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Bešenić, Tibor; Vujanović, Milan; Baleta, Jakov; Pachler, Klaus; Samec, Niko; Hriberšek, Matjaž

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Research Article

Tibor Bešenić*, Milan Vujanović, Jakov Baleta, Klaus Pachler, Niko Samec, and
Matjaž Hriberšek

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Abstract: Mass transfer between the phases is a cornerstone of many technological processes and presents a topic whose understanding and modelling is of high importance. For instance, absorption of gases in liquid droplets is an underlying phenomenon for the desulfurization of flue gases in wet scrubbers. Wet scrubbing is an efficient cleaning method where the liquid is sprayed in a stream of rising gases, removing pollutants due to the concentration difference between the gas phase and droplets. A model for absorption in water droplets has been developed to describe the complex physical and chemical interactions during the exposure to flue gases. The main factors affecting the absorption are the mass transfer of pollutants through the gas–droplet interface and the aqueous phase chemistry in a droplet. The mass transfer coefficient, which has been modeled with several approaches, is the most significant parameter regulating the absorption dynamic into the droplet, while the in-droplet chemistry controls the maximum quantity of dissolved pollutants. Dissociation of sulfur dioxide and the chemical reactions in seawater have been described by the equilibrium reactions. Afterward, the influence of the mass transfer coefficient has been investigated, and the model has been validated against the literature data on a single droplet scale. Obtained results are comparable with the experimental measurements and indicate the applicability of the model for the design and development of industrial scrubbers.

Keywords: gas absorption, mass transfer coefficient, flue gas desulfurization, penetration theory, film theory

1 Introduction

A deeper understanding of the underlying physical and chemical phenomena is essential for designing better, more efficient, and sustainable technology and equipment. Improving the processes can help in achieving the crucial goals of modern society, such as mitigating the harmful impact to the environment and paving a way to a sustainable future. For example, the presence of pollutants in flue gases is an open issue, and sulfur oxides are among the most harmful gaseous pollutants. They have a detrimental impact on the environment, leading to respiratory and health effects for humans, causing excessive plant exfoliation, acidifying the natural waters, enhancing the corrosion in the exposed metals, and being precursors to the acid rains. European Union set the goals for SO_2 emission reductions for each country, while the MARPOOL limited the allowed sulfur levels in the fuel used for ships and these restrictions will get stricter and include more territories [1]. sulfur dioxides form during the combustion from fuel sulfur and exit the combustion chamber with flue gases. Primary measures for reducing their emissions include the low sulfur fuel, its removal before combustion, or influencing the parameters in the combustion chamber to prevent its formation. However, the more prevalent solution is exhaust gas after treatment. Removal of sulfur dioxide, as the dominating among the sulfur species, is usually done by the wet scrubbing process, where the polluted gas is placed in contact with the scrubbing liquid. Most common wet scrubber designs include packed beds, plate scrubbers, venturi scrubbers, and spray towers. Spray towers are

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*** Corresponding author: Tibor Bešenić,** The Department of Energy, Power and Environmental Engineering, Faculty of Mechanical Engineering and Naval Architecture, University of Zagreb, Zagreb, Croatia, e-mail: tibor.besenic@fsb.hr
Milan Vujanović: The Department of Energy, Power and Environmental Engineering, Faculty of Mechanical Engineering and Naval Architecture, University of Zagreb, Zagreb, Croatia, e-mail: milan.vujanovic@fsb.hr
Jakov Baleta: Department of Mechanical Metallurgy, Faculty of Metallurgy, University of Zagreb, Sisak, Croatia, e-mail: baleta@simet.hr
Klaus Pachler: AVL List GmbH, Graz, Austria, e-mail: klaus.pachler@avl.com
Niko Samec, Matjaž Hriberšek: Chair for Power, Process and Environmental Engineering, Faculty of Mechanical Engineering University of Maribor, Maribor, Slovenia, e-mail: matjaz.hribersek@um.si, niko.samec@um.si

several meters high vessels, where the liquid is sprayed in the gas stream from an array of nozzles, to increase the gas–liquid interfacial surface area. The droplets absorb SO_2 from flue gases and are collected at the bottom and treated. Commonly used liquids include solutions of the alkali in the water, such as limestone slurry, magnesium hydroxide, hydrated lime or caustic soda, although even seawater and pure water have the absorption capabilities [2]. Depending on the design and the liquid medium used, the SO_2 removal efficiency in spray towers can reach over 90% [3]. Seawater is convenient as a scrubbing agent in power plants near the sea [4], as well as in the marine applications on ships powered by sulfur-containing heavy oil [5].

Previously, researchers had tackled problems pertinent to the spray scrubber applications, both on the experimental and modelling sides. The topic of sulfur dioxide absorption in liquid droplets has been investigated for some years now, from the atmospherical point of view and the simplified chemical models in the beginning [6], to the numerical modelling in the recent years [7]. Maurer [8] investigated the solubility, and the equilibrium concentrations of sulfur dioxide in water, Walcek and Pruppacher [6] studied the atmospherical absorption of SO_2 by clouds and raindrops, while Walcek et al. [9] performed experiments on the freely falling droplets in rain shaft. Among others, Saboni and Alexandrova [10] developed a mathematical model for absorption of SO_2 in pure water droplets, while Abdulsattar et al. [11] modelled chemistry of a sulfur dioxide–seawater systems. More recently, Andreasen and Mayer [12] compared model predictions of the equilibrium SO_2 concentrations in pure and seawater, while Marocco [13] implemented a model for limestone slurry spray tower in the computational fluid dynamics framework.

A significant scientific focus has been placed on the mass transfer phenomena between the gas and liquid phase during the last century, as well as on the aqueous-phase chemistry. Numerous scientific articles have been published concerning the physics and chemistry of bubbles, drops, liquid sheets, and contactors. However, reliable experimental data for single droplet absorption of gases are not abundant, while numerical models are focused on liquid–liquid systems, and even the pertinent models include a significant amount of assumptions and unknown parameters. On the other hand, chemical models are predominantly focused on the lime slurry sprays, with only a small number concerning the seawater chemistry.

Taking the above-stated into account, present work aimed to develop a model for the sulfur dioxide absorption in pure water droplets and to study the available approaches for modelling of mass transfer coefficient, to develop a tool for simulation of the removal of harmful emissions.

First, the Mathematical model is presented. It is divided into the modelling of chemical reactions in the aqueous phase, and the overview of the approaches used for the mass transfer modelling. After that, the simulation results and the comparison with three sets of experimental data are shown. Finally, the summation of the work and model is given in the Conclusion.

2 Mathematical model

When considering the sulfur dioxide absorption by liquid droplets, two main parts must be taken into consideration. First are the chemical reactions, or the equilibrium concentration of the absorbed pollutant, indicating the maximum amount of the chemical species that can be absorbed in a liquid. Second is the mass transfer across the gas–liquid interface, controlling the absorption dynamics. The idea behind the approach used in this work was to implement the single-droplet, lumped-parameters model for pure water, combining the liquid-phase chemistry and different models for mass transfer absorption. The model can be used as the basis of a tool for the design and optimization of the industrial equipment for the removal of pollutant emissions.

2.1 Equilibrium conditions

When a gas mixture is in contact with the liquid, its constituents dissolve in the liquid phase, and the amount is proportional to their partial pressure in the gas phase. This is Henry's law, which is valid for dilute solutions and is suitable for the considered application of sulfur dioxide absorption from flue gases. Sander [14] gave a detailed introduction to the Henry's constant: physical background, variations of Henry's law constants and dimensions, dependence on the temperature and solution composition, and the compilation of Henry's constants for various species and water as a solution. Henry's solubility constant is defined as:

$$H_{SO_2}^{cp} = c_{SO_2} P^{-1}. \tag{1}$$

In the above expression, c_a is the concentration of a species in the aqueous phase (mol/m_{aq}^3), P is the pressure (Pa). Temperature dependence of Henry's constant can be described with the van't Hoff equation:

$$H(T) = H^{\ominus} \exp\left(\frac{-\Delta H}{R}\left(\frac{1}{T} - \frac{1}{T^{\ominus}}\right)\right). \tag{2}$$

where the exponent ⊖ denotes the reference values of the temperature and Henry's constant, while $-\Delta H/R$ is tabulated. Equation (2) and the parameters presented are valid only for a limited temperature range. At present, the value of 1.2×10^{-2} is used for $H^{\ominus}$ and the value of $-\Delta H/R$ is 2,850 K.

2.2 Liquid-phase chemistry

To determine the dissolution of sulfur dioxide in water, Henry's law alone is not enough, since it does not include the influence of chemical reactions in the liquid phase. SO_2 is well soluble in pure water, but subsequent reactions further increase maximum dissolved concentrations. In aqueous solutions, sulfur dioxide undergoes dissociation reactions forming bisulfite and sulfite ions.

First reaction is the dissolution of gaseous SO_2 into the unreacted, aqueous form, governed by Henry's law. Next is the dissociation of SO_2, forming the bisulfite and the hydronium ion (here, the H_3O^+ will be used instead of H^+). The reactions below are fast and are in equilibrium, and the reaction quotient for the first dissociation is:

$$Q_I = \frac{c_{HSO_3^-} c_{H_3O^+}}{c_{SO_{2aq}}}, \quad (3)$$

with its temperature dependence from Marocco [15].

The dissociation of sulfur dioxide in water occurs in two stages, and the second one is the reaction of bisulfite with water, forming sulfite. Below is the equilibrium constant of this reaction, with its temperature dependence obtained from Rabe and Harris [16].

$$Q_{II} = \frac{c_{SO_3^{2-}} c_{H_3O^+}}{c_{HSO_3^-}}, \quad (4)$$

The second dissociation reaction is slower than the first one and can often be neglected due to the low concentrations of sulfite ions.

After defining variables and calculation of the auxiliary values, a loop through the range of partial pressures calculates the SO_{2aq} using Henry's constant. This concentration is the input parameter in the system of equations calculating the species' concentration. Since there are three unknowns (HSO_3^-, SO_3^{2-}, and H_3O^+), and there are only two dissociation reactions, the additional equation is needed for "closing" the system. In literature, electroneutrality is commonly used as an additional condition in aqueous solutions:

$$c_{H_3O^+} = c_{HSO_3^-} + 2c_{SO_3^{2-}}. \quad (5)$$

After solving the system, the total sulfur concentration is summed up and converted from moles per kilogram to gram per mole.

$$\text{Total sulfur} = c_{SO_{2eq}} + c_{HSO_3^-} + c_{SO_3^{2-}}. \quad (6)$$

2.3 Mass transfer dynamics

As stated previously, besides the equilibrium concentration, another parameter influencing the mass exchange across the interface is the mass transfer coefficient. Its modelling for different technical applications is still an open topic. Choosing the appropriate model for the mass transfer is a complex issue, since droplets can display various intricate physical phenomena on small time and length scales [17]. The correct model for the mass transfer in droplets is challenging to develop, since there are myriad of parameters that influence the transport, such as droplet velocity and size, turbulence, droplet pulsations, and internal circulation [9]. Ambient conditions greatly influence the absorption process and can modify the mass transfer by orders of magnitude. Furthermore, interaction with the wall has a major impact on droplet properties and the gas–fluid interaction [18]. A significant amount of research has been done on the topic of interface mass transfer during the better part of the last century [19], but a simple and comprehensive unified model is still not available. A major share of the research is focused on general mass transfer across straight surfaces: in agitated systems, or for rising bubbles, extension of these models to droplets falling with terminal velocity is questionable. Usually, the model can be confidently used on a narrow set of conditions that it is derived for and changing some of the dimensionless numbers can lead to major discrepancies with experimental data.

The total mass transfer coefficient $K_{SO_2,tot}$ is constituted out of gas ($k_{SO_2,g}$) and liquid ($k_{SO_2,l}$) side mass transfer coefficients. The resistance of each phase influences the overall coefficient, and their ratio determines whether the mass transfer is liquid- or gas-side dominated. This depends on the application and the parameters, but in the present case, the liquid side proved to be the dominating one, which is confirmed by the literature [20]. In this work, the lumped-parameter model has been assumed, which is commonly used in the computational fluid dynamic models for absorption, but the internal distribution is neglected this way. Thus, there is a discrepancy between the literature and the present model for the investigation of mass transfer controlling side, because detailed models from the literature assume radial concentration distribution in the droplet or discretize the interior. In the below equation, E_{SO_2} is the enhancement factor that accounts for a higher driving force in the liquid film compared with the absorption without chemical reactions [21]:

$$\frac{1}{K_{SO_2,tot}} = \frac{1}{k_{SO_2,g}} + \frac{H_{SO_2}}{E_{SO_2} k_{SO_2,l}}. \tag{7}$$

Here, the gas side mass transfer coefficient is calculated by the Ranz–Marshall equation [21], which is a commonly used approach:

$$\mathrm{Sh} = \frac{k_{SO_2,g} R T d}{D_{SO_2,g}} = 2 + 0.69\,\mathrm{Re}_l^{0.5}\mathrm{Sc}^{0.33}. \tag{8}$$

Sh, Re, and Sc are the droplet Sherwood, Reynolds, and Schmidt dimensionless numbers, respectively; R is the universal gas constant, T is temperature, d is droplet diameter, and $D_{SO_2,g}$ is the binary diffusivity of SO_2 in the air.

One of the first models for liquid side mass transfer was based on the film theory, described by Whitman [22], developed for the absorption of gases in horizontal stirred surfaces. It assumes that the liquid surface is steady, that no turbulent motion exists inside the droplet, and that the mass transfer is limited by the diffusion across the film near the surface. In the equation below, $D_{SO_2,l}$ is the diffusivity coefficient of SO_2 in water:

$$k_l^{\text{film}} = 10\frac{D_{SO_2,l}}{d}. \tag{9}$$

In reality, there is a significant amount of circulation inside falling droplets caused by shear stress, which makes the film theory unsuitable for a range of applications. Internal circulation increases the mass transfer, and even small droplets can have larger k_l than predicted by film theory [23]. Furthermore, experimental data have shown that the proportionality between the mass transfer coefficient and diffusivity is not linear, but it scales with the power of 1/2 [19].

One of the more commonly used approaches is the penetration theory by Higbie, which assumes that the internal turbulence extends to the surface, with eddies bringing portions of fresh liquid to the interface [24]. Penetration theory takes into account the unsteadiness of the mass transfer process with the contact time parameter, which is difficult to estimate for specific applications and even harder to generalize.

$$k_l^{\text{P.T.}} = 2\sqrt{\frac{D_{SO_2,l}}{t\pi}} \tag{10}$$

An additional expansion of the penetration theory is the surface renewal theory introduced by Danckwerts [25]. In it, a surface element can be replaced at any instant of its life time, which accounts for the unpredictability of the turbulent flow inside the droplet:

$$k_l^{\text{S.R.}} = \sqrt{D_{SO_2,l}\, s}\,. \tag{11}$$

where s is a surface renewal rate, which is not generally known, just as the contact time t in penetration theory. These parameters need to be handled carefully since they significantly influence the values of k, but there is no general model for all applications and conditions.

Another expansion of the model is possible by including the influence of droplet oscillations and surface stretch [26]. Angelo et al. developed the model that also scales with $D^{1/2}$ and includes the stretching of the surface via parameter α, and models the oscillation time τ by Lamb's model [27], with surface tension σ and liquid density ρ:

$$\begin{aligned} k_l^{\text{Angelo}} &= \sqrt{\frac{D_{SO_2,l}}{\pi t}\left(1 + \alpha + \frac{3\alpha^2}{8}\right)} \\ \tau &= \frac{\pi}{4}\sqrt{\frac{\rho_l d^3}{\sigma}}. \end{aligned} \tag{12}$$

Lastly, a model by Amokrane et al. [28] includes the interfacial velocity $u_\star$ and oscillation frequency ω, which relates shear stress with the mass transfer coefficient.

$$\begin{aligned} k_l^{\text{Amokrane}} &= \omega\sqrt{\frac{D_{SO_2,l} u_\star}{d}} \\ u_\star &= \sqrt{\frac{\tau}{\rho_l}} \end{aligned} \tag{13}$$

A large number of mass transfer models are available, such as Newman's model for stationary drops or Kronig–Brink model for creeping internal flows, but the models presented here are most common and more suitable for the application under consideration.

Finally, the molar flux across the interface is calculated as follows, with P' being the partial pressure:

$$N_{\mathrm{SO_2}} = K_{\mathrm{SO_2,tot}}\left(P'_{\mathrm{SO_2}} - H_{\mathrm{SO_2}} c_{\mathrm{SO_{2(aq)}}}\right). \tag{14}$$

3 Results

The approach by Andreasen and Mayer [12] has been followed for the equilibrium approach. They provide results of their model for the equilibrium concentrations of SO_2 absorbed in pure water. The model is applicable not only for the water droplet but also for the equilibrium conditions at the water–gas interface. However, it does not provide information regarding the dynamics of absorption into the droplet.

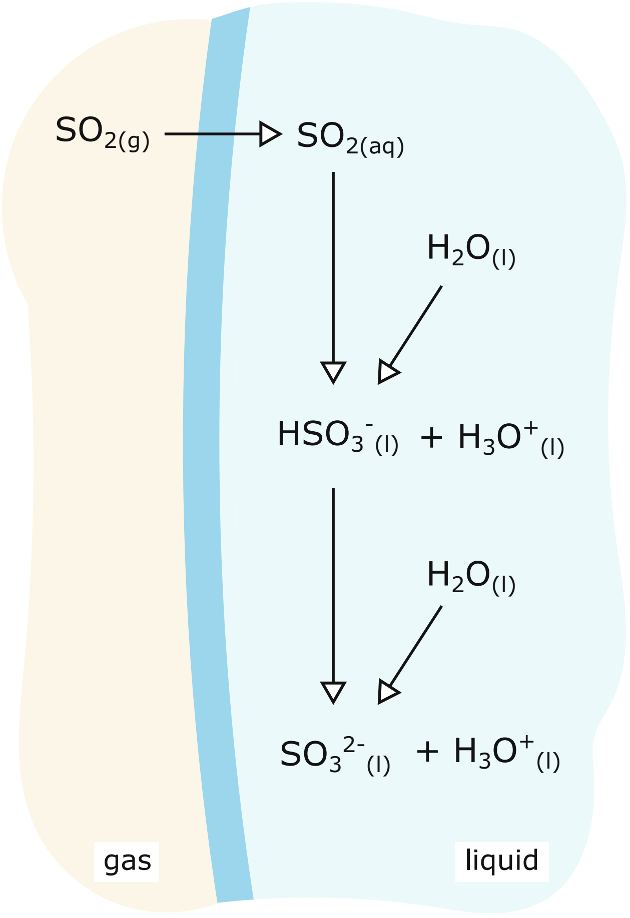

Figure 1: Chemical reactions in liquid phase.

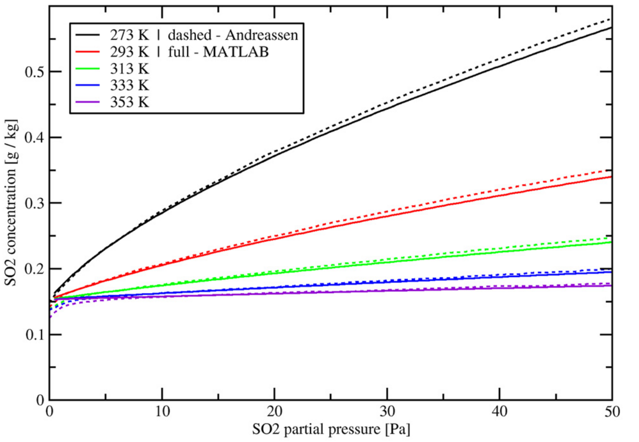

Figure 2: Comparison of the MATLAB model with the literature data [12].

In Figure 1, the sulfur dioxide concentrations are given for the range of SO_2 partial pressures from 0 to 50 Pa and several temperatures from 273 to 353 K. According to equation (6), sulfur dioxide concentration is the total sulfur, combining the contribution of the dissolved $SO_{2(aq)}$, HSO_3^- and the SO_3^{2-}, in gram per kilogram. Comparison of the MATLAB model with the results by Andreasen and Mayer is given for the pure water in Figure 2. It can be seen that the implemented lumped parameter model in MATLAB reproduces the equilibrium concentrations quite well. The increase due to the rise of partial pressure is not linear (as would be with Henry's law only), but curved due to the chemical reactions in the liquid phase. The high concentration at near-zero pressure would be avoided if the resolution of calculation pressures was increased. It can be concluded that the agreement is satisfactory and that the results are correctly reproduced.

Different equations used for the liquid side mass transfer coefficient are expected to provide varied results and, as the liquid side has a bigger influence on the total coefficient in this application, this choice has a critical effect on the overall mass transfer dynamics. Comparison of the coefficients by the presented approaches and the literature values has been made, and they differ significantly. This is expected since a single equation cannot cover a wide array of conditions for all the droplets in the spray.

Experiments for determining the absorbed sulfur dioxide for a single drop are challenging to perform. There are issues with obtaining droplet sizes, duration of the contact time with the atmosphere, accurate measurement on a droplet scale, and at the end, there is a question of how wide is the range of parameters that the results are valid. Here, the model results will be compared with three sets of data.

One is for low sulfur dioxide concentrations in the gas phase (97 and 1.035 ppm), droplet diameter of 2.88 mm, and long exposure times by Saboni and Alexandrova [10]. The second is the experimental investigation by Kaji et al. [29]

for short exposure times, large droplets, and higher SO_2 concentrations. The last are results for smaller diameters and range of SO_2 concentrations in the gas phase (300 µm and 1–100% SO_2) by Walcek [9]. The single droplet is suspended in a stream of gas or dropped from a high shaft, and after some time in contact with the sulfur-rich atmosphere, the concentration of transferred sulfur dioxide is calculated. Reproduced results are presented in Figures 3 and 4.

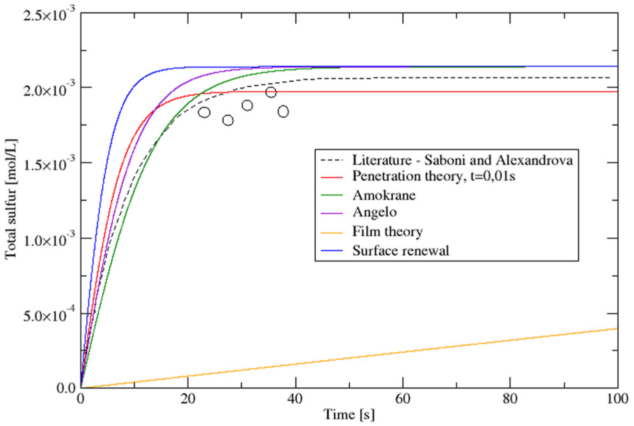

Figure 3: Comparison with literature data [10]. Single falling droplet, *d* = 2.2 mm; 97 ppm SO_2.

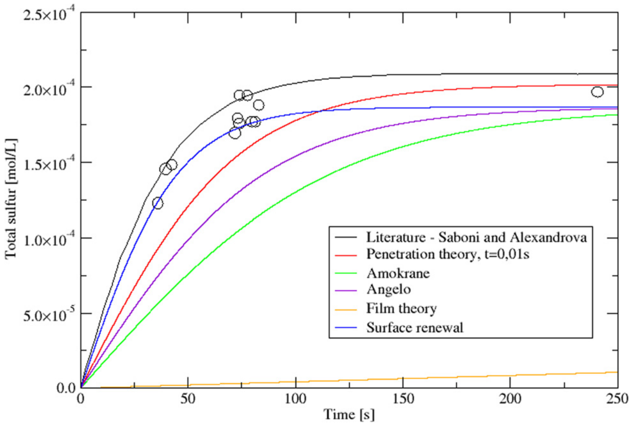

Figure 4: Comparison with literature data [10]. Single falling droplet, *d* = 2.2 mm; 1.035 ppm SO_2.

For both experimental cases of Saboni and Alexandrova, the penetration theory with short contact time shows very good agreement with the literature data. Besides the film theory, which is theoretically more suitable for long contact times, all models are meant to be used for the short exposures of the drop. Experiments above show the contact time around 20 s, which is too high for the application in scrubbers, but both the literature model and experimental data can be reached with presented models.

It can be seen that for higher concentrations and for a longer time, the transferred quantity increases and that the equilibrium concentration is asymptotically approached after sufficient exposure time. In the second case, with very small SO_2 concentrations, discrepancies between the models and the literature data are more significant.

A similar experiment by Kaji et al. [29] was replicated. Here, only short time absorption has been investigated in a short drop tube, as shown in Figure 5 the penetration theory and surface renewal theory display good agreements with the experimental data, while other approaches showed greater discrepancies and are therefore not depicted.

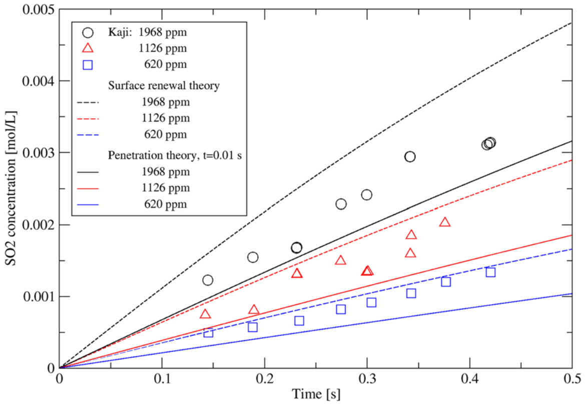

Figure 5: Comparison with literature data [29]. Single falling droplet, *d* = 2.2 mm; *T* = 20°C; 620, 1,126 and 1,968 ppm SO_2, penetration and surface renewal theory.

For comparison with the literature data by Walcek, film theory model showed better results than others.

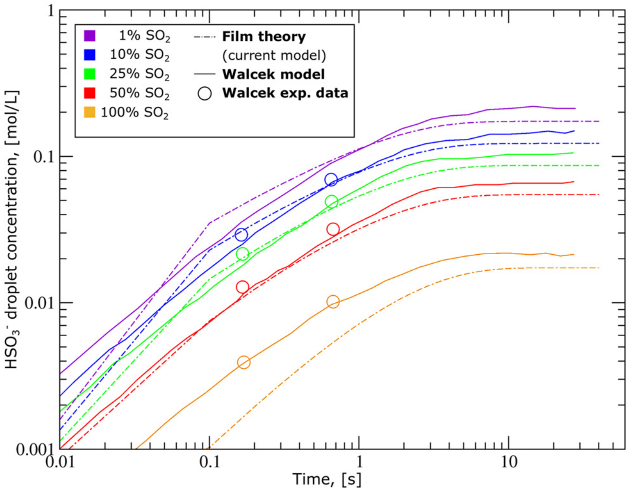

Figure 6: Comparison with literature data [6]. Single falling droplet, *d* = 300 µm; 1–100% SO_2); film theory.

It produced faster absorption dynamics at the beginning of the simulation, as well as small overpass at the end, as shown in Figure 6. The discrepancy between the models and better results by the film theory can be possibly explained by the smaller droplet sizes (300 µm). The results confirm the known fact that the influence of the droplet size, SO_2 concentration, and velocities has a great and complex impact on the absorption dynamics and that at present the unified model cannot envelop all the conditions that can be encountered.

4 Conclusion

The goal of this work was the implementation of the SO_2 absorption model in water droplets, to develop a tool for the design and optimization of industrial scrubbers. A single-droplet, lumped-parameter absorption model has been described with the in-droplet chemistry of sulfur dioxide in pure water. An analysis and comparison of the liquid side mass transfer coefficient models were performed, with models ranging from rudimentary ones that model diffusive transport only to the more complex approaches taking into account phenomena such as flow recirculation and droplet oscillation. In total, four models for the liquid-side mass transfer coefficient were investigated and compared with three sets of experimental data. In the case of low sulfur dioxide concentrations and big droplets with long exposure times, penetration theory with short contact time proved to have the best agreement. When large droplets and short exposure times in atmosphere of higher SO_2 concentrations were observed, penetration theory with short contact time and surface renewal theory achieved the best matching with the measured value, while only in the case of small droplets and high SO_2 concentrations film theory proved to be the better approach. In the presented work, it is shown that the implemented model reproduces the results comparable with the experimental data, with the temperature, concentration, droplet size, and velocity conditions representative for the real industrial cases. It represents a solid basis for the next phase of the work that will consist of applying the developed model to the real industrial case within the CFD framework.

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