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Conjugate mass transfer during gas absorption by falling liquid droplet with internal circulation

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Abstract

We considered conjugate mass transfer during absorption of a gas by a falling droplet with internal circulation. Gaseous phase is assumed to contain inert admixtures, and resistance to mass transfer in both phases is taken into account. Mass flux is directed from a gaseous phase to a droplet, and the interfacial shear stress causes a fluid flow inside the droplet. Droplet deformation under the influence of the interface shear stress is neglected. Absorbate accumulation in the bulk of dispersed phase is taken into account. The problem is solved in the approximations of a thin concentration boundary layer in the dispersed and continuous phases. The bulk of a droplet, beyond the diffusion boundary layer is completely mixed, and concentration of absorbate is homogeneous and time-dependent in the bulk. The thermodynamic parameters of a system are assumed constant. By combining the generalized similarity transformation method with Duhamel's theorem, the system of transient conjugate equations of convective diffusion for absorbate transport in liquid and gaseous phases with time-dependent boundary conditions is reduced to Volterra integral equation of the second kind which is solved numerically. Theoretical results are compared with the available experimental data.

Keywords: Diffusion; Gas absorption; Droplet; Mass transfer; Inert admixtures; Internal circulation

1. Introduction

Removal of gases from polluted air and gaseous streams by water drops is an important mass transfer operation in air pollution control and engineering. Mass transfer to droplets is of fundamental importance in many naturally occurring phenomena and industrial processes involving sprays, e.g. atmospheric physics, wet deposition. Gas scavenging by atmospheric water droplets include absorption of SO_2, CO_2, HCl, NH_3, NO_x (nitric oxide and nitrogen dioxide), CFCs, methane and some other gases. Sources of these gases in the atmosphere are briefly reviewed in the following.

Sulfur dioxide (SO_2) is emitted from a smokestack as a result of various fossil fuels combustion, e.g. crude oil and coal. Gaseous SO_2 usually contains small amounts of SO_3, and then it is converted to sulfuric acid in the presence of water, forming acid fogs and acid rains. Fundamental mechanism of the acid rain formation partially results from sulfur oxidation and absorption by water droplets in the atmosphere. Comprehensive

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Nomenclature

D_i molecular diffusion coefficient, $m^2 s^{-1}$
D square root of the diffusivities ratio, $\sqrt{D_1/D_2}$
k coefficient in Eq. (1)
$N_i = -D_i\rho_i(\partial x_i/\partial y)$ mass flux density, $kg\,m^{-2}\,s^{-1}$
m distribution coefficient
$Pe_i = kRU/D_i$ Peclet number for a moving droplet
r radial coordinate, m
R droplet radius, m
$Re = 2UR\rho_2/\mu_2$ external flow Reynolds number for a moving droplet
$Sc = v_i/D_i$ Schmidt number
t time, s
$T = tUk/R$ dimensionless time
U translational velocity of a droplet, $m\,s^{-1}$
v_r, v_θ velocity components, $m\,s^{-1}$
V droplet volume, m^3
x mass fraction of an absorbate
x_{10} initial value of mass fraction of absorbate in a droplet
$X_1 = (x_1 - mx_2(\infty))/(x_{10} - mx_2(\infty))$ dimensionless mass fraction of an absorbate in the liquid phase
$X_2 = (x_2 - x_2(\infty))/(x_{10} - mx_2(\infty))$ dimensionless mass fraction of an absorbate in the gaseous phase
$X_b(\tau_1) = (x_b(t) - mx_2(\infty))/(x_{10} - mx_2(\infty))$ dimensionless mass fraction of an absorbate in the bulk of a droplet
y distance from the surface of a droplet, m
$Y = y/R$ dimensionless distance from the surface of a droplet

Greek symbols

$\gamma = \rho_1/\rho_2$ densities ratio
δ_i thickness of a diffusion boundary layer, m
$\Delta_i = \delta_i/R$ dimensionless thickness of a diffusion boundary layer
λ variable, Eqs. (16), (17)
η_i similarity variable, Eq. (7)
θ angle, rad
μ_i dynamic viscosity of a fluid, $kg\,m^{-1}\,s^{-1}$
v_i kinematic viscosity of a fluid, $m^2\,s^{-1}$
ρ_i density at the bulk of fluid, $kg\,m^{-3}$
$\tau_i = tD_i/R^2$ dimensionless time

Subscripts

0 value at the inlet
1 liquid phase
2 gaseous phase
∞ value at infinity
b value in the bulk of a droplet
r radial direction
s value at the interface
θ tangential direction

description of this particular mass transfer process is conducive to understanding the wet acid deposition in the environment.

Oxides of nitrogen (NO_x) such as nitric oxide and nitrogen dioxide, and carbon dioxide (CO_2) may be generated by different types of combustors, e.g., boilers and furnaces. The major source of CO_2 on geologic time scales is volcanic activity.

The main source of atmospheric ammonia is agriculture (see, e.g. Van Der Hoek, 1998), and the remaining minor sources are industries, humans, pets, wild animals, landfills and households products (see Sutton et al., 2000). Agricultural emissions of ammonia are associated mainly with animals waste (mainly cattle, poultry, pigs and sheep) and with fertilizers, mainly ammonium sulfate, ammonium nitrate, ammonia, urea and ammonium phosphate, (see, e.g. Buijsman and Erisman, 1988). The contribution of vehicles to non-agricultural NH_3 emissions has been considered to be negligible until 1995 (Sutton et al., 1995). In the last few years, however, an increase of NH_3 emission due to the introduction of petrol-engine vehicles equipped with catalytic converters has been reported in the literature (see Moeckli and Sigrist, 1996; Fraser and Cass, 1998).

Hydrogen chloride (HCl) is generated during combustion of municipal solid waste (MSW) containing certain types of plastics (Rhyner et al., 1995, pp. 262–267). When the hydrogen chloride is transferred to particles containing moisture, strong acids are formed. These acidic aerosols may cause serious corrosion and erosion of the equipment. In order to remove this hazardous air pollutant (HAP), water is often blended with basic chemicals such as NaOH and KOH to wash the flue gas. As a result, HCl is captured from the flue gas into the absorbing material.

Major sources of methane are such as rice agriculture, cattle and biomass burning.

Chlorofluorocarbons (CFCs) have been used for many applications, but mostly for refrigeration and air conditioning. In earlier years, they were used as aerosol propellants, and in more recent years they have been used for blowing polyurethane foams. A large amount

of CFC-11 is believed to be tied up in closed-cell polyurethane foams used for insulation in homes and buildings.

Due to differences in solubility of atmospheric gases in water and due to differences in the initial concentrations of the absorbed gases in the gaseous phase mass transfer during absorption of one atmospheric gas by water droplets in the presence of inert admixture can be continuous-phase controlled, liquid phase controlled or conjugate. Continuous-phase controlled mass transfer by falling droplets was discussed in a survey by Kaji et al. (1985) and by Altwicker and Lindhjem (1988) while liquid-phase controlled mass transfer was discussed, e.g., by Amokrane and Caussade (1999). Majority of theoretical approaches for investigating gaseous-phase mass transfer are based on quasi-steady state assumption which employs the steady-state values of the instantaneous continuous phase mass transfer coefficient together with an unsteady-state mass balance (see, e.g. Hales (1972, 1978), Waltrop et al. (1991), Saboni and Alexandrova, (2001)). Expression for the stationary coefficient of mass transfer in a gaseous phase is usually adopted by different authors from Frössling (1938) or Garner and Keey (1958) for mass transfer from a falling solid sphere for $Re \gg 1$ and $Pe \gg 1$ or from the theoretical analysis of Levich (1962) for mass transfer from a falling sphere for $Pe \gg 1$ and small Reynolds numbers. For small Peclet numbers, the correlation of Acrivos and Taylor (1962) is often used while for intermediate Peclet numbers different interpolation formulas are often employed (see, e.g. Zhang and Davis (1987)). Note that all usually used theoretical correlations for stationary mass transfer in a continuous phase are derived for a constant value of concentration of the transferred component at the interface between a droplet and gas, and in all the existing approaches the internal circulation in liquid droplets is not taken into account.

LeClair et al. (1972) found that falling drops in air exhibit a vigorous internal circulation, and that such circulation significantly affects the transport of solvable gases across gas–liquid interface and into the interior of a drop. The maximum internal velocity near the droplet surface was found to be close to $u_s \approx (1/25)U$, where U is the terminal fall velocity of the droplet. Although the flow fields inside and outside a falling liquid droplet in a gas were available since 1972 (see LeClair et al. (1972)), Baboolal et al. (1981) was the first to incorporate them into a model of gas absorption. Baboolal et al. (1981) integrated numerically the convective diffusion equation using the flow fields generated by an accurate numerical solution of the steady state Navier–Stokes equations for a flow around a falling droplet. Walcek and Pruppacher (1984a) solved simultaneously the convective diffusion equations for the diffusion of a trace gas outside and inside the droplet. Their computations showed that for SO_2 concentrations in air in the range of ppb SO_2, absorption by a falling water droplet is gaseous-phase controlled process while for concentrations in the percent range the resistance to diffusion is almost entirely in the liquid phase. At intermediate concentration of SO_2 diffusion resistance to mass transfer in both phases must be taken into account. Computations of Walcek and Pruppacher (1984a) also showed that at ambient SO_2 concentrations in ppb(v) range, droplets larger than a few tens of microns cannot be considered in equilibrium with SO_2. Fractional liquid resistance to mass transfer for droplets falling in a gaseous phase with various concentrations of SO_2 was analyzed by Alexandrova et al. (2004) using a quasistationary model for mass transfer. Alexandrova et al. (2004) showed that the fractional liquid resistance is independent of the droplet diameter. On the other hand, Alexandrova et al. (2004) found that at high concentration of SO_2 in the gaseous phase ($>1\%$) the internal resistance to diffusion dominates while at low concentrations of an absorbate (<1000 ppb) the external resistance to diffusion dominates.

Effect of a droplet's internal circulation on the rate of liquid phase controlled SO_2 absorption was calculated numerically and analyzed by Chen (2001a). Considering the absorption of sulfur dioxide by a droplet in a cloud or fog with various velocities, three different Reynolds numbers, viz., $Re = 0.643, 1.287$, and 12.87 were studied and the results were compared. The results of Chen (2001a) indicate that for the Reynolds number of 0.643, sulfur dioxide always penetrates toward the droplet centerline throughout the entire absorption period. The latter behavior is explained by that mass transfer is determined by diffusion along the radial direction. In contrast, when the Reynolds number is 12.87, the strength of the vortex flow inside the droplet is strong. Because of this circulation, most of the time the concentration contours are parallel to the streamlines, and the lowest SO_2 concentration is attained at the vortex center. As a consequence, the diffusion distance is reduced by a factor of three, and the absorption time required for a droplet to reach saturation is considerably reduced. Note that Chen (2001a) investigated SO_2 absorption from a mixture of gases containing an inert gas by a water droplet with internal circulation as a liquid-phase controlled mass transfer. Later Chen (2004) analyzed ammonia absorption from mixture of gases containing inert gas by water droplet as a conjugate mass transfer. Analysis of the influence of the internal circulation on the rate of mass transfer by falling droplets was further developed by Chen (2001b) for raindrops. Gas absorption by a liquid aerosol in a stationary environment was investigated by Chen (2002).

Accumulation of the dissolved atmospheric gases in a falling water droplet during absorption and circulation

of liquid inside a droplet caused by shear stress at the interface poses a conjugate problem of unsteady convective diffusion with time-dependent boundary conditions. Analytical solution of this problem requires application of more sophisticated methods in the theory of heat and mass transfer (see, e.g. Bartels and Churchill (1942) and Ruckenstein (1967)).

In all previous studies conjugate mass transfer during gas absorption by falling liquid droplets was investigated either analytically using quasistationary model (see, e.g. Alexandrova et al., 2004) or by solving numerically a system of governing partial differential equations (see Chen, 2004, Walcek and Pruppacher, 1984a). In the approach suggested in this study, we developed an alternative model for an unsteady mass transfer during binary gas absorption by falling droplets. The suggested approach involves application of a generalized similarity transformation to a system of nonstationary equations of convective diffusion and a Duhamel's theorem. The problem is reduced to a numerical solution of Volterra integral equation of the second kind. The obtained solution can be used for validating comprehensive simulation procedures for gas absorption by falling liquid droplet involving ventilation (see, e.g. Beard and Pruppacher, 1971, Pruppacher and Klett 1997, p. 540), dissociation (Jacobson, 2005) and aqueous chemistry with diffusion-limited mass transfer (Pandis and Seinfeld, 1989; Jacobson, 1997).

2. Description of the model

Consider absorption of a soluble gas from a mixture containing an inert gas by a moving droplet. At time $t=0$, a droplet begins to absorb gas from a host medium occupying a large volume as compared with the droplet's volume. Resistance to mass transfer in both phases is taken into account. The problem is solved in the approximation of an infinite dilution of absorbate in the absorbent. The thermodynamic parameters of a system are assumed constant. Convective diffusion is determined by fluid velocity at the droplet's surface. The thicknesses of diffusion boundary layers in both phases are assumed small compared with the droplet's size and tangential molecular mass transfer rate along at the surface of a spherical droplet is assumed small compared with a molecular mass and heat transfer rates in the normal direction. The bulk of a droplet, beyond the diffusion boundary layers, is completely mixed and concentration of solute is homogeneous in the bulk. The assumptions that there is circulation inside a droplet and that the droplet has a spherical shape are valid in the following range of the falling in air water droplet radii, Reynolds numbers and velocities: $0.1\,\mathrm{mm}\leqslant R\leqslant 0.5\,\mathrm{mm}$, $10\leqslant Re\leqslant 300$ and $0.7\,\mathrm{m\,s^{-1}}\leqslant U\leqslant 4.5\,\mathrm{m\,s^{-1}}$. Effects of turbulent diffusion and turbulent flow in the air must be taken into account for droplets with $R>0.8\,\mathrm{mm}$ (see Amokrane and Caussade (1999)), and they are not discussed in the present study. Analysis of fluid flow around a moving droplet (LeClair et al., 1972) showed that at different Reynolds numbers the tangential and radial fluid velocity components in the vicinity of a gas–liquid interface can be approximated by the following equations (see Pruppacher and Klett (1997, p. 392)):

$$v_\theta = -kU\sin\theta, \quad v_\mathrm{r} = \frac{2kU}{R}y\cos\theta, \tag{1}$$

where coefficient k is equal to 0.04 in the range of external flow Reynolds numbers ($Re=2UR\rho_\mathrm{G}/\mu_\mathrm{G}$) from 10 to 300 (see Pruppacher and Klett (1997, p. 386)). Eqs. (1) are valid for $y\ll R$. The dependence of the terminal fall velocity of liquid droplets on their diameter is available in Pruppacher and Klett (1997, pp. 415–421). Following the approach suggested by Ruckenstein (1967) and developed in our previous studies (see, e.g. Elperin and Fominykh (1999, 2003, 2004)) we arrive at the nonstationary equations of convective diffusion for the liquid and gaseous phases which accounts for convection in radial and tangential directions:

$$\frac{\partial X_i}{\partial\tau_i}+Pe_i\left\{-\sin\theta\frac{\partial X_i}{\partial\theta}+2Y\cos\theta\frac{\partial X_i}{\partial Y}\right\}=\frac{\partial^2 X_i}{\partial Y^2}, \quad i=1,2. \tag{2}$$

The initial and boundary conditions to Eqs. (2) read:

$$X_2=0 \quad \text{as } Y\to\infty, \tag{3}$$

$$X_1=X_\mathrm{b}(\tau_1) \quad \text{as } Y\to-\infty, \tag{4}$$

$$X_1=mX_2 \quad \text{at } Y=0, \tag{5}$$

$$N_1=N_2 \quad \text{at } Y=0, \tag{6}$$

where

$$N_i=-D_i\rho_i\frac{\partial x_i}{\partial y}, \quad X_\mathrm{b}(\tau_1)=\frac{x_\mathrm{b}(t)-mx_2(\infty)}{x_{10}-mx_2(\infty)},$$

$$y=r-R, \quad y\ll R$$

(see Fig. 1),

$$X_1=\frac{x_1-mx_2(\infty)}{x_{10}-mx_2(\infty)}, \quad X_2=\frac{x_2-x_2(\infty)}{x_{10}-mx_2(\infty)},$$

$$Pe_i=\frac{RkU}{D_i}, \quad Y=y/R, \quad \tau_i=\frac{tD_i}{R^2}$$

and coefficient m is a dimensionless Henry constant that characterizes the solubility of gases in liquids. The equations of convective diffusion (2) describe the change in concentration near the gas-droplet interface on both sides of the droplet. The form of these equations suggests that its solution may be obtained using of a similarity transformation (see Ruckenstein (1967)).

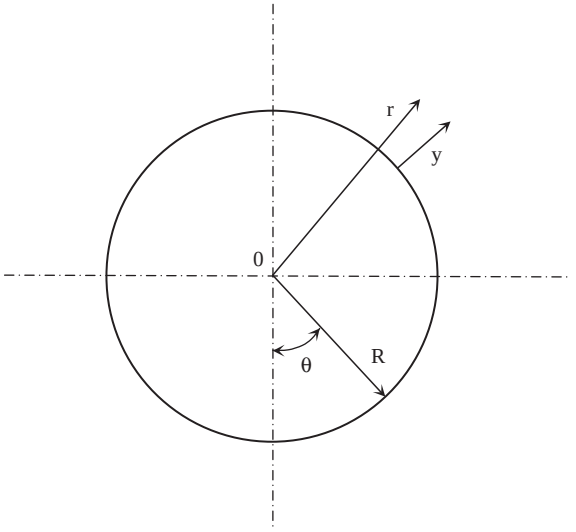

Fig. 1. Schematic view of a falling droplet.

3. Method of solution

Since the boundary condition (4) to Eq. (2) is a function of time, the solution can be found by combining the similarity transformation method with Duhamel's theorem. Eqs. (2) are solved by assuming first that the concentration of the absorbate in the bulk of liquid phase in Eq. (4) is constant. Introduce self-similar variables (see Ruckenstein et al. (1971)):

$$\eta_i = \frac{y}{\delta_i(t,\theta)} = \frac{Y}{\Delta_i}. \tag{7}$$

Variables η_i allow to transform a system of partial differential Eq. (2) into a system of ordinary differential equations:

$$\frac{\mathrm{d}^2 X_i}{\mathrm{d}\eta_i^2} + 2\eta_i \frac{\mathrm{d}X_i}{\mathrm{d}\eta_i} = 0, \tag{8}$$

where

$$\Delta_i^2 = \frac{4}{Pe_i \sin^4(\theta)} \left\{ \cos(\theta) - \frac{1}{3}\cos^3(\theta) - \left[\frac{1 - \mathrm{tg}^2(\theta/2)\exp(2T)}{1 + \mathrm{tg}^2(\theta/2)\exp(2T)} - \frac{1}{3}\left(\frac{1 - \mathrm{tg}^2(\theta/2)\exp(2T)}{1 + \mathrm{tg}^2(\theta/2)\exp(2T)} \right)^3 \right] \right\}, \tag{9}$$

$T = Pe_i\tau_i = tUk/R$ is a dimensionless time. Boundary conditions to Eqs. (8) read:

$$X_2 = 0 \quad \text{as } \eta_2 \to \infty, \tag{10}$$

$$X_1 = X_\mathrm{b}(\tau_1) \quad \text{as } \eta_1 \to -\infty, \tag{11}$$

$$X_1 = mX_2 \quad \text{at } \eta_1 = \eta_2 = 0, \tag{12}$$

$$N_1 = N_2 \quad \text{at } \eta_1 = \eta_2 = 0, \tag{13}$$

where $N_i = -D_i\rho_i(\partial x_i/\partial y)$. Solutions of Eqs. (8), $\bar{x}_i$, with a constant boundary condition at $y \to -\infty$ read:

$$\bar{x}_1 = A_1 \,\mathrm{erf}|\eta_1| + A_2, \tag{14}$$

$$\bar{x}_2 = A_3 \,\mathrm{erf}(\eta_2) + A_4, \tag{15}$$

where A_1, A_2, A_3, and A_4 are constants of integration and $\mathrm{erf}(z) = (2/\sqrt{\pi})\int_0^z \mathrm{e}^{-\tau^2}\,\mathrm{d}\tau$. Substituting Eqs. (14)–(15) into boundary conditions (10)–(13) and assuming that x_b is independent of time, allows to determine the constants of integration:

$$A_1 = \frac{x_\mathrm{b} - mx_2(\infty)}{1 + \gamma mD}, \quad A_2 = \frac{mx_2(\infty) + x_\mathrm{b}\gamma mD}{1 + \gamma mD},$$

$$A_3 = \frac{\gamma D[mx_2(\infty) - x_\mathrm{b}]}{1 + \gamma mD}, \quad A_4 = \frac{x_2(\infty) + x_\mathrm{b}\gamma D}{1 + \gamma mD}.$$

Applying Duhamel's theorem to Eqs. (14) and (15) we obtain a solution of Eqs. (2) with time-dependent boundary conditions at the infinity in the following form:

$$X_1(Y,\theta,T) = \frac{\partial}{\partial T}\int_0^T \left[1 - \frac{1}{1 + m\gamma D}\,\mathrm{erfc}\left(\frac{Y}{\Delta_1(\theta, T-\lambda)} \right) X_\mathrm{b}(\lambda) \right] \mathrm{d}\lambda \tag{16}$$

$$X_2(Y,\theta,T) = \frac{\partial}{\partial T}\int_0^T \left[\frac{\gamma D}{1 + m\gamma D}\,\mathrm{erfc}\left(\frac{Y}{\Delta_1(\theta, T-\lambda)} \right) X_\mathrm{b}(\lambda) \right] \mathrm{d}\lambda, \tag{17}$$

where $X_\mathrm{b}(\lambda) = (x_\mathrm{b}(\lambda) - mx_2(\infty)/x_{10} - mx_2(\infty))$ and $\mathrm{erfc}(x) = 1 - \mathrm{erf}(x)$. The variable x_b is an unknown function of time which can be determined by means of an integral material balance over the droplet (see Uribe-Ramirez and Korchinsky (2000)):

$$-V\rho_1 \frac{\mathrm{d}x_\mathrm{b}}{\mathrm{d}t} = 2\pi R^2 \int_0^\pi N_1|_{Y=0} \sin\theta\,\mathrm{d}\theta. \tag{18}$$

Substituting expression for the absorbate concentration in the dispersed phase (Eq. 16) into Eq. (18), we obtain after integration the expression for the dimensionless mass fraction of an absorbate in the bulk of a droplet as a function of a dimensionless time T, droplet Peclet number, diffusion and density ratio and distribution coefficient m:

$$X_\mathrm{b}(T) = 1 + \frac{3}{Pe_1\sqrt{\pi}(1 + m\gamma D)} \times \int_0^T X_\mathrm{b}(\lambda) \int_0^\pi \frac{\sin\theta}{\Delta_1(\theta, T-\lambda)}\,\mathrm{d}\lambda, \tag{19}$$

where $Pe_1 = (UkR/D_1)$. Eq. (19) is a Volterra integral equation of the second kind (see, e.g. Press et al., 1992).

Eq. (19) can be written in the following form:

$$f(\tau) = \int_0^\tau f(\zeta)K(\tau,\zeta)\,\mathrm{d}\zeta + g(\tau). \tag{20}$$

After discretization on a uniform mesh with an increment h and using trapezoidal integration rule, Eq. (20) can be reduced to a system of linear algebraic equations with a triangular matrix:

$$f(0) = g(0),$$
$$(1 - 0.5hK_{i,i})f_i = h\left(0.5K_{i,0} + \sum_{j=1}^{i-1} K_{ij}f_j\right) + g_i, \tag{21}$$

where $i = 1,\ldots,N$, $f_i = f(ih)$, $g_i = g(ih)$, $K_{ij} = K(ih,jh)$.

In order to solve the system of Eqs. (21), it is required to calculate $N^2/2$ values of the kernel K_{ij}. Since $K(\tau,\zeta) = K(\tau-\zeta)$ or $K_{ij} = K((i-j)h)$, then $K_{i+1,j} = K((i+1-j)h) = K((i-(j-1))h) = K_{i,j-1}$. Consequently, at each time step i, it is necessary to calculate only one value of the kernel K_{i0} because all the other values were already determined: $K_{i,1} = K_{i-1,0}$, $K_{i,2} = K_{i-1,1}$. Therefore when condition $K(\tau,\zeta) = K(\tau-\zeta)$ is valid it is necessary to calculate only N values of the kernel. Numerical calculations were performed with $N = 400$.

The mass fraction of absorbate in the bulk of dispersed phase, x_b is only a function of time. The theoretical bulk mass fractions of absorbate in the dispersed phase calculated from Eq. (19) with the function $\Delta_1(\theta,T)$ given by Eq. (9) was compared with the experimental data of Altwicker and Lindhjem (1988) for carbon dioxide absorption from the mixture with N_2 gas by falling droplets of water with a diameter $d = 1.2$ mm (see Fig. 2) and with the experimental data of Amokrane and Caussade (1999) for SO_2 absorption from the mixture with N_2 gas by falling droplets of water with a diameter $d = 0.6$ mm (see Fig. 3). In both experiments droplets exhibited steady internal circulation (see e.g. Walcek et al. (1984b)). The droplet velocity and the external Reynolds number for a falling drop in the experiments of Altwicker and Lindhjem (1988) was equal to $4\,\mathrm{m\,s^{-1}}$ and 340, and numerical calculations were performed for $Pe_1 = 5\times10^4$. In the experiments of Amokrane and Caussade (1999) droplet velocity and external flow Reynolds number were equal to $2.7\,\mathrm{m\,s^{-1}}$ and 115, and numerical calculations are performed for $Pe_1 = 2.8\times10^4$, $x_2(\infty) = 4.34\times10^{-3}$. As can be seen from Figs. 2 and 3, the theoretical predictions show fairly good agreement with the experimental results. Further validation and improvement of the suggested model can be carried out using new experimental results including simultaneous measurements of the rate of absorption and fluid velocity at the droplet interface.

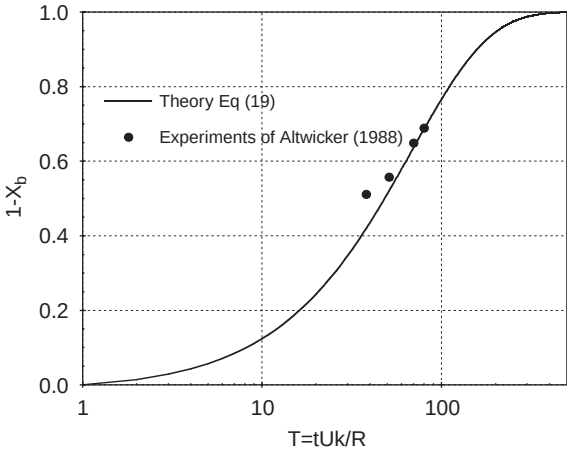

Fig. 2. Dependence of the concentration of the dissolved gas in the bulk of a water droplet $1 - X_b(T) = (x_b(t) - x_{10})/(mx_2(\infty) - x_{10})$ vs. time for absorption of CO_2 by water in the presence of inert admixture.

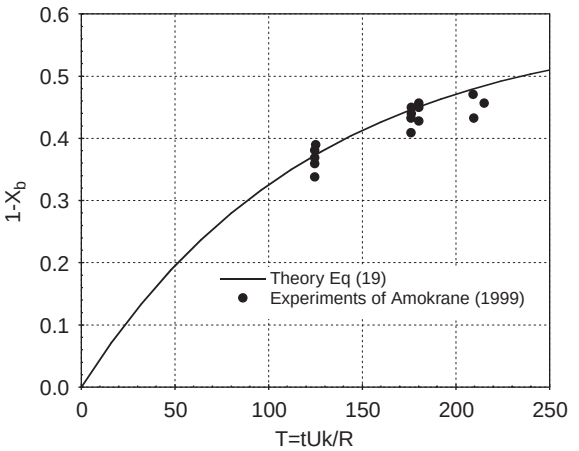

Fig. 3. Dependence of the concentration of the dissolved gas in the bulk of a water droplet $1 - X_b(T) = (x_b(t) - x_{10})/(mx_2(\infty) - x_{10})$ vs. time for absorption of SO_2 by water in the presence of inert admixture.

4. Summary and conclusions

We considered conjugate mass transfer during solvable gas absorption by a falling droplet with the internal circulation from a mixture containing an inert gas using approximation of a thin concentration boundary layer in the liquid and gaseous phases. Absorbate accumulation in the bulk of the liquid phase is taken into account. The bulk of a droplet, beyond the diffusion boundary layer is completely mixed and concentration of absorbate is homogeneous and time-dependent in the bulk.

The system of transient partial parabolic differential equations of convective diffusion in liquid and gaseous phases with time-dependent boundary conditions was solved by combining the similarity transformation method with Duhamel's theorem. It is shown that the dependence of the radius-averaged concentration vs. time in a falling droplet is determined by Volterra integral equation of the second kind which is easier to solve than the original system of partial differential equations. The obtained theoretical results were compared with the experimental data of Altwicker and Lindhjem (1988)) for carbon dioxide absorption from the mixture with inert gas and with the experimental data of Amokrane and Caussade (1999) for SO_2 absorption from the mixture with N_2 gas by falling droplets of water with internal circulation. Theoretical predictions and experimental data show fairly good agreement.

The derived solution can be used in calculations of scavenging of hazardous gases in atmosphere by rain and incorporated into the computer codes. The simple form of the obtained solutions allows to use them to analyze the dependence of the rate of mass transfer on different parameters, e.g., upon a radius of a droplet, diffusion coefficient, etc. The obtained results allow to construct a database that can be used for training artificial neural networks (ANN) for atmospheric pollutant level analysis (see Lu et al. (2002)). The obtained solution can be used for validating modeling procedures for solving more complicated problems of gas absorption by falling liquid droplets.

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