$k_F a_v$	= liquid-film mass transfer coefficient	[l/min]
$k_s a_v$	= solid-phase mass transfer coefficient	[l/min]
Pe	= Peclet number of axial dispersion based on particle diameter	[—]
q	= amount adsorbed on particle	[cc/g]
Re	= Reynolds number based on particle diameter	[—]
Sc	= Schmidt number	[—]
t	= time	[min]
u	= superficial velocity	[cm/min]
V_F	= velocity of adsorption front defined by Eq. (4)	[cm/min]
Z	= bed length	[cm]
β	= adsorption coefficient	[cc/g]
γ	= packed density	[g/cc]
ε_b	= bed voidage	[—]
ε_{pt}	= total porosity of particle	[—]

θ	= time, $t - \varepsilon_b Z/u$	[min]
ρ_p	= apparent density of particle	[g/cc]
ρ_t	= true density of particle	[g/cc]
τ_t	= tortuosity factor, $\varepsilon_{pt} D_m/D_e$	[—]

Literature Cited

- 1) Kawazoe, K. and T. Kawai: *Kagaku Kōgaku*, **32**, 1122 (1968).
- 2) Longsworth, L. G.: *J. Phys. Chem.*, **67**, 689 (1963).
- 3) Miyauchi, T. and Kikuchi: *Chem. Eng. Sci.*, **30**, 343 (1975).
- 4) Satterfield, C. N.: "Mass Transfer in Heterogeneous Catalysis", M.I.T. Press (1970).
- 5) Shiotsuka, T., T. Togami and I. Yokoyama: *Kagaku Kōgaku*, **34**, 44 (1970).
- 6) Takeuchi, K. and Y. Uraguchi: *J. Chem. Eng. Japan*, **10**, 297 (1977).
- 7) Wilson, E. J. and C. J. Geankoplis: *Ind. Eng. Chem., Fundam.*, **5**, 9 (1966).

BOUNDARY OF HYDRODYNAMIC FLOW REGION AND GAS-PHASE MASS-TRANSFER COEFFICIENT IN PACKED COLUMN WITH COCURRENT DOWNWARD FLOW

SUSUMU FUKUSHIMA AND KATSUHIKO KUSAKA

Department of Chemical Engineering, Kansai University, Suita 564

Introduction

Cocurrent flow operation is to be preferred in cases where the mean driving force is not affected by flow direction, as in the absorption of an acid gas by a highly alkaline solution. In particular, this case is an operation under a high gas and a low liquid flow rate. The estimation of gas-phase volumetric coefficients of mass transfer, $k_g a_p$, for spray, pulse and wavy flow regions is thus important in designing and operating the columns.

For cocurrent downward operations, Reiss⁵⁾ and Gianetto *et al.*⁴⁾ have presented empirical equations for $k_g a_p$ in terms of dissipated energy and other factors. Shende and Sharma⁸⁾ have also reported empirical equations in terms of gas and liquid flow rates. There are few reports of empirical equations for gas-phase mass-transfer coefficient k_g . Gianetto *et al.*⁴⁾ have only reported the equation in terms of dissipated energy and other factors for the pulse flow region.

It was presented in the earlier papers¹⁾ that the

dependence of interfacial area a_p on the gas Reynolds number Re_g and the liquid Reynolds number Re_l is distinctly different in spray, pulse, wavy, dispersed bubble and trickle flow regions, and that the empirical equations of hydrodynamic flow boundaries are obtained by combining two of the equations for a_p . These equations of flow boundaries were also obtained from the equations of liquid-phase volumetric coefficients of mass transfer, $k_l^* a_p$, in a similar fashion to that described in the early papers²⁾.

We had three objectives in this investigation. The first was to obtain the empirical equations of hydrodynamic flow boundaries by analysing the literature data on $k_g a_p$ in the fashion mentioned above. The second was to obtain an empirical equation of k_g . The third was to compare the predicted values with the experimental data on the absorption rate of sulfur dioxide from factory exhaust gas into water.

1. Gas-Phase Volumetric Coefficients of Mass Transfer and Hydrodynamic Flow Boundary

Figure 1 shows the dependence of $k_g a_p$ on Re_l and Re_g obtained from rearrangement of the literature

Received December 2, 1977. Correspondence concerning this article should be addressed to S. Fukushima.

B.: Berl saddles, P.R.: Pall rings, R.: Raschig rings, S.: spheres

Fig. 1 Rearrangement of the literature data on gas-phase volumetric coefficients of mass transfer

data, where the liquid holdup Φ_l is estimated by the equations in the earlier papers¹⁾. For ammonia gas absorption into 2 N- Na_2SO_4 solution for 0.6 cm spheres and Berl saddles, the Gianetto data⁴⁾ are significantly divided into A, B, E and H regions. For sulfur dioxide gas absorption into 2N- NaOH solution for 5/8 in. Pall rings, the Shende data⁶⁾ pertain to A and B regions. The Wen data⁷⁾, on dehumidification of air by CaCl_2 solution for various sizes of Raschig rings, also pertain to A region.

The empirical equations of $k_g a_p$ for A, B and E regions were thus obtained, on the assumption that the value of k_g is proportional to the square root of diffusivity in gas-phase D_g , as follows:

For flow region A

$$\frac{k_g a_p d_p^2}{D_g (1 - \Phi_l / \epsilon)} = 1.9 \times 10^{-1} \phi^{-0.7} Re_l^{0.15} Re_g^{0.9} Sc_g^{1/2} (d_p/T)^{-0.2} \quad (1)$$

For flow region B

$$\frac{k_g a_p d_p^2}{D_g (1 - \Phi_l / \epsilon)} = 1.7 \times 10^{-1} \phi^{-0.7} Re_l^{0.7} Re_g^{0.6} Sc_g^{1/2} (d_p/T)^{-0.2} \quad (2)$$

Fig. 2 Boundaries of hydrodynamic flow regions for 0.6 cm spheres

For flow region E

$$\frac{k_g a_p d_p^2}{D_g (1 - \Phi_l / \epsilon)} = 1.9 \times 10^{-2} \phi^{-1.7} Re_l^{0.6} Re_g^{1.3} Sc_g^{1/2} \quad (3)$$

The equations of hydrodynamic flow boundaries may be thus given by combining two of these equations for $k_g a_p$ as follows:

For boundary A-B

$$1.1 = Re_l^{0.55} Re_g^{-0.3} \quad (4)$$

For boundary A-E

$$10 = \phi^{-1.0} Re_l^{0.45} Re_g^{0.4} (d_p/T)^{0.2} \quad (5)$$

For boundary B-E

$$8.9 = \phi^{-1.0} Re_l^{-0.1} Re_g^{0.7} (d_p/T)^{0.2} \quad (6)$$

Figure 2 indicates that the predicted boundaries (thick lines) according to Eqs. (4), (5) and (6) are fairly close to those (thin lines) of spray-pulse, spray-wavy and pulse-wavy boundaries given by combining two of the equation on a_p in the earlier papers¹⁾, respectively, for 0.6 cm spheres, where $d_p/T=0.075$. It is thus found that the spray, pulse and wavy flow regions correspond to A, B and E regions, respectively. Furthermore, the close resemblance between thick and thin lines suggests that the k_g is roughly presented by the same equation for spray, pulse and wavy flow regions.

Even though the H region is placed at low Re_l and Re_g , the dependence of $k_g a_p$ on Re_l for H region is much larger than that for other regions. To obtain reasonable boundaries of pulse-trickle and wavy-trickle flows, the $k_p a_p$ at low Re_l and Re_g must be further investigated.

2. Gas-Phase Mass-Transfer Coefficient

The equation of k_g was obtained by dividing $k_g a_p$ by a_p , where the value of a_p for respective flow regions is given by the equation in the earlier papers¹⁾. Hence, the equation for pulse flow region is obtained from Eq. (2) as follows:

$$Sh_g = \frac{k_g d_p}{D_g} = 8.0 \times 10^3 \phi^{-0.4} Re_g^{0.4} Sc_g^{1/2} (d_p/T)^{2.3} \quad (7)$$

Table 1 Comparison of predictions with the data on absorption of SO₂ into water in pulse flow region

Re_g [—]	Re_l [—]	P_{Ain} [10 ² atm]	$(k_l^*a_p)_{cal}$ [10 ³ sec ⁻¹]	$(k_g'a_p)_{cal}$ g-mole [10 ⁵ cm ³ ·sec·atm]	C_{Aout} [10 ³ M]		$K'_{OG}a_p$ g-mole [10 ⁶ cm ³ ·sec·atm]		N_A [10 ⁴ $\frac{g-mole}{sec}$]	
					obs.	cal.	obs.	cal.	obs.	cal.
180	53	20.7	21	12	180	180	26	26	50	50
180	132	9.09	45	20	53.8	53	58	61	37	37
180	202	1.83	64	25	8.2	8.5	70	82	5.7	5.9
180	313	9.36	93	32	25.8	29	73	120	44	49
393	53	6.31	30	20	78.4	78	41	43	22	22
393	132	2.73	64	33	27.4	27	100	100	19	19

Although the equations for spray and wavy flow regions were also obtained from Eqs. (1) and (3), the predicted values of k_g from those equations do not show much difference in comparison with the values given by Eq. (7).

There are available data on k_g given by Gianetto *et al.*⁴⁾ for 0.6 cm Berl saddles and by Shende and Sharma⁶⁾ for 1 in. Pall rings and 1 in. Intalox saddles. Other data on k_g for spray, pulse and wavy flow regions were also obtained by dividing the literature data on $k_g a_p$ by a_p estimated from the equation in the earlier papers¹⁾. The correlation of Eq. (7) and these data agree within $\pm 30\%$ error, except for the Wen data⁷⁾ at high Re_l , as shown in Fig. 3. The observed value of $k_g a_p$ given by Wen *et al.*⁷⁾ decreases as Re_l increases at high Re_l in Fig. 1, but this abnormal result has not been found in other investigations.

3. Physical Absorption of Sulfur Dioxide with Water

3.1 Experimental

The absorption of SO₂ from exhaust gas of a chemical factory into water was performed at $26.4 \pm 0.5^\circ\text{C}$ in a packed column, 10 cm in diameter and 100 cm in height, using 1/2 in. ceramic spheres, as described in the earlier papers¹⁾. The partial pressure of SO₂ at the column inlet, P_{Ain} , was varied from 0.0183 to 0.207 atm, where the total pressure is 1 atm. On the assumption that the bulk of liquid is in piston flow, the overall volumetric coefficient of mass transfer, $K'_{OG} a_p$, was determined by the difference of SO₂ content in liquid phase at the inlet and the outlet.

The diffusivities of SO₂ for gas and liquid phase at 30°C are respectively 0.14 cm²/sec given by Shende and Sharma⁶⁾ and 2.5×10^{-5} cm²/sec given by Whitney and Vivian⁸⁾. The effects of temperature on these diffusivities for gas and liquid phase are given by the Fuller³⁾ and by the Stokes-Einstein equations, respectively.

3.2 Comparison of predictions with the data

Table 1 compares predictions with experimental data on absorption rate N_A . These operating conditions are placed in the pulse flow region from the map of hydrodynamic flow boundaries. The predicted values of $K'_{OG} a_p$ are thus obtained by combination of

B.: Berl saddles, I.: Intalox saddles, P.R.: Pall rings, R.: Raschig rings, S.: spheres

Fig. 3 Correlation of Eq. (7) and the data for spray, pulse and wavy flow regions

k_g given by Eq. (7), k_l^* and a_p given by respective equations in the earlier papers^{1,2)}. The predicted N_A are in good agreement with the data.

This result indicates that the empirical equations of k_g , k_l^* and a_p are useful in predicting absorption rate.

Conclusions

1. The empirical equations for spray-pulse, spray-wavy and pulse-wavy flow boundaries were obtained from analysis of gas-phase volumetric coefficient of mass transfer. The predicted boundaries agreed fairly well with these boundaries as given from analysis of interfacial area in the earlier work.

2. The gas-phase mass-transfer coefficients are presented by the same equation for spray, pulse and wavy flow regions.

3. The predicted absorption rates of sulfur dioxide from exhaust gas in water are in good agreement with the experimental data.

Acknowledgment

The authors express special appreciation to Messrs. M. Kumagai and T. Nakajima for their experimental assistance, and to Mr. M. Noda, president and Mr. M. Murakami, chief of research, Daito Chemical Company for offering an opportunity for gas absorption experiments.

Nomenclature

a_p	= interfacial area per unit volume of packed column	[cm ⁻¹]
C_{Aout}	= concentration of sulfur dioxide in water at outlet of column	[g-mole/l]
D_g	= diffusivity in gas	[cm ² /sec]
d_p	= packing diameter	[cm]
d_s	= equivalent diameter of packing based on sphere	[cm]
G	= superficial mass velocity	[g/cm ² ·sec]
K'_{OG}	= overall mass-transfer coefficient based on gas phase	[g-mole/cm ² ·sec·atm]
k_g	= gas-phase mass-transfer coefficient	[cm/sec]
k'_g	= gas-phase mass-transfer coefficient	[g-mole/cm ² ·sec·atm]
k_l^*	= liquid-phase mass-transfer coefficient	[cm/sec]
N_A	= absorption rate of sulfur dioxide	[g-mole/sec]
P_{Ain}	= partial pressure of sulfur dioxide at inlet of column	[atm]
Re	= Reynolds number, $d_s G / \mu$	[—]
S	= geometrical surface of packing	[cm ²]
Sc_g	= Schmidt number in gas phase, $\mu_g / \rho_g D_g$	[—]
Sh_g	= Sherwood number in gas phase, $k_g d_p / D_g$	[—]

T	= column diameter	[cm]
ε	= void fraction in packed column	[—]
μ	= viscosity	[g/cm·sec]
ρ	= density	[g/cm ³]
Φ	= holdup of fluid per unit volume of packed column	[—]
ϕ	= surface shape factor of packing, S/d_p^2	[—]

<Subscripts>

cal.	= calculated
g	= gas-phase
l	= liquid-phase

Literature Cited

- 1) Fukushima, S. and K. Kusaka: *J. Chem. Eng. Japan*, **10**, 461 (1977).
- 2) Fukushima, S. and K. Kusaka: *ibid.*, **10**, 468 (1977).
- 3) Fuller, E. N., P. D. Schettler and J. C. Giddings: *Ind. Eng. Chem.*, **58**, No. 5, 19 (1966).
- 4) Gianetto, A., V. Specchia and G. Baldi: *AIChE J.*, **19**, 916 (1973).
- 5) Reiss, L. P.: *Ind. Eng. Chem., Process Des. Dev.*, **6**, 486 (1967).
- 6) Shende, B. W. and M. M. Sharma: *Chem. Eng. Sci.*, **29**, 1763 (1974).
- 7) Wen, C. Y., W. S. O'Brien and L. T. Fan: *J. Chem. Eng. Data*, **8**, 42 (1963).
- 8) Whitney, R. P. and J. E. Vivian: *Chem. Eng. Progr.*, **45**, 323 (1949).

AN EFFECTIVE SUB-OPTIMAL FORMULATION OF THE SINGULAR AND/OR BANG-BANG CONTROL PROBLEM

TAKEICHIRO TAKAMATSU AND YOSHIKI SHIMIZU

Department of Chemical Engineering, Kyoto University, Kyoto 606

In a recent study Jacobson *et al.*³⁾ have proposed a computational method to solve the singular and/or bang-bang control problem in a sub-optimal manner.

This method has been applied to the optimal control problem of chemical processes by Edgar and Lapidus¹⁾ and it has been shown to be effective especially in the large dimensional cases with multiple controls.

Along with this approach, Nishida *et al.*⁵⁾ have proposed the piece-wise *Max-H* strategy to deal with the optimal control problem of dynamical systems.

In these studies more emphasis has been paid on generality and easiness of the algorithm than on strict-

ness of the optimality. From the technical viewpoint sub-optimal formulation is preferable because of uncertainties in the system model or incomplete realization of the optimal policy.

This paper will present another approximation method which has computational advantages over previous methods.

1. Statement of the Problem

We are concerned with the following optimal problem:

$$\text{Min}_{u \in U} J = K(x(t_f)) + \int_{t_0}^{t_f} L(x, t) dt \quad (1)$$

subject to

Received October 31, 1977. Correspondence concerning this article should be addressed to Y. Shimizu, Research Reactor Institute, Kyoto Univ., Kumatori-cho Sennan-gun, Osaka 590-04.