

# Effect of an absorbate concentration level on the coupled mass and heat transfer during short gas plugs dissolution

Tov Elperin \*, Andrew Fominykh

Pearlstone Center for Aeronautical Engineering Studies, Department of Mechanical Engineering, Ben-Gurion University of the Negev,  
 P.O. Box 653, Beer-Sheva 84105, Israel

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**Abstract**—We studied gas absorption from a rising gas plug when the concentration level of the absorbate in the absorbent is finite (finite dilution of absorbate approximation). It is shown that in the case of the finite dilution the lateral convective term in the equation of convective mass transfer in the absorbate must be taken into account. It is found that the mass transfer rate increases with the increase of the absorbate concentration level. Isothermal and nonisothermal absorptions are considered whereby the latter is described by the coupled equations of mass and heat transfer. It is found that the mass transfer rate decreases when the dimensionless heat of absorption increases. © 2000 Éditions scientifiques et médicales Elsevier SAS

**nonisothermal absorption / mass and heat transfer / rising gas plug / finite dilution of absorbate in the absorbent**

## Nomenclature

$a$	thermal diffusivity of a liquid . . . . .	$\text{m}^2 \cdot \text{s}^{-1}$
$b$	coefficient in equation (37)	
$c$	molar density . . . . .	$\text{mol} \cdot \text{m}^{-3}$
$c_A$	absorbate concentration . . . . .	$\text{mol} \cdot \text{m}^{-3}$
$c_p$	specific heat . . . . .	$\text{kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$
$D$	molecular diffusion coefficient . . . . .	$\text{m}^2 \cdot \text{s}^{-1}$
$d$	coefficient in equation (37) . . . . .	$\text{K}^{-1}$
$d_{\text{ch}}$	channel diameter . . . . .	$\text{m}$
$g$	acceleration of gravity . . . . .	$\text{m} \cdot \text{s}^{-2}$
$K$	$= c_p / (dL)$ , dimensionless number	
$L$	heat of absorption . . . . .	$\text{kJ} \cdot \text{kg}^{-1}$
$L_B$	length of a gas plug . . . . .	$\text{m}$
$Le$	$= D/a$ , Lewis number	
$N_{A_r}$	mass flux density of component $A$ in radial direction . . . . .	$\text{kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$
$N_{A_z}$	mass flux density of component $A$ in axial direction . . . . .	$\text{kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$
$Pe$	$= U_{\infty} d_{\text{ch}} D^{-1}$ , Peclet number for a gas plug	
$Pe^*$	$= 8/3(Re Pr)$ , Peclet number for a falling liquid film	

$Pr$	$= \nu/a$ , Prandtl number	
$Q_c$	mass flux . . . . .	$\text{kg} \cdot \text{s}^{-1}$
$Q_{c_0}$	mass flux in the approximation of the infinite dilution . . . . .	$\text{kg} \cdot \text{s}^{-1}$
$Q_{c_0}^{\text{is}}$	mass flux for isothermal absorption in the approximation of the infinite dilution . . . . .	$\text{kg} \cdot \text{s}^{-1}$
$Q_T$	heat flux . . . . .	$\text{kJ} \cdot \text{s}^{-1}$
$Q_{T_0}$	heat flux in the approximation of infinite dilution . . . . .	$\text{kJ} \cdot \text{s}^{-1}$
$Re$	$= 8u_s \delta / (3\nu)$ , Reynolds number for liquid film	
$r$	radial coordinate . . . . .	$\text{m}$
$R$	channel radius . . . . .	$\text{m}$
$R_i$	ratio between terms in equation (13)	
$Sh$	Sherwood number	
$Sh_0$	Sherwood number under the assumption of the infinite dilution	
$s$	length of an arc measured from a gas plug's nose . . . . .	$\text{m}$
$X$	$= (x_A - x_{A_0}) / (x_{A_s} - x_{A_0})$ , dimensionless weight fraction of the absorbate	
$x_A$	weight fraction of the absorbate	
$x_{A_s}$	weight fraction of the absorbate at the gas–liquid interface	
$x_{A_0}$	weight fraction of the absorbate at the inlet	

\* Correspondence and reprints.  
 elperin@menix.bgu.ac.il

$x'_{A0}$	equilibrium weight fraction of absorbate at the initial temperature	
$X_A^*$	molar fraction of the absorbate at the gas-liquid interface	
$X_{Ai}$	molar fraction of the absorbate at the inlet	
$T$	temperature of a liquid . . . . .	K
$T'_0$	equilibrium temperature at the initial concentration . . . . .	K
$u_s$	interfacial velocity in the liquid film . . . . .	$m \cdot s^{-1}$
$U_\infty$	velocity of a gas plug rising in a stagnant fluid . . . . .	$m \cdot s^{-1}$
$v_r, v_z$	velocity components . . . . .	$m \cdot s^{-1}$
$z$	axial coordinate . . . . .	m

$\varphi$	dimensionless average mass velocity	
$\psi$	stream function . . . . .	$m^3 \cdot s^{-1}$
$\Psi$	$= \psi / (2\sqrt{D\mu})$ , similarity variable	
$\Psi'$	$= \psi' / (2\sqrt{a\mu})$ , similarity variable	
$\Omega$	enhancement factor	

*Subscripts*

s	value at the interface
$\infty$	value at infinity
0	value at the inlet
A	absorbate
B	absorbent

*Greek symbols*

$\alpha$	coefficient of heat transfer . . . . .	$kJ \cdot m^{-2} \cdot s^{-1} \cdot K^{-1}$
$\alpha_0$	coefficient of heat transfer in the approximation of the infinite dilution of the absorbate . . . . .	$kJ \cdot m^{-2} \cdot s^{-1} \cdot K^{-1}$
$\beta$	finite dilution nonisothermal mass transfer coefficient . . . . .	$m \cdot s^{-1}$
$\beta^{is}$	finite dilution isothermal mass transfer coefficient . . . . .	$m \cdot s^{-1}$
$\beta_0$	infinite dilution nonisothermal mass transfer coefficient . . . . .	$m \cdot s^{-1}$
$\beta_0^{is}$	infinite dilution isothermal mass transfer coefficient . . . . .	$m \cdot s^{-1}$
$\delta$	thickness of a liquid film . . . . .	m
$\theta_1$	$= \beta^{is} / \beta_0^{is}$ , correction factor which accounts for the effect of the finite concentration level of the absorbate on the rate of mass transfer in a case of isothermal absorption	
$\theta_2$	$= \beta / \beta_0^{is}$ , correction factor which accounts for the effects of the temperature change and the finite concentration level of the absorbate on the rate of mass transfer	
$\theta_3$	$= \beta / \beta_0$ , correction factor which accounts for the effect of the finite concentration level of the absorbate on the rate of mass transfer in a case of nonisothermal absorption	
$\theta_4$	$= \beta / \beta^{is}$ , correction factor which characterizes an influence of temperature change on the rate of mass transfer in a case of the absorption with a finite concentration level of the absorbate	
$\lambda$	thermal conductivity of a liquid . . . . .	$kJ \cdot m^{-1} \cdot s^{-1} \cdot K^{-1}$
$\mu$	function determined by equation (15)	$m^4 \cdot s^{-1}$
$\mu_0$	shape factor for a gas plug . . . . .	$m^4 \cdot s^{-1}$
$\rho$	density of solution . . . . .	$kg \cdot m^{-3}$
$\phi$	velocity potential . . . . .	$m^2 \cdot s^{-1}$

**1. INTRODUCTION**

Mass and heat transfer between a rising bubble and a liquid is of great importance for design and analysis of two-phase absorbers. Examples of industrial operations where effects of heat release during absorption may be important include absorption of ammonia into water, absorption of carbon dioxide and hydrogen sulfide into amine solutions, water vapor absorption (hygroscopic condensation) by aqueous solutions of LiBr and LiCl and chlorinating of organic liquids. Commercial applicability of nonisothermal absorption in energy recovery schemes is highly dependent on the overall transfer rates which can be achieved. Examples of fast growing worldwide applications of thermal technology are refrigeration and air-conditioning. Energy efficiency and new fluids without adverse environmental effects are the two most important challenges that the latter technologies must overcome. Heat release effects are usually important, and hence a deeper understanding of the simultaneous heat and mass transfer is required in order to increase the efficiency and to reduce the environmental impact. One of the examples of absorption with a finite concentration level of the absorbate in the absorbent is water vapor absorption (hygroscopic condensation) by aqueous solutions of LiBr and LiCl. Hygroscopic condensation is governed by mass transfer during nonisothermal absorption. Brauner et al. [1, 2] and Brauner [3] evaluated the effect of the absorbate concentration in systems with a finite absorbate dilution on the rates of heat and mass transfer during nonisothermal absorption. At finite absorbate dilution the lateral convective term cannot be ignored. Brauner et al. [2] showed that a nonisothermal absorption by a falling liquid film in the approximation of the thin concentration and temperature boundary layers in the liquid phase can be described by the following equations of mass and heat transfer:

$$u_s \frac{\partial c_A}{\partial x} = D \frac{\partial}{\partial y} \left[ \frac{1}{(1 - c_A/c)} \frac{\partial c_A}{\partial y} \right] \quad (1)$$

$$u_s \frac{\partial T}{\partial x} = a \frac{\partial^2 T}{\partial y^2} \quad (2)$$

with the corresponding boundary conditions at the interface and at infinity. Here  $u_s$  is an interfacial velocity in the liquid film,  $c_A$  is absorbate concentration,  $c$  is a molar density,  $x$  and  $y$  are coordinates. The effect of a finite dilution is described by the ratio  $c_A/c \neq 0$  in equation (1). The above system of partial differential equations was reduced to the system of ordinary differential equations by using the similarity transformation. The solution of equation (1) was obtained by Brauner et al. [2] in the form of the Volterra integral equation while the solution of equation (2) has a form of the error function. It was shown that the enhancement factor  $\Omega$  which is equal to the ratio of the actual absorption mass transfer coefficient to that obtained under the assumption of the infinite dilution of the absorbate in absorbent is determined by the following expression:

$$\Omega = \frac{Sh}{Sh_0} = \frac{1}{3} \frac{\sqrt{\pi}}{(X_A^* - X_{Ai})} \left[ \frac{x}{\delta} \frac{Pe^*}{Le} \right]^{1/2} \quad (3)$$

where  $X_A^*$  is a molar fraction of absorbate at the gas–liquid interface,  $X_{Ai}$  is a molar fraction of the absorbate at the inlet,  $Pe^*$  is Peclet number for a falling liquid film (for details see [2]). Analysis of equation (3) shows that models using the assumption of the infinite absorbate dilution may considerably underestimate the rates of heat and mass transfer during hygroscopic condensation. The assumption of short exposure distance in a falling film (see [1, 2]) was abandoned by Brauner in [3]. Brauner [3] investigated the case of the developing concentration and thermal boundary layers in a liquid film downstream and took into account the increase of a liquid film thickness due to the hygroscopic condensation. Brauner [3] employed the integral transform approach for solving simultaneously the continuity, diffusion and energy equations for the cases of an isothermal and adiabatic wall.

Combined mass and heat transfer during pure gas absorption from a rising gas plug was investigated by Elperin and Fominykh [4] in the approximations of the thin thermal and concentration boundary layers in the liquid phase and of the infinite dilution of the absorbate. Expressions for mass and heat transfer coefficients obtained in [4] for the case of absorption without heat release recover the formulas for the isothermal absorption derived previously by van Heuven and Beek [5]. Mass transfer during nonisothermal absorption in a gas–liquid plug flow was investigated in [6–9]. Infante Ferreira [6, 7]

investigated combined heat and mass transfer in a vertical plug flow absorbers on the base of a system of equations of momentum, heat and mass transfer which were solved numerically. Elperin and Fominykh [8, 9] derived the recurrent formulas for the concentration and temperature in the  $n$ th liquid plug and determined expressions for mass and heat fluxes from the  $n$ th gas plugs and the total mass and heat fluxes from  $N$  gas plugs in a linear cluster of plugs under the assumption of a perfect liquid mixing in a liquid plug by a vortex and homogeneous distribution of temperature and concentration of the dissolved gas in each liquid plug. Case with the inhomogeneous distributions of temperature and concentration of the dissolved gas in each liquid plug was analyzed in [8]. Results obtained in [9] show that contribution of small spherical bubbles in the liquid plug to mass and heat transfer in gas–liquid plug flow is considerably higher than the contribution of a gas plug. It was also found that the increase of volumetric mass transfer coefficient with the increase of the gas superficial velocity measured in [10] is determined by the increase of a void fraction and a number of spherical gas bubbles in the liquid plug with a gas superficial velocity increase.

In all the above referenced studies heat and mass transfer during gas absorption from gas plugs was investigated in the approximation of the infinite dilution of the absorbate in the absorbent. In the present work gas absorption from a short rising gas plug is investigated in the approximation of finite dilution of the absorbate in the absorbent.

## 2. DESCRIPTION OF THE MODEL

Consider a gas plug rising with a constant velocity  $U_\infty$  in a vertical channel filled with liquid. Schematic view of a gas plug and a system of coordinates attached to a rising gas plug are shown in *figure 1*. Gas plug is assumed to be short, i.e.  $L_B \leq 2d_{ch}$ . In the vicinity of the nose and sides of a gas plug, where mass and heat transfer is to be calculated, the usual assumption of streamline flow is followed. Consider the coupled mass and heat transfer during absorption of a pure soluble gas from a rising gas plug in a channel accompanied by a heat release. The problem is solved in the approximation of a finite dilution of absorbate in the absorbent. The thermodynamic parameters of a system are assumed constant, and only the resistance to mass and heat transfer in the liquid phase is taken into account. Assume that heat released during absorption is dissipated in a liquid phase where it causes an increase of the liquid temperature.

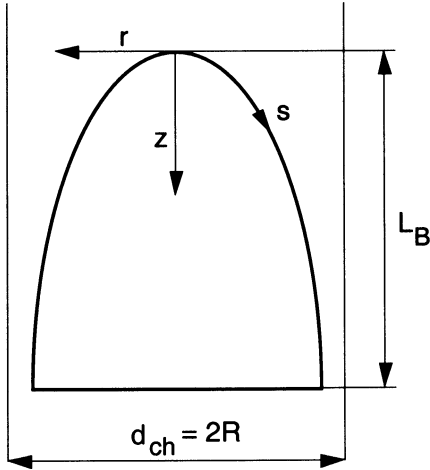


Figure 1. Schematic view of a short gas plug.

Thus, coupling between mass and heat transfer occurs. The equilibrium condition at the gas–liquid interface is described by a linear dependence of the concentration on temperature. Assume that development of thin diffusion and temperature boundary layers in a liquid starts at the leading edge of a gas plug, and convective diffusion and heat transfer are determined by fluid velocity at the gas plug surface. The gas plug–liquid interface is assumed to be a surface of revolution obtained by the rotation of a curve  $r(z)$  around a  $z$ -axis. The equation of a curve  $r(z)$  is derived in [11]. A differential mass balance equation for the absorbing component in cylindrical coordinates reads:

$$\frac{\partial N_{Az}}{\partial z} + \frac{1}{r} \frac{\partial}{\partial r} (r N_{Ar}) = 0 \quad (4)$$

Expression for mass flux of absorbate in radial direction  $N_{Ar}$  in equation (4) reads (see [12]):

$$N_{Ar} = -D\rho \frac{\partial x_A}{\partial r} + x_A(N_{Ar} + N_{B_r}) \quad (5)$$

Equation (5) shows that the diffusion flux of absorbate  $N_{Ar}$  in radial direction relative to stationary coordinates is a sum of two fluxes:  $x_A(N_{Ar} + N_{B_r})$  which is a mass flux of absorbate resulting from the bulk motion of fluid and  $-D\rho \partial x_A / \partial r$ , which is a mass flux of absorbate resulting from the diffusion superimposed on the bulk flow. Assume that the flux of the absorbent at gas–liquid interface is negligibly small and density of solution is constant. Then following Rice [13] we found the expression for the mass flux of the absorbate  $N_{Ar}$ :

$$N_{Ar} = -D\rho \frac{\partial x_A}{\partial r} + x_A N_{Ar}|_s + x_A \rho v_r \quad (6)$$

and at the interface

$$N_{Ar}|_s = -\frac{D\rho}{1 - x_{As}} \left( \frac{\partial x_A}{\partial r} \right)_s \quad (7)$$

Finally, expression for  $N_{Ar}$  can be written as follows:

$$N_{Ar} = -D\rho \frac{\partial x_A}{\partial r} - \frac{D\rho x_A}{1 - x_{As}} \left( \frac{\partial x_A}{\partial r} \right)_s + x_A \rho v_r \quad (8)$$

Following the same procedure as for  $N_{Ar}$  component we obtain the following formula for  $N_{Az}$ :

$$N_{Az} = -D\rho \frac{\partial x_A}{\partial z} - \frac{D\rho x_A}{1 - x_{As}} \left( \frac{\partial x_A}{\partial z} \right)_s + x_A \rho v_z \quad (9)$$

Equation of continuity in cylindrical coordinates reads:

$$\frac{\partial v_z}{\partial z} + \frac{1}{r} \frac{\partial}{\partial r} (r v_r) = 0 \quad (10)$$

Substituting expressions for  $N_{Ar}$  and  $N_{Az}$  (equations (8) and (9)) into equation (4) and using equation (10) we arrive at the following equation of convective diffusion in cylindrical coordinates:

$$\begin{aligned} v_r \frac{\partial x_A}{\partial r} + v_z \frac{\partial x_A}{\partial z} = D \left( \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial x_A}{\partial r} \right) + \frac{\partial^2 x_A}{\partial z^2} \right) \\ + D \left( \frac{1}{1 - x_{As}} \left( \frac{\partial x_A}{\partial r} \right)_s \frac{1}{r} \frac{\partial}{\partial r} (r x_A) \right) \\ + \frac{1}{1 - x_{As}} \left( \frac{\partial x_A}{\partial z} \right)_s \frac{\partial x_A}{\partial z} \end{aligned} \quad (11)$$

For small mass transfer rates the velocity components  $v_r$  and  $v_z$  in equation (11) can be determined from the solution of the hydrodynamic problem with zero mass flux at the interface. Validity of such an approach was demonstrated by Bird et al. [12] for sublimation of a semi-infinite plate of a volatile solid into an unbounded gaseous stream. In order to simplify equation (11) we introduce the stream function  $\psi$  and velocity potential  $\phi$  as follows:

$$\begin{aligned} v_z &= \frac{1}{r} \frac{\partial \psi}{\partial r} = \frac{\partial \phi}{\partial z} \\ v_r &= -\frac{1}{r} \frac{\partial \psi}{\partial z} = \frac{\partial \phi}{\partial r} \end{aligned}$$

Using the assumption of potential flow

$$\begin{aligned} \frac{\partial^2 \phi}{\partial z^2} + \frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r} \frac{\partial \phi}{\partial r} = 0 \\ \frac{\partial^2 \psi}{\partial z^2} + \frac{\partial^2 \psi}{\partial r^2} - \frac{1}{r} \frac{\partial \psi}{\partial r} = 0 \end{aligned} \quad (12)$$

equation (11) can be rewritten as follows:

$$\begin{aligned} \frac{\partial x_A}{\partial \phi} = D \left\{ r^2 \frac{\partial^2 x_A}{\partial \psi^2} + \frac{r^2}{1-x_{A_s}} \left( \frac{\partial x_A}{\partial \psi} \right)_s \frac{\partial x_A}{\partial \psi} \right. \\ + D \left\{ \frac{\partial^2 x_A}{\partial \phi^2} + \frac{1}{1-x_{A_s}} \left( \frac{\partial x_A}{\partial \phi} \right)_s \frac{\partial x_A}{\partial \phi} \right. \\ + \frac{x_A v_r}{(1-x_{A_s})v^2 r} \left( \frac{\partial x_A}{\partial \phi} \right)_s + \frac{2v_z}{v^2} \frac{\partial x_A}{\partial \psi} \\ \left. \left. + \frac{x_A v_z}{(1-x_{A_s})v^2} \left( \frac{\partial x_A}{\partial \psi} \right)_s \right\} \right\} \quad (13) \end{aligned}$$

where  $v^2 = v_r^2 + v_z^2$ . Equation (13) can be simplified by neglecting molecular diffusion along the streamlines so that the third, the fourth and the fifth terms in the right-hand side of equation (13) can be omitted. Assuming that diffusion is confined to a thin film surrounding the gas plug and taking into consideration that  $Pe \gg 1$ , where  $Pe = U_\infty d_{ch}/D$ ,  $U_\infty = 0.35\sqrt{gd_{ch}}$  (see, e.g., [14]), we neglect the sixth and the seventh terms in the r.h.s. of equation (13). The validity of all these assumptions is discussed in the Appendix. Introducing these simplifications equation (13) can be reduced to

$$\frac{\partial x_A}{\partial \phi} = D \left\{ r^2 \frac{\partial^2 x_A}{\partial \psi^2} + \frac{r^2}{1-x_{A_s}} \left( \frac{\partial x_A}{\partial \psi} \right)_s \frac{\partial x_A}{\partial \psi} \right\} \quad (14)$$

Taking into account that diffusion boundary layer is very thin, and following [15, 16] we assume that  $r$  is a function of  $\phi$  only. Introduce a new variable  $\mu$  defined by  $\partial\mu/\partial\phi = r^2$ . Function  $\mu$  is determined by integration along the gas plug surface,  $s$  being the length of arc measured from the gas plug nose (see *figure 1*) and  $v = \partial\phi/\partial s$  (for details see [15, 16]):

$$\mu \left( \frac{z}{d_{ch}} \right) = \int_{\phi_0}^{\phi} r^2 d\phi' = \int_0^s r^2 v ds' \quad (15)$$

Using simple geometric arguments we find that

$$\begin{aligned} \mu_0 = \mu \left( \frac{\delta L_B}{\delta d_{ch}} \right) \\ = \left( \int_0^{L_B/d_{ch}} \left( \frac{r(z)}{d_{ch}} \right)^2 \sqrt{1 + \left( \frac{dr(z)/d_{ch}}{dz/d_{ch}} \right)^2} \right. \\ \left. \cdot \sqrt{\frac{2gz}{d_{ch}}} d \left( \frac{z}{d_{ch}} \right) \right)^{1/2} \quad (16) \end{aligned}$$

where  $\mu_0$  is a shape factor of a gas plug which was introduced in [5] and tabulated in [15, 16]. Combining equations (14) and (15) yields:

$$\frac{\partial x_A}{\partial \mu} = D \frac{\partial^2 x_A}{\partial \psi^2} + \frac{D}{1-x_{A_s}} \left( \frac{\partial x_A}{\partial \psi} \right)_{\psi=0} \frac{\partial x_A}{\partial \psi} \quad (17)$$

The last term in the r.h.s. of equation (17) accounts for the effects of the finite concentration level of the absorbate. This term is not equal to zero in the approximation of a small but finite concentration level of the absorbate ( $x_{A_s} \ll 1$ ), and it vanishes in the approximation of the infinite dilution of the absorbate in absorbent ( $x_{A_s} \rightarrow 0$ ). Following similar approach we arrived at the energy equation taking into account that  $Le \ll 1$ :

$$\frac{\partial T}{\partial \mu} = a \frac{\partial^2 x_A}{\partial \psi^2} \quad (18)$$

Initial and boundary conditions which accompany equations (17) and (18) read:

$$T = T_0, \quad x_A = x_{A_0} \quad \text{at } \mu = 0 \quad (19)$$

$$T = T_s, \quad x_A = x_{A_s} \quad \text{at } \psi = 0 \quad (20)$$

$$T = T_0, \quad x_A = x_{A_0} \quad \text{at } \psi \rightarrow \infty \quad (21)$$

Introducing new variables

$$X = \frac{x_A - x_{A_0}}{x_{A_s} - x_{A_0}}, \quad \Psi = \frac{\psi}{2\sqrt{D\mu}}$$

equations (17), (19)–(21) can be transformed as follows:

$$\frac{d^2 X}{d\Psi^2} + 2(\Psi - \varphi) \frac{dX}{d\Psi} = 0 \quad (22)$$

$$X = 1 \quad \text{at } \Psi = 0 \quad (23)$$

$$X = 0 \quad \text{at } \Psi \rightarrow \infty \quad (24)$$

where

$$\varphi = -\frac{1}{2} \frac{x_{A_s} - x_{A_0}}{1 - x_{A_s}} \left( \frac{dX}{d\Psi} \right)_{\Psi=0} \quad (25)$$

Solution of equation (22) with boundary conditions (23), (24) reads (see [12]):

$$\frac{x_A - x_{A_0}}{x_{A_s} - x_{A_0}} = \frac{1 - \operatorname{erf}(\Psi - \varphi)}{1 + \operatorname{erf} \varphi} \quad (26)$$

where  $\varphi$  is a dimensionless mass average velocity. Using equations (25) and (26) we find that

$$\left( \frac{dX}{d\Psi} \right)_{\Psi=0} = - \left[ \frac{\sqrt{\pi}}{2} e^{\varphi^2} (1 + \operatorname{erf} \varphi) \right]^{-1} \quad (27)$$

$$\frac{x_{A_s} - x_{A_0}}{1 - x_{A_s}} = \varphi \sqrt{\pi} e^{\varphi^2} (1 + \operatorname{erf} \varphi) = 2\varphi f(\varphi) \quad (28)$$

$$x_{A_s} = \frac{2\varphi f(\varphi) + x_{A_0}}{1 + 2\varphi f(\varphi)} \quad (29)$$

Taking into consideration that

$$Q_c^{is} = -2\pi\rho D \int_0^{\mu_0} \frac{1}{1-x_{A_s}} \left( \frac{\partial x_A}{\partial \psi} \right)_{\psi=0} d\mu$$

$$= 4\pi D^{1/2} \rho \varphi \mu_0^{1/2} \quad (30)$$

$$\beta^{is} = \frac{Q_c^{is}}{S\rho(x_{A_s} - x_{A_0})} = \frac{4\pi D^{1/2} \varphi \mu_0^{1/2}}{S(x_{A_s} - x_{A_0})} \quad (31)$$

and (see [5])

$$Q_{c_0}^{is} = -2\pi\rho D \int_0^{\mu_0} \left( \frac{\partial x_A}{\partial \psi} \right)_{\psi=0} d\mu$$

$$= 4\pi^{1/2} D^{1/2} \rho \mu_0^{1/2} (x_{A_s} - x_{A_0}) \quad (32)$$

$$\beta_0^{is} = \frac{Q_{c_0}^{is}}{S\rho(x_{A_s} - x_{A_0})} = 4\pi^{1/2} D^{1/2} \mu_0^{1/2} S^{-1} \quad (33)$$

we find that

$$\theta_1 = \frac{\beta^{is}}{\beta_0^{is}} = \frac{Q_c^{is}}{Q_{c_0}^{is}} = \frac{\sqrt{\pi} \varphi}{x_{A_s} - x_{A_0}} = \frac{\sqrt{\pi}}{2f(\varphi)(1-x_{A_s})} \quad (34)$$

where  $\theta_1$  is the correction factor which accounts for the effect of the finite concentration level of the absorbate on the rate of mass transfer for a case of isothermal absorption. If  $x_{A_0} = 0$  we can rewrite equation (34) as follows:

$$\theta_1 = \frac{\sqrt{\pi} (1 + 2\varphi f(\varphi))}{2f(\varphi)} \quad (35)$$

Function  $f(\varphi)$  and correction factor  $\theta_1$  determined by equations (28) and (35) are presented in *table I*. Function  $f(\varphi)$  and correction factor  $\theta_1$  are presented in *table II*

TABLE I

$\varphi$	$f(\varphi)$	$2\varphi f(\varphi)$	$\theta_1$
0.0	0.8862	0.0	1.0
0.001	0.8872	0.002	1.0009
0.005	0.8912	0.0089	1.0033
0.01	0.8963	0.0179	1.006
0.1	0.9957	0.1992	1.0673
0.2	1.1278	0.4511	1.1403

for absorption of different gases in water at normal temperature and pressure conditions (NTP). Inspection of *table II* shows that absorption of CO<sub>2</sub>, H<sub>2</sub>S, Cl<sub>2</sub> by water can be described using the approximation of the infinite dilution of absorbate in the absorbent ( $x_{A_s} \rightarrow 0$  and  $\varphi \rightarrow 0$ ). The same is valid for N<sub>2</sub> and O<sub>2</sub> absorption by water at NTP. However, in order to describe SO<sub>2</sub> absorption by water at NTP the approximation of the finite dilution of the absorbate in the absorbent ( $\varphi \ll 1$  and  $x_{A_s} \ll 1$ ) must be used.

The solution of equation (18) with initial and boundary conditions (19)–(21) reads:

$$T = (T_0 - T_s) \operatorname{erf}(\Psi') + T_s \quad (36)$$

where  $\Psi' = \psi/(2\sqrt{a\mu})$ . For the case of nonisothermal absorption the unknown values of concentration and temperature at the gas–liquid interface are found from the following equations:

$$x_A = dT + b \quad \text{at } \psi = 0 \quad (37)$$

$$\lambda \int_0^{\mu_0} \left( \frac{\partial T}{\partial \psi} \right)_{\psi=0} d\mu$$

$$= L\rho D \int_0^{\mu_0} \frac{1}{1-x_{A_s}} \left( \frac{\partial x_A}{\partial \psi} \right)_{\psi=0} d\mu \quad \text{at } \psi = 0 \quad (38)$$

Equation (37) describes a condition of equilibrium at the gas–liquid interface (see, e.g., [17]), and equation (38) implies that all heat released during absorption is dissipated in the liquid phase. Note that for most gases coefficient  $d$  in equation (37) is negative. Equations (26), (36)–(38) yield expressions for  $T_s$  and  $x_{A_s}$ :

$$T_s - T_0 = \frac{T'_0 - T_0}{1 - K/(\theta_1\sqrt{Le})} \quad (39)$$

$$x_{A_s} - x_{A_0} = \frac{x'_{A_0} - x_{A_0}}{1 - \theta_1\sqrt{Le}/K}$$

where  $Le = D/a$ ,  $K = c_p/(dL)$ ,  $x'_{A_0} = dT_0 + b$  is the equilibrium concentration at the initial temperature,  $T'_0 = (x_{A_0} - b)/d$  is the equilibrium temperature at the initial concentration. Mass flux from a gas plug

TABLE II

Gas	$x_{A_s}$	$\varphi$	$f(\varphi)$	$2\varphi f(\varphi)$	$\theta_1$	$Le$	$K$
CO <sub>2</sub>	$1.69 \cdot 10^{-3}$	0.001	0.8872	0.002	1.00099	$1.37 \cdot 10^{-2}$	120
H <sub>2</sub> S	$3.85 \cdot 10^{-3}$	0.002	0.8882	0.00355	1.0043	$10^{-2}$	42
Cl <sub>2</sub>	$7.29 \cdot 10^{-3}$	0.004	0.8901	0.0071	1.0027	$8.7 \cdot 10^{-3}$	33
SO <sub>2</sub>	0.113	0.07	0.9607	0.1345	1.0465	$1.05 \cdot 10^{-2}$	0.58



and mass transfer coefficient can be determined from equations (26) and (33):

$$\begin{aligned} Q_c &= -2\pi\rho D \int_0^{\mu_0} \frac{1}{1-x_{A_s}} \left( \frac{\partial x_A}{\partial \psi} \right)_{\psi=0} d\mu \\ &= \frac{\theta_1 S \beta_0^{\text{is}}}{1 - \theta_1 \sqrt{Le}/K} (x'_{A_0} - x_{A_0}) \end{aligned} \quad (40)$$

Taking into consideration that

$$\beta = \frac{Q_c}{(x'_{A_0} - x_{A_0})\rho S} \quad (41)$$

equations (40) and (41) yield the correction factor  $\theta_2$  which accounts for the effects of temperature change and finite concentration level of the absorbate on mass transfer rate during absorption:

$$\theta_2 = \frac{\beta}{\beta_0^{\text{is}}} = \frac{1}{\theta_1^{-1} - \sqrt{Le}/K} \quad (42)$$

where  $\beta_0^{\text{is}} = 4\pi^{1/2} D^{1/2} \mu_0^{1/2} S^{-1}$  (see [5]). Taking into account (for details see [4]) that

$$\beta_0 = \frac{Q_{c_0}}{S\rho(c'_0 - c_0)} = \frac{\beta_0^{\text{is}}}{1 - \sqrt{Le}/K} \quad (43)$$

equations (31), (40), (41) and (43) yield

$$\theta_3 = \frac{\beta}{\beta_0} = \frac{1 - \sqrt{Le}/K}{\theta_1^{-1} - \sqrt{Le}/K} \quad (44)$$

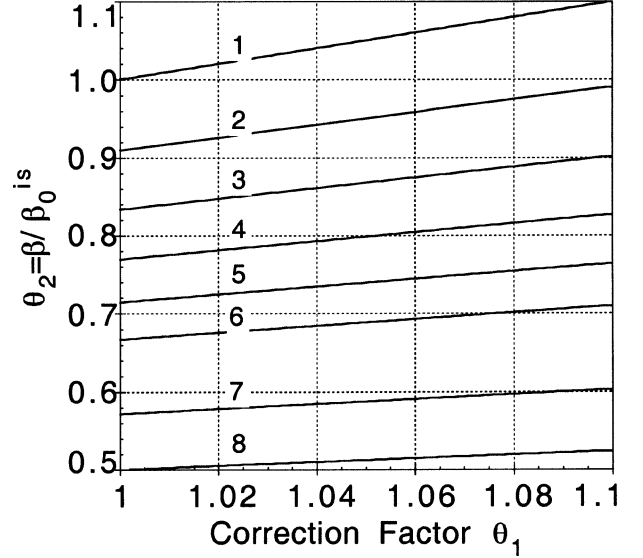
and

$$\theta_4 = \frac{\beta}{\beta_0^{\text{is}}} = \frac{\theta_1^{-1}}{\theta_1^{-1} - \sqrt{Le}/K} \quad (45)$$

where  $\theta_3$  is the correction factor which accounts for the effect of a finite concentration level of the absorbate on the rate of mass transfer for a case of nonisothermal absorption, and  $\theta_4$  is the correction factor which accounts for the effect of temperature change on the rate of mass transfer for a case of absorption with a finite concentration level of the absorbate. Heat flux and heat transfer coefficient can be determined from equations (36) and (39) as follows:

$$\begin{aligned} Q_T &= -2\pi\lambda \int_0^{\mu_0} \left( \frac{\partial T}{\partial \psi} \right)_{\psi=0} d\mu \\ &= \frac{4\pi^{1/2} \lambda a^{-1/2} \mu_0^{1/2}}{1 - K/(\theta_1 \sqrt{Le})} (T'_0 - T_0) \end{aligned} \quad (46)$$

$$\alpha = \frac{Q_T}{S(T'_0 - T_0)} = \frac{4\pi^{1/2} \lambda a^{-1/2} \mu_0^{1/2} S^{-1}}{1 - K/(\theta_1 \sqrt{Le})} \quad (47)$$



**Figure 2.** Dependence of  $\theta_2 = \beta/\beta_0^{\text{is}}$  versus  $\theta_1$ . (1)  $\sqrt{Le}/|K| = 0$ ; (2)  $\sqrt{Le}/|K| = 0.1$ ; (3)  $\sqrt{Le}/|K| = 0.2$ ; (4)  $\sqrt{Le}/|K| = 0.3$ ; (5)  $\sqrt{Le}/|K| = 0.4$ ; (6)  $\sqrt{Le}/|K| = 0.5$ ; (7)  $\sqrt{Le}/|K| = 0.75$ ; (8)  $\sqrt{Le}/|K| = 1.0$ .

Taking into consideration that (see [4])

$$\alpha_0 = \frac{Q_{T_0}}{S(T'_0 - T_0)} = \frac{4\pi^{1/2} \lambda a^{-1/2} \mu_0^{1/2}}{S(1 - K/\sqrt{Le})} \quad (48)$$

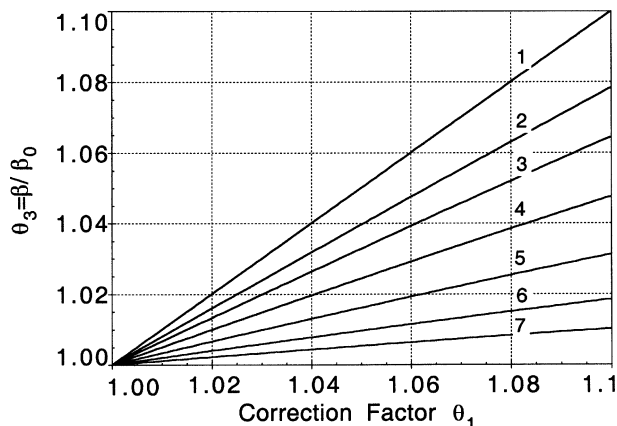
equations (47) and (48) yield

$$\frac{\alpha}{\alpha_0} = \frac{1 - \sqrt{Le}/K}{\theta_1^{-1} - \sqrt{Le}/K} = \theta_3 \quad (49)$$

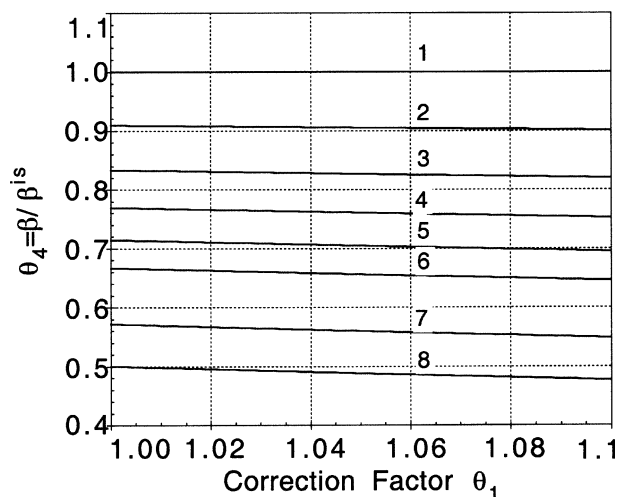
Dependences of  $\theta_2$ ,  $\theta_3$  and  $\theta_4$  versus  $\theta_1$  for different values of parameter  $\sqrt{Le}/|K|$  are shown in figures 2–4.

### 3. DISCUSSION AND CONCLUSIONS

We showed that for gas absorption from a rising gas plug when the absorbate concentration level is finite the convective term in equation of diffusion in the lateral direction must be taken into account. It is found that the correction factor  $\theta_1$  which accounts for the effect of the finite concentration level of the absorbate for a case of isothermal absorption depends only on gas solubility and initial concentration of the absorbate in the absorbent. Equation (40) implies the increase of the transfer rate with the increase of the concentration level. The correction factor  $\theta_1$  accounts for the enhancement of mass transfer coefficient due to the effect of the



**Figure 3.** Dependence of  $\theta_3 = \beta/\beta_0$  versus  $\theta_1$ . (1)  $\sqrt{Le}/|K| = 0$ ; (2)  $\sqrt{Le}/|K| = 0.25$ ; (3)  $\sqrt{Le}/|K| = 0.5$ ; (4)  $\sqrt{Le}/|K| = 1$ ; (5)  $\sqrt{Le}/|K| = 2$ ; (6)  $\sqrt{Le}/|K| = 4$ ; (7)  $\sqrt{Le}/|K| = 8$ .



**Figure 4.** Dependence of  $\theta_4 = \beta/\beta^{is}$  versus  $\theta_1$ . (1)  $\sqrt{Le}/|K| = 0$ ; (2)  $\sqrt{Le}/|K| = 0.1$ ; (3)  $\sqrt{Le}/|K| = 0.2$ ; (4)  $\sqrt{Le}/|K| = 0.3$ ; (5)  $\sqrt{Le}/|K| = 0.4$ ; (6)  $\sqrt{Le}/|K| = 0.5$ ; (7)  $\sqrt{Le}/|K| = 0.75$ ; (8)  $\sqrt{Le}/|K| = 1.0$ .

additional convective term in equation (14). Increase of  $\theta_1$  with the increase of gas solubility is shown in *tables I and II*. Equations (39) show that the interfacial equilibrium temperature increases with the increase of dimensionless heat of absorption  $\sqrt{Le}/|K|$  while the corresponding concentration decreases. The latter is valid for most gases when  $d < 0$  and  $K < 0$ . The effect of heat and mass coupling parameter  $\sqrt{Le}/|K|$  on the rates of mass transfer is shown in *figures 2 and 3*. For small  $\sqrt{Le}/|K|$  mass transfer dominates, and the concentration level effects significantly the relative coefficient of mass transfer. For large  $\sqrt{Le}/|K|$  heat transfer dominates, and

the effect of the concentration level vanishes. Thus, the slopes of the curves in *figures 2 and 3* decrease with the increase of the dimensionless heat of absorption  $\sqrt{Le}/|K|$ . Inspection of *figure 4* shows that the increase of heat release causes the reduction of the rate of mass transfer during absorption. The latter effect becomes more pronounced with the increase of concentration level of the absorbate in the absorbent. Inspection of the results presented in *table II* shows that absorption of such commonly used gases as  $\text{CO}_2$ ,  $\text{H}_2\text{S}$  and  $\text{Cl}_2$  by water at NTP can be described in the approximation of the infinite dilution of absorbate in the absorbent ( $\varphi \rightarrow 0$  and  $x_{A_s} \rightarrow 0$ ). The same is valid for  $\text{N}_2$  and  $\text{O}_2$  absorption by water. However, absorption of  $\text{SO}_2$  by water at NTP can be described only using an approximation of the finite dilution of the absorbate in the absorbent ( $\varphi \ll 1$  and  $x_{A_s} \ll 1$ ).

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**APPENDIX**

Consider the simplifying assumptions used in deriving equation (14). The second term in the right-hand side of equation (13) is retained, i.e. it was assumed that

$$R_1 = \left[ \frac{1}{1-x_{As}} \left( \frac{\partial x_A}{\partial \psi} \right)_s \frac{\partial x_A}{\partial \psi} \right] \left( \frac{\partial^2 x_A}{\partial \psi^2} \right)^{-1} \sim 1 \quad (A.1)$$

The third, the fourth, the fifth, the sixth and the seventh terms in the right-hand side of equation (13) were neglected, i.e. it was assumed that

$$R_2 = \left( \frac{\partial^2 x_A}{\partial \phi^2} \right) \left( r^2 \frac{\partial^2 x_A}{\partial \psi^2} \right)^{-1} \ll 1 \quad (A.2)$$

$$R_3 = \left[ \frac{1}{1-x_{As}} \left( \frac{\partial x_A}{\partial \phi} \right)_s \frac{\partial x_A}{\partial \phi} \right] \left( r^2 \frac{\partial^2 x_A}{\partial \psi^2} \right)^{-1} \ll 1 \quad (A.3)$$

$$R_4 = \left[ \frac{x_A v_r}{(1-x_{As})v^2 r} \left( \frac{\partial x_A}{\partial \phi} \right)_s \right] \left( r^2 \frac{\partial^2 x_A}{\partial \psi^2} \right)^{-1} \ll 1 \quad (A.4)$$

$$R_5 = \left[ \frac{2v_z}{v^2} \frac{\partial x_A}{\partial \psi} \right] \left( r^2 \frac{\partial^2 x_A}{\partial \psi^2} \right)^{-1} \ll 1 \quad (A.5)$$

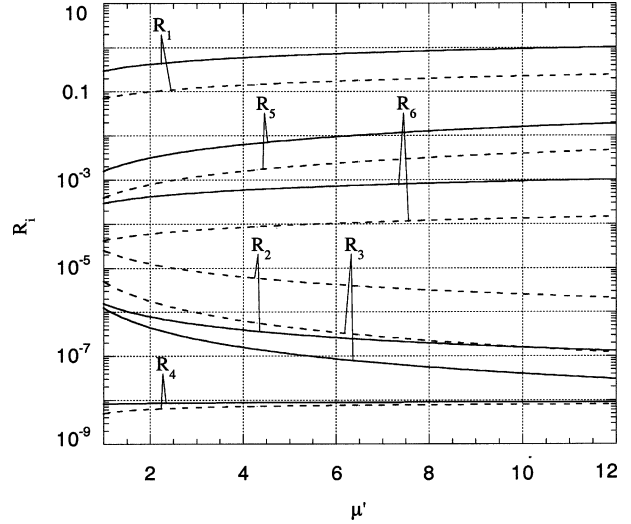


Figure 5. Dependence of  $R_i$  ( $i = 1, \dots, 6$ ) versus  $\mu'$  along two streamlines  $\psi'$ : dashed curves,  $2.5 \cdot 10^{-3}$ ; solid curves,  $10^{-2}$ .

$$R_6 = \left[ \frac{x_A v_z}{(1-x_{As})v^2} \left( \frac{\partial x_A}{\partial \psi} \right)_s \right] \left( r^2 \frac{\partial^2 x_A}{\partial \psi^2} \right)^{-1} \ll 1 \quad (A.6)$$

The terms  $R_1, \dots, R_6$  were obtained by differentiating equation (26) and following the approach employed in [18]. Figure 5 shows the obtained values of  $R_1, \dots, R_6$  plotted against  $\mu'$  for representative values of  $\psi'$  and for  $Pe = 2.74 \cdot 10^5$ , where  $\psi' = \psi / (U_\infty R^2)$  and  $\mu' = \mu / (U_\infty R^3)$ . Taking into consideration that the coefficient of molecular diffusion for all gases in liquids is of the order of  $D \sim 10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ , Peclet number for gas plug, dissolving in liquid is always high and it is of the order of  $Pe \sim 10^5$ . The value of  $Pe = 2.74 \cdot 10^5$  corresponds to a gas slug of  $\text{CO}_2$  rising in a channel ( $d_{ch} = 1 \text{ cm}$ ) filled with water. Results presented in figure 5 show that it is reasonable to assume that  $R_1 \sim 1$  while  $R_2, \dots, R_6 \ll 1$  for small values of  $D$  (large  $Pe$ ).