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SO_2 absorption and desorption by an accelerating water droplet undergoing vaporization

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Abstract

Heat and mass transfer between a water droplet and polluted air has been studied during the beginning of its free fall. Experimental results are compared with predicted ones from a model based on the Whitman film theory. The purpose of this article is to extend our previous model to predict the SO_2 absorption and desorption by drops (1–5 mm) falling in air containing low gas concentration and including the natural droplet vaporization and cooling. The measured droplet temperature and sulfur concentration after small contact time (0.35–2.43 s) concords with model predictions. It is observed that the initial vaporization and its corresponding cooling effect increase the initial SO_2 absorption rate and decrease the initial SO_2 desorption rate.

Keywords: Absorption; Heat transfer; Mass transfer; Droplet; Vaporization; Cooling

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1. Introduction

Some knowledge of the simultaneous evaporation and gas absorption is necessary in any application where gas–liquid absorption occurs. Drying, purification of toxic atmospheres or water curtains are typical examples in which the transfer between a continuous phase (the gas) and a dispersed phase (the droplets) occurs. Many experimental investigations have been carried out in the area of air-pollution control. Studies, both computational and empirical, of gas absorption by droplets have been performed, and reviews of the current understanding are available in literature (Beard and Pruppacher, 1971; Chang and Davis, 1974; Pruppacher and Rasmussen, 1979; Saboni, 1991; Amokrane et al., 1994; Widmann and Davis, 1997; Sirignano, 1999; Chen, 2001b). They put in advance that transport of trace gases from polluted air into a droplet is controlled by molecular diffusion and by convection outside and inside the droplet. Moreover, in the case of falling droplets, the aerodynamic drag on the surface creates internal circulation and makes droplets larger than 0.5 mm oscillate. That facilitates the diffusion of the absorbed gas (Altwicker and Lindhjem, 1988; Saboni, 1991; Chen, 2001b). For droplets falling in a highly soluble gas medium, the main transfer resistance is located in the gas phase, but according to Saboni and Alexandrova (2001); and Alexandrova et al. (2004), the global resistance is the sum of the resistance in the liquid with the resistance in the gas. At high concentration (few %) the resistance in the liquid dominates whereas at low concentration (few ppb), the resistance in the gas phase dominates. In the present work, experiments are conducted at low and intermediate gas concentration where the mass transfer resistance is located in the gas and in the liquid (few ppm).

Models have been proposed and a survey of the published studies (Altwicker and Lindhjem, 1988; Saboni and Alexandrova, 2001) shows that a lot of good numerical models, as well as experimental correlations for prediction of the mass transfer coefficient in the gas film are available.

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Nomenclature

C_D	drag coefficient
C_p	heat capacity, J kg^{-1} K^{-1}
d	droplet diameter, m
D	molecular diffusivity, m^2 s^{-1}
g	acceleration due to gravity, m s^{-2}
h	convective heat transfer, W m^{-2} K^{-1}
Hr	humidity, %
k	mass transfer coefficient, m s^{-1}
k_H	SO_2 liquid/gas equilibrium constant $k_H = 10^{(\frac{1376.1}{T}-6.521)}8.32T$ (mol/mol)
k_{E1}	$H_2SO_3 \leftrightarrow HSO_3^- + H^+$ equilibrium constant $k_{E_1} = 10^{(\frac{853}{T}-4.74)}$ (mol/l)
Le	Lewis number
L_v	enthalpy of vaporization, J kg^{-1}
$\dot{m}$	vaporized mass flow, kg s^{-1}
Nu	Nusselt number
Pr	Prandtl number
$\dot{Q}_{amb}$	heat power, W
Re	Reynolds number
Re^*	Reynolds at the interface
S_l	droplet surface, m^2
Sc	Schmidt number
Sh	Sherwood number
t	time, s
T	temperature, K
u	velocity, m s^{-1}
U_∞	terminal velocity, m s^{-1}
u^*	liquid interfacial friction velocity, m s^{-1}
V_l	volume of droplet, m^3
Y	mass fraction
ρ	density, kg m^{-3}
ω	constant
λ	thermal conductivity, W m^{-1} K^{-1}
ν	kinematic viscosity, m^2 s^{-1}
τ	time scale, s
ΔH	enthalpy of reaction of absorption, kJ kg^{-1}

Subscripts

abs.	absorption
acc.	acceleration
cool.	cooling
l	liquid phase
g	gas phase
s	droplet surface
sat.	at saturation
vap.	vaporization
∞	ambient air
SO_2	sulfur dioxide
w	water

To predict the transfer in the liquid, Chen (2001a,b, 2004) investigated numerically the transport of absorbed gas (SO_2 or NH_3) in the case of a liquid droplet in convective flow. He suggests, that the transport mechanisms in the droplet is a consequence of radial diffusion and internal circulation by the way of internal vortexes. Sulfur dioxide absorption is mainly governed by internal circulation when convective flow is relatively fast.

Saboni (1991) proposed a quasi steady state model based on local scales, interfacial liquid friction velocity and droplet diameter including the resistance to mass transfer in the liquid phase. The model was validated experimentally by Amokrane et al. (1994), in the case of sulfur dioxide absorption by water drops falling through air containing high gas concentration (few %).

A considerable number of works made it possible to propose a set for correlation on heat and mass transfer in the case of stagnant droplets, droplets at terminal velocity or droplet at intermediate and constant velocity. Only few experimental studies have been undertaken at the beginning of the gas–liquid contact (Erens et al., 1994), when the droplet temperature converges to the wet bulb temperature, however this corresponds to common applied or experimental situations when droplets are generated in columns or towers containing mixture of dry air and gas. In Smolík et al. (2001) the influence of evaporation on the effectiveness of the transfer of matter has been the subject of investigation. The author presents an overview of existing work and proposed a correlation valid for intermediate Reynolds number when droplets are submitted to pure evaporation, without gas absorption. Widmann and Davis (1997) were interested in the case of droplets crossing air polluted with SO_2 and calculated the time variation of the concentration in air and in the droplet. The authors investigated very small droplets (10^{-5} m) and considered that the sequence of drop can be approximated as a line or rod source with respect to the gas phase, that is not directly usable in the present case. Effect of water vaporization or condensation on SO_2 absorption was also investigated with a mathematical model by Huckaby and Ray (1989) in the case of stagnant droplet. The authors found that droplet evaporation enhances gas absorption at short contact time.

In order to investigate the effect of droplet vaporization onto the absorption process, Saboni's model was extended to the case of a uniform but time varying droplet temperature. The extended model is presented in Section 2. To validate it, experiments were carried out onto the cooling of free falling droplets with gas absorption or desorption. The experimental device is presented in details in Section 3. The time variation of the droplet temperature was measured through a vertical tunnel of 2.3 m containing air not saturated with water vapor. The absorption of SO_2 by large droplets was measured during their fall with the same device for various gas concentrations and for various droplets diameters. The natural reversible desorption was also measured from initially polluted droplets, during their fall in unsaturated pure air. Results at intermediate contact time are also reported. Comparisons between experimental and predicted results and discussion are presented in Section 4.

2. Theory

The droplet is assumed to be a sphere at uniform temperature falling down with zero initial velocity in a stagnant gaseous mixture of air, water vapor and SO_2. The droplet undergoes heat transfer, vaporization, SO_2 absorption or desorption and acceleration.

Three systems are considered: the droplet, the surface and the surrounding air. Transport in the interior of the droplet is significantly affected by the drop internal circulation produced by the gas shear at the droplet surface (Chen, 2001a; Chen and Lu, 2003; Sirignano, 1999). Then, exchanges between the three systems are introduced by considering convection through gas film and liquid film formed on each sides of the surface. The droplet temperature and droplet SO_2 concentration are considered uniform but time variable. This is also the case for the surface where reactions of absorption and evaporation occur. Temperature, humidity and SO_2 concentration in air and outside the gas film are constant.

2.1. Heat and mass transfer

2.1.1. Transfer between the surface and surrounding air

The heat flux arriving to the surface by air convection is

$$\dot{Q}_{\mathrm{amb}} = S_1 h_{\mathrm{g}}(T_\infty - T_{\mathrm{s}}). \tag{1}$$

The water vapor mass flow crossing the gas film at the surface is

$$\dot{m}_{\mathrm{w}} = S_1 k_{\mathrm{gw}} \rho_{\mathrm{g}} (Y_{\mathrm{sg,w}} - Y_{\infty,\mathrm{w}}). \tag{2}$$

The mass flow of SO_2 crossing the gas film at the surface is

$$\dot{m}_{\mathrm{SO_2}} = S_1 k_{\mathrm{gSO_2}} \rho_{\mathrm{g}} (Y_{\mathrm{sg,SO_2}} - Y_{\infty,\mathrm{SO_2}}). \tag{3}$$

2.1.2. Transfer between the droplet and its surface

The time variation of the dissolved SO_2 concentration in the droplet is calculated from migration of SO_2 in the liquid film:

$$V_1 \rho_1 \frac{\mathrm{d}Y_{1,\mathrm{SO_2}}}{\mathrm{d}t} = S_1 k_1 \rho_1 (Y_{1,\mathrm{SO_2}} - Y_{\mathrm{sl,SO_2}}). \tag{4}$$

The time variation of the droplet temperature is calculated from the heat flux between the droplet and the surface:

$$V_1 \rho_1 C_p \frac{\mathrm{d}T_1}{\mathrm{d}t} = S_1 h_1 (T_1 - T_{\mathrm{s}}). \tag{5}$$

2.1.3. Transfer at the surface

The vaporized water mass flow is calculated from heat balance at the surface:

$$L_{\mathrm{v}} \dot{m}_{\mathrm{w}} = \dot{Q}_{\mathrm{amb}} + S_1 h_1 (T_1 - T_{\mathrm{s}}) + \dot{m}_{\mathrm{SO_2}} \Delta H. \tag{6}$$

It is known that the temperature of a moving droplet is about the temperature of the surface (Sirignano, 1999). That indicates that introducing a surface temperature different from droplet temperature has a marginal effect on predicted results (absorption of SO_2, water vaporization, time variation of the droplet temperature). Then, Eqs. (4) and (6) can be simplified into

$$V_1 \rho_1 C_p \frac{\mathrm{d}T_1}{\mathrm{d}t} = L_{\mathrm{v}} \dot{m}_{\mathrm{w}} - \dot{Q}_{\mathrm{amb}} - \dot{m}_{\mathrm{SO_2}} \Delta H. \tag{7}$$

Absorbed SO_2 concentration in the liquid at the interface is calculated by assuming gas–liquid equilibrium at the interface (Chen, 2001b). In this case, SO_2 is immediately transformed into H_2SO_3, HSO_3^- and SO_3^{2-} (Huckaby and Ray, 1989; Amokrane et al., 1994; Saboni and Alexandrova, 2001; Alexandrova et al., 2004).

Therefore, in the liquid phase, concentration of SO_2 corresponds to the sum of concentration of species H_2SO_3, HSO_3^- and SO_3^{2-}. The low concentration encountered in the present case limits the second dissociation, and the total concentration is correctly approximated by the sum of concentration of H_2SO_3 and HSO_3^-. This sum is expressed in equivalent mass of SO_2.

Equation describing SO_2 equilibrium at the interface is in this case:

$$\rho_1 Y_{\mathrm{sl,SO_2}} = k_{\mathrm{H}} \rho_{\mathrm{g}} Y_{\mathrm{sg,SO_2}} + \sqrt{k_{\mathrm{H}} k_{\mathrm{E1}} \rho_{\mathrm{g}} Y_{\mathrm{sg,SO_2}}}. \tag{8}$$

The corresponding equilibrium constant k_{H} and k_{E1} are introduced as function of temperature at the surface.

In Eqs. (1)–(3), heat and mass transfer coefficient in the gas phase are

$$k_{\mathrm{g}} = \frac{Sh_{\mathrm{g}} D_{\mathrm{g}}}{d} \tag{9}$$

and

$$h_{\mathrm{g}} = \frac{Nu_{\mathrm{g}} \lambda_{\mathrm{g}}}{d}. \tag{10}$$

The Sherwood number in the gas phase is calculated from the Reynolds and Schmidt numbers in the gas phase with the expression proposed by Kulmala et al. (1995).

$$Sh_{\mathrm{g}} = 2.00 + 0.514 Re_{\mathrm{g}}^{0.5} Sc_{\mathrm{g}}^{0.33}. \tag{11}$$

The Nusselt number in the gas phase is calculated by using the analogy between heat and mass transfer:

$$Nu_{\mathrm{g}} = 2.00 + 0.514 Re_{\mathrm{g}}^{0.5} Pr_{\mathrm{g}}^{0.33}. \tag{12}$$

With regard to the transfer in the liquid phase, preceding works of Saboni (1991); Amokrane et al. (1994); Saboni and Alexandrova (2001) and Marion et al. (2003) give a correlation implying the coefficient of mass transfer k_1 of absorbed SO_2 in a moving droplet through the number of Sherwood:

$$Sh_l = \frac{k_l d}{D_L} = \omega Re_l^{*1/2} Sc_l^{1/2}. \tag{13}$$

In this equation, ω is a constant determined experimentally and D_l is the coefficient of diffusion of SO_2 in water. The authors found $\omega = 0.8$. Re_l^* is the interfacial Reynolds number and u^* is the interfacial friction velocity.

$$Re_l^* = \frac{u^* d}{\upsilon_l}, \tag{14}$$

$$u^* = U_\infty \sqrt{\frac{\rho_g}{\rho_l} \frac{C_D}{2}}. \tag{15}$$

The global drag coefficient C_D of a droplet in free fall has been proposed by Berry and Pranger (1974) by the following expression:

$$\begin{aligned}\ln(Re_g) = {} & -3.126 + 1.013 \ln(C_D Re_g^2) \\ & - 0.01912 \left(\ln(C_D Re_g^2)\right)^2.\end{aligned} \tag{16}$$

This relation is valid for a Reynolds number ranging between 1 and 3550 and for $C_D Re_g^2$ ranging between 2.4 and 10^7.

2.2. Equation of motion

The droplet is subjected to its weight, to the Archimede force and to the drag force. Then, the acceleration of the droplet is

$$\frac{du}{dt} = \frac{(\rho_l - \rho_g)}{\rho_l} g - \frac{3}{4d} \frac{\rho_g C_D}{\rho_l} u^2. \tag{17}$$

The droplet temperature history, droplet concentration and droplet velocity are calculated by integrating Eqs. (4), (7) and (17), respectively.

Chiang et al. (1992) investigates the case of a cold droplet injected into a hot gas stream with a numerical analysis. The authors observed that the effect of variable properties can not be neglected for the large temperature difference from the droplet surface to the far stream environment. In the present work, physical and chemical constants are introduced as functions of the liquid or the gas temperature, and the main influence of the temperature is limited to the chemical equilibrium at the surface. We used properties of pure water and pure air and coefficients of diffusion of SO_2 in water or in air.

3. Experiments

The experimental device is presented in Fig. 1. It is made of a Plexiglas column 2.3 m height and 104 mm internal diameter. The air in the column is set at the desired temperature and humidity. The rate of flow is sufficiently low to be neglected (0.14 m/s) but it is strong enough to homogenize the SO_2 concentration anywhere in the column. Air with desired SO_2 concentration up to 2000 ppm is introduced into the column. A chemical cell analyzer measures the gas concentration in the column. The temperature and humidity are measured at the top, in the middle and at the bottom of the column. The droplets are generated thanks to a specific device made of a glass tube connected to a capillary (0.30 mm in diameter). Their initial temperature can be different from the air temperature by using a heater before injection. For all the experiments, we used demineralized water. The drops were identical and fell with zero initial velocity. Their individual diameters were measured just at the beginning of the free fall by using a video camera focused onto the bottom of the injector. We used a color camera JVC TK-C7101EG and a Cosmicar objective of 75 mm preceded by a lengthening-piece of 4 cm. A strong back lighting made the obturator of the camera operate at very short exposure time, which gives a clear image of the drop in spite of its displacement. The real diameter was obtained by using the image of a calibration grid registered before the experiment. The image analyses of droplets were carried out with LEICA IM50 software. To measure the instantaneous droplet temperature, droplets were collected into a calorimeter. This collector is equipped with a 0.2 mm thermocouple. A Keithley 7700 card recorded the corresponding signal. The collector initially contained kerosene in order to form a film on the collected water. The presence of this organic film avoided the additional water evaporation and additional absorption of SO_2. During an experiment, a small pipe evacuated a fraction of the collected water to reduce the thermal response time of the unit. Then the collector was quickly at the temperature of the impacting droplet. For temperature measurement only, it can be placed at various heights. This gave access to the temperature profile of the droplet during the initial phase of its free fall.

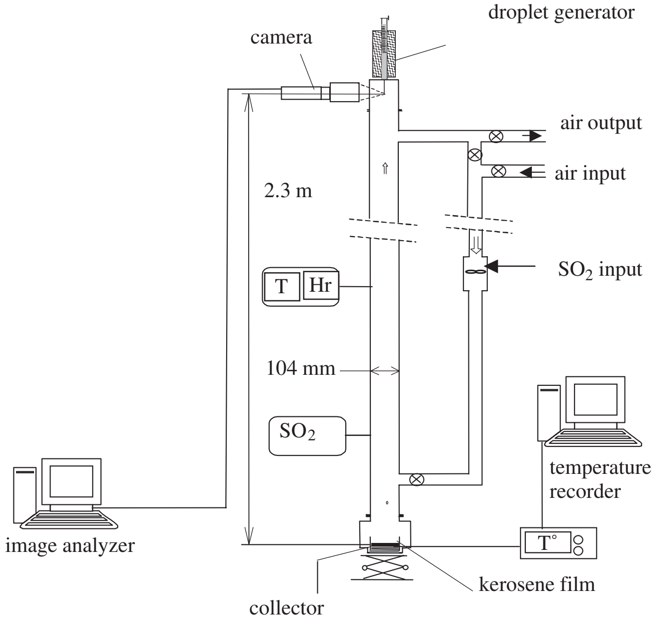

Fig. 1. Experimental device.

Table 1
Experimental conditions

	Absorption		
Panel A			
Drop size (mm)	2.04	3.09	4.31
Height (m)	2.3	2.3	2.3
Contact time (s)	0.76	0.73	0.71

	Desorption									
Panel B										
Drop size (mm)	4.31	4.31	4.31	4.31	4.31	4.57	4.57	4.57	4.57	4.57
Height (m)	2.3	3.98	8.03	12.93	16.3	2.3	3.98	8.03	12.93	16.3
Contact time (s)	0.72	0.97	1.49	2.06	2.45	0.71	0.97	1.48	2.05	2.43

	Cooling							
Panel C								
Drop size (mm)	2.04	2.04	2.04	2.04	1.80	1.80	1.80	1.80
Height (m)	0.55	1.35	1.85	2.35	0.51	0.91	1.31	1.71

The collected artificial sulfur acid rain was oxidized by hydrogen peroxide (H_2O_2) to form sulfuric acid (H_2SO_4). This operation avoided the natural reversible desorption of SO_2 when acid rain was separated from kerosene for analysis.

For reversible desorption, experiments were performed in the laboratory atmosphere. Droplets were generated using water containing initially dissolved sulfur dioxide. In this case, the 4.3–4.57 mm (in) diameter droplets free fall occurred over 16.3 m. Three intermediate levels allowed to operate with falling times varying from 0.7 to 2.4 s. The ambient temperature was measured in the surrounding of both the injector and the collector. For a run, the maximum temperature variation measured between the top and the bottom of the tunnel was 2 °C. The initial concentrations of dissolved SO_2 ranged from 0.5×10^{-3} to 1.9×10^{-3} mol/l.

For all the experiments (absorption or desorption), the concentration of sulfuric acid was determined by measurement of turbidity, using a spectrophotometer Elios operating at 605 nm. The calibration curve of the spectrophotometer was established beforehand using standard solutions of sulfuric acid.

Experimental conditions are summarized in Table 1. Contact time was calculated from integration of the equation of motion and considered the instantaneous droplet diameter and the falling distance.

4. Results and discussion

4.1. Validation of the present model

The measured time variation of the droplet temperature is presented in Fig. 2. Experiment 1 is obtained from water droplets of 2.0 mm, initially at 20 °C in free fall in air at 20 °C and 9% Hr. Experiment 2 is obtained from water droplets of 1.8 mm, initially heated-up to 36 °C and falling in air at 20 °C and 5% Hr. The initial phase of cooling predicted with the present model is in accordance with our experimental results. The experimental results of Yao (1974) and the predicted droplet temperature with our present model are also presented in Fig. 3. The good agreement of experimental and simulated data could indicate that the present model correctly predicts the effect of heat and mass transfer for water.

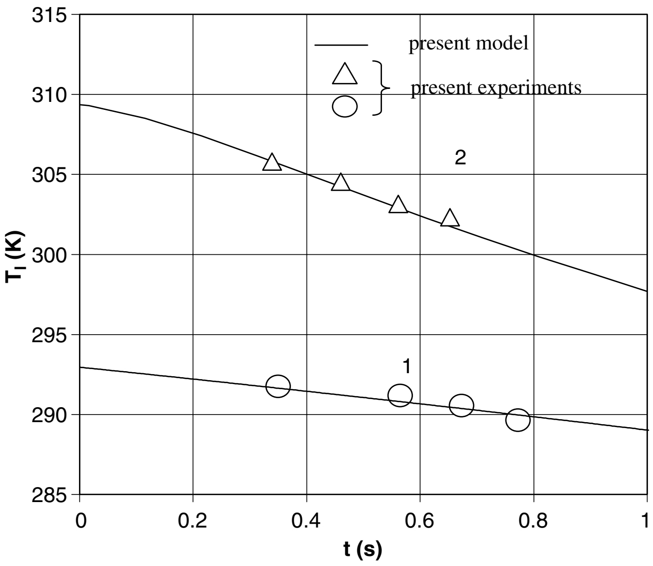

Fig. 2. Measured temperature of water droplets in free fall in air at 20 °C and for (1) 2 mm droplets at 20 °C in air at 9% Hr, and (2) 1.8 mm droplets at 36 °C in air at 5% Hr.

In the case of absorption with cooling effect, the measured concentration of total sulfur dioxide absorbed is plotted versus the gas concentration in the column in Fig. 4. Results are obtained for the maximum free fall available (2.3 m) in polluted air at 18.6 °C and 38.5% Hr for 2.04, 3.09 and 4.31 mm initial droplet diameters. Droplets do not reach terminal velocity, however, Kaji et al. (1985) observed that there is already internal agitation during the formation of a drop. The latter remains present

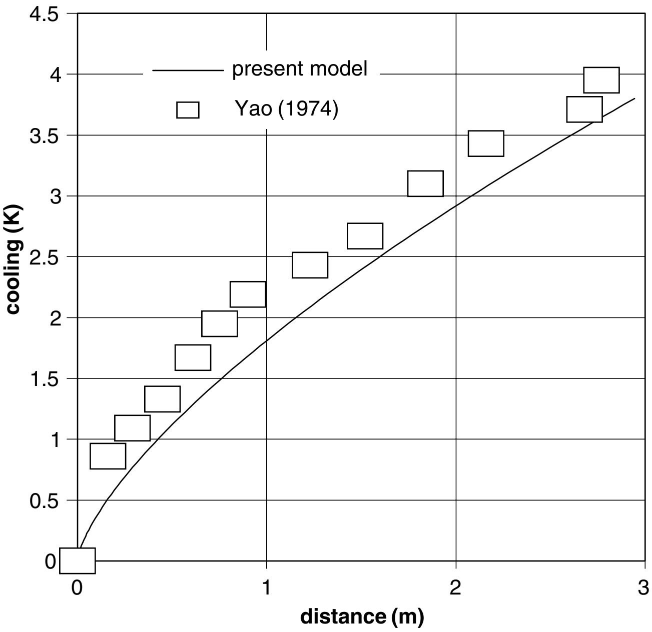

Fig. 3. Cooling of a 4 mm water drop mm in free fall initial temperature = 40.74 °C, air at 23.44 °C and 36% Hr.

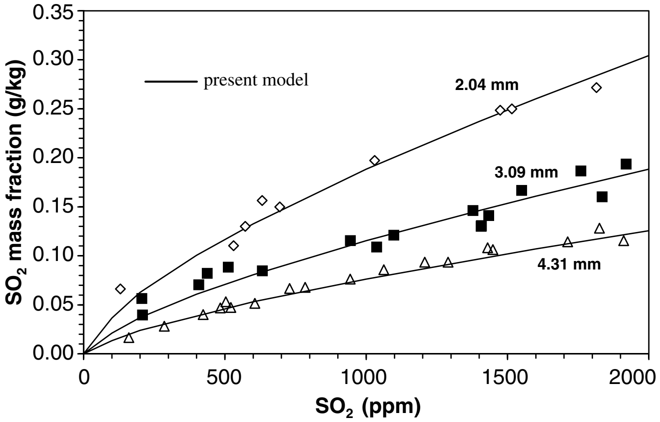

Fig. 4. Equivalent SO_2 mass fraction measured into 2.04, 3.09 and 4.31 mm water droplets after 2.3 m free fall in air at 18.6 °C and 38.5% Hr.

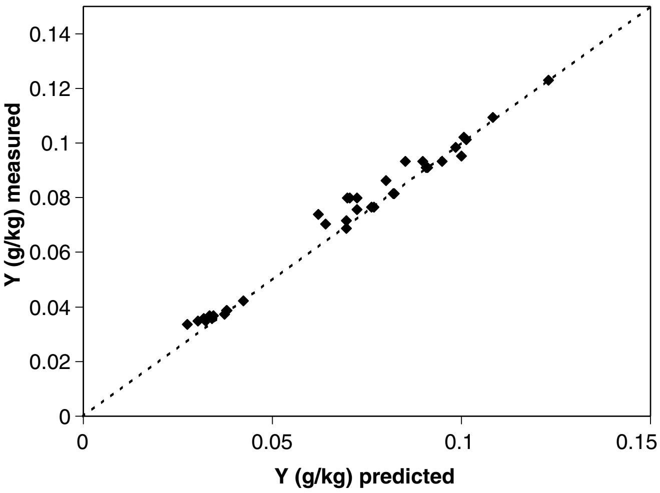

Fig. 5. Experimental and predicted equivalent SO_2 mass fraction in droplets falling in air and submitted to natural vaporization.

during all the fall because of the aerodynamic efforts exerted on the surface. Consequently, initial turbulence makes initial transfer resistance in the liquid being small even at the beginning of the fall when droplet velocity is small. This result is introduced in the present work by using a small internal resistance (calculated at terminal velocity) during the total fall of the droplet as in Amokrane et al. (1994).

The measured SO_2 mass fraction in the collector is plotted against the predicted value in Fig. 5. We notice that the present model fits the experimental results correctly with $\omega = 1.1$. Then, without including the time variation of the droplet temperature, good adequacy between experimental and predicted results would be obtained with $\omega = 1.2$. The mass transfer between the gas phase and the moving water droplets depends not only on hydrodynamics, but also on the physical and chemical properties of the diffusing gas. That is to say, it depends on the instantaneous local temperature. And this is because of the strong influence of the temperature on the chemical equilibrium. One can notice that for 2.04 mm droplets, the droplet temperature varies from 291.5 to 289.5 K during the fall and the equilibrium concentration at the interface increases up to 6% when concentration in the air is 1000 ppm. To further investigate the effect of vaporization and cooling, the model was used to calculate the SO_2 absorption rate into a 1 mm water droplet falling in dry air (20% Hr) during the first second of the fall. The predicted result is shown in Fig. 6 and a comparison is established with the predicted result in wet air (100% Hr). The gas concentration is 1000 ppm SO_2. It can be seen that the absorption rate calculated including the time variation of the droplet temperature is bigger than the absorption rate at constant temperature. The vaporization of the droplet causes its natural cooling and increases the value of the equilibrium constant at the interface and therefore, the concentration in the liquid at the interface.

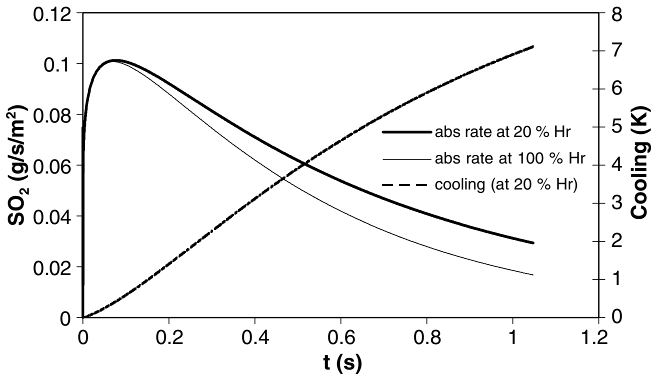

Fig. 6. Absorption rates and cooling for 1 mm water droplets falling in the air at $T_{air} = 293$ K containing 1000 ppm SO_2, Hr = 20% and 100%, and Tinitiale = 293 K (20 °C).

The model proposed by Saboni (1991) and validated by Amokrane et al. (1994) includes a constant determined experimentally at $\omega = 0.8$ emerging from a comparison with other literature models and with the experimental data of Walcek et al. (1984); Kaji et al. (1985); Altwicker and Lindhjem (1988) and Garner and Lane (1959). We notice that this corresponds to long residence time of the droplet (Walcek et al., 1984), very short residence time (Kaji et al., 1985), large droplets (Garner and Lane, 1959) and high gas concentration and large droplets experiments (Amokrane et al., 1994). Then, for all the experiments, the droplet temperature is roughly constant and its time variation is not significantly present during the absorption process. Therefore, the droplet is considered at the initial temperature or at the wet bulb temperature. In the present work, the time constant characterizing the variation of the droplet temperature is found to be around the time constant characterizing the SO_2 absorption and $\omega = 1.1$ is found to be the best way to fit the experimental results.

When the mass transfer goes from liquid to air (desorption), the concentration in the liquid decreases. The same basic theory which describes absorption should also describe desorption. The only factor which changes in the theory are the external and initial concentrations. Fig. 7 shows the total sulfur dioxide concentration versus elapsed time measured for 4.57 mm droplets falling in pure air at 21 °C and 40% Hr and when water droplets initially contain SO_2. Data at 0.7 s are obtained in the column; other results are obtained outside the column, in laboratory atmosphere. We notice that due to zero initial velocity, droplets accelerate over the 16.3 m of fall and they are about to terminal velocity when they hit the collector. At 2.4 s, (maximum contact time available) the total sulfur concentration measured is about 20% lower than the initial concentration. The measured and predicted time variation of the desorption rate are reported in Fig. 8. Once again, predicted results, with time variation of the droplet temperature correctly follow the experimental ones when $\omega = 1.1$. To investigate the effect of droplet vaporization, the predicted desorption rate without vaporization is shown in the same figure. We notice that the desorption rate in the air at 40% is lower than the desorption rate in the air at 100% (or constant droplet temperature). For example at $t = 2$ s, the difference is 6% and reaches 7% at $t = 3$ s, the corresponding cooling is 2.7 K at $t = 2$ s and 3.9 K at $t = 3$ s.

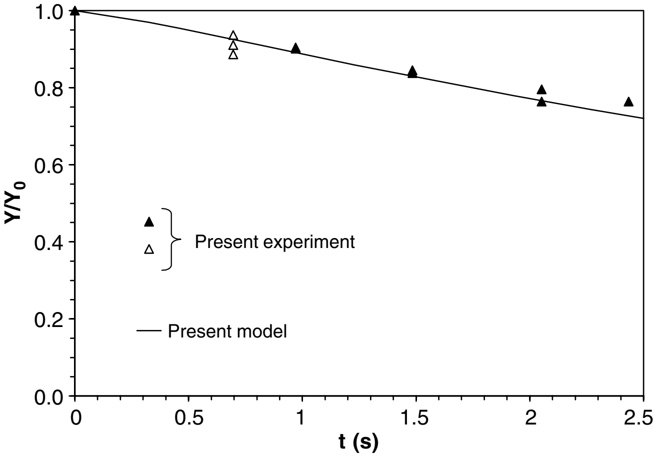

Fig. 7. Experimental and predicted time variation of the total equivalent SO_2 mass fraction inside a 4.57 mm in diameter drop falling in air at 21 °C and 40% Hr. Initial mass fraction of SO_2 is $Y_0 = 0.091$ g/kg.

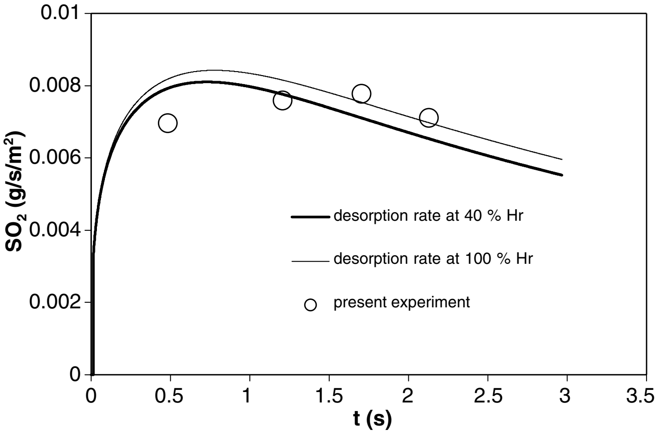

Fig. 8. Predicted and measured desorption rates of a 4.57 mm droplet containing initially 0.091 g/kg of equivalent SO_2, and falling in air at 294 K and Hr = 40%. Predicted value at 100% Hr is also present.

5. Conclusion

In the present approach, heat and mass transfer are considered between an evaporating and accelerating water droplet and stagnant polluted dry air. The governing equations describing the coupled heat and mass transfer are solved to find the cooling, the vaporization and the absorption (or the desorption) of individual droplets. In the liquid phase, a model based on local scales, interfacial liquid friction velocity and drop size diameter is used. In the continuous gas phase a more classical model is applied. The effect of vaporization was enhanced at intermediate concentration where resistance to mass transfer is located in the liquid and in the gas phase. To support the present model, three types of experiments were carried out. The first one was adapted to measure the natural cooling of a droplet in free fall in dry air. The second one was adapted to measure the absorption of gas by droplets of known diameter falling in dry air. The last one gave the desorption rate from droplets with an initial concentration of sulfur dioxide falling through SO_2-free air. We found that our measured droplet cooling and absorbed/desorbed gas concentration after small contact times agrees with our quasi steady state model predictions. We note that the cooling time of the droplet was about the same as the time for SO_2 absorption and desorption. The natural initial vaporization and its cooling effect increased slightly (about 6% in our experiments) the initial absorption rate and decreased slightly the initial desorption rate. This effect is explained by the strong influence of the temperature onto the equilibrium concentrations at the interface.

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