rate is represented in the figure 7.

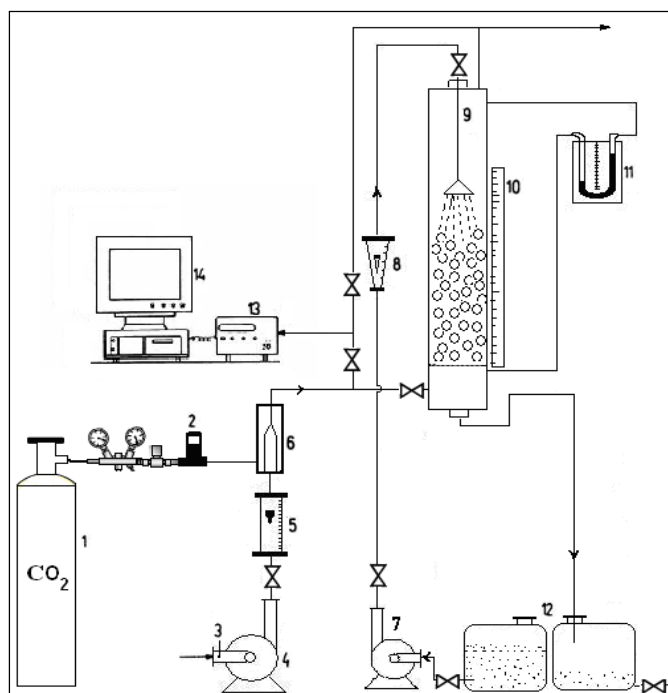


Figure 7. Flow diagram of the experimental equipment
 1-carbon dioxide cylinder, 2-carbon dioxide mass flow meter, 3-air temperature controller, 4-air blower, 5-gas rotameter, 6-gas mixing tube, 7-liquid pump, 8-liquid rotameter, 9-three phase fluidized bed absorber column, 10-graduet scale, 11-manometer, 12-slution tanks, 13-gas analyzer, 14-IBM computer.

The column was made of glass with an interior diameter of 0.14 m and high 1.10 m. The glass column was used for better visual observation of the phenomena occurring in the fluidized bed. Gas mixture is the continuous phase and was introduced at predetermined flow rate into the column. NaOH solution of specified concentration as dispersed phase was sprayed from the liquid distributor over the top of the fluidized bed.

The solid materials which were used as solid phase are plastic hollow balls of 1.0 cm diameter and with the density of 170 kg/m³. The experiments were conducted in a fully fluidized state. The fluidized bed height was read on the scale. The liquid and air flow rates were measured by rotameters and carbon dioxide flow rate was measured and controlled by mass flow meter. The composition of carbon dioxide in the entering and leaving gas phase was measured using a calibrate BINOS 120 gas analyzer. The experimental conditions are presented in table 8.

Table 8. The experimental conditions (T = 293 K, P = 1 bar)

Variable	Range
Column diameter D_c [m]	0.14
Diameter of solid particle d_p [m]	0.01
Solid particle density [Kg/m ³]	170
Static solid bed height H_0 [m]	0.12
Gas velocity v_g [m/s]	1.1; 2.11
CO ₂ mole fraction in the gas mixture	0.05; 0.08; 0.1
Liquid flow [l/h]	100; 200
Concentration of NaOH solution, [mol/l]	0.5; 1.0

The absorption rate has been calculated from the measured inlet ($Y_{CO_2}^0$) and outlet (Y_{CO_2}) concentration of the carbon dioxide in the gas phase.

REFERENCES

1. A. Shabani, S.T. Asl and B.H.Shahraki, *International Journal of Chemical Engineering and Application*, **2010**, 1(1), 117.
2. S. Dragan, *Stud. Univ. Babes-Bolyai, Chemia*, **2015**, 60, 3, 183.
3. P.V. Danckwerts, "Gas-Liquid Reactions", New York, McGraw-Hill, **1970**, Chap.9, 206.
4. G.H. Joosten and P. Danckwerts, *Chem. Eng. Science*, **1973**, 28, 453.

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5. M. Drăgan, S. Drăgan, I. Siminiceanu. *Stud. Univ. Babes-Bolyai, Chemia*, **2000**, 45 (1-2), 11.
6. M.H. de Brito, U. von Stockar, A.M. Bangerter and M. Laso. *Ind. Eng. Chem. Res.*, **1994**, 33 (3), 647.
7. E.R.Tsai, P.Schultheis, A. Kettner, J.C.Lewis, A.F. Seibert, R.B.Eldridge and G.T. Rochelle, *Ind. Eng. Chem. Res.*, **2008**, 47, 1253.
8. R. Tsai, PhD Dissertation, Austin, University of Texas. **2010**.
9. M.M., Sharma, P.V. Danckwerts, *Br.Chem.Eng.*, **1970**, 15, 522.
10. K. Yang, G. Chu, H. Zou, B. Sun, L., J.F. Shao, *Chem. Eng. Journal*, **2011**, 168, 1377.
11. P.V.L. Baret, PhD Thesis, University of Cambridge, **1967**.
12. G. Astarita, D.V. Savage, A. Bisio, "Gas treating with chemical solvents", John Wiley & Sons, **1983**. Chap. 9, 210.
13. R. Pohorecki, W. Moniuk, *Chem. Eng. Sci.*, **1988**, 43 (7), 1677.