Experimental and numerical analysis of mass transfer in liquid films under counter-current gas

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Résumé :

Dans cet article, nous étudions l'absorption de CO_2 dans les écoulements diphasiques en canalisation, quand un film liquide mince est cisaillé par un écoulement de gaz en contre-courant. Les deux phases étant réactives, des réactions chimiques se produisent au niveau de l'interface gaz-liquide et jouent un rôle important dans les transferts associés. Nous présentons des résultats expérimentaux sur le taux d'absorption de CO_2 du gaz vers le liquide dans des canalisation inclinées. Il est démontré que l'augmentation de la vitesse de gaz entraîne une augmentation du taux d'absorption, alors que le taux d'absorption diminue lorsque le débit du liquide augmente. Ces résultats expérimentaux sont ensuite utilisés pour valider notre code numérique de simulation directe.

Abstract :

In this paper, we investigate the absorption of CO_2 in two-phase channel flows, when a thin liquid film is sheared by a counter-current gas flow. Since the two phases are reactive, chemical reactions occur at the gas-liquid wavy interface and enhance the mass transfer. We present experimental results for an inclined channel and show that increasing the gas load leads to an increase of the absorption rate, whereas the absorption rate decreases when the liquid load increases. The experimental results will also serve as validation for our level-set flow solver.

Mots clefs : thin films, gas flow, mass transfer, carbon capture

1. Introduction

Two-phase flow dynamics affects several industrial processes and technologies. In the chemical industry, the capture of carbon dioxide through absorption units relies upon the interaction between a thin liquid

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film and the flue gas. These two phases flow in a counter-current fashion, resulting that the interface might be disturbed with occurrence of waves. The waviness of the interface increases the interacting area and enhances the mass transfer between the two phases, e.g. more CO_2 is absorbed into the liquid [1]. In addition, if the phases are reactive, and this is often the case of absorption and distillation technologies, the chemical reactions at the gas-liquid interface also influence the mass transfer rate. In this work, we investigate the absorption of carbon dioxide into an aqueous solution of monoethanolamine. The experimental set-up consists of an inclined channel where a thin film falling under the action of gravity is sheared by a counter-current gas flow driven by an applied pressure gradient. Through titration of samples, we measure the absorption occurring at different flow conditions, by varying the liquid flow rate, the gas speed and its composition, as well as the pressure inside the channel. It is observed that an increase of the liquid load leads to a decrease of the absorption rate, whereas when the gas speed is increased, so does the absorption rate. Interestingly, the amount of CO_2 absorbed into the liquid is not affected by the CO_2 starting concentration of the solvent, at least until saturation is reached. Aiming to support the experimental results with numerical simulations, we implement a novel module for mass transfer in our level-set flow solver taking into account a variant of the ghost-fluid formalism. Our in-house flow solver applies the level-set method along with a continuous surface-tension model; it is also developed to run on super-computer through distributed or shared-memory architecture. The hydrodynamics module of this flow solver has been already validated against linear and non-linear theory, as well as experiments. With the implementation of the mass-transfer module, we validate the scenarios with and without hydrodynamics by comparing the species concentration in the bulk flow to the analytical solution. In a final stage, we implement the chemical reaction numerical module to perform analysis of the absorption rate in reactive flows, and try to reproduce the above-mentioned experimental results in order to explore the active role of the waves at the interface.

2. The experimental set-up

The experimental set-up is depicted in Fig. 1. It consists of an inclined channel with liquid falling from the top against a counter-current gas flow. The test-cell is inclined at $38°$ with respect to the horizontal. The liquid is collected into a reservoir at the bottom of the test-cell and is re-injected in the channel through a pump, which can be controlled in order to investigate the effect of several liquid loads. Pressure in the gas and temperature in the liquid are controlled and maintained constant during the run. The first version of this experimental apparatus was used for mass transfer with textured walls [2].

The liquid used in our experimental campaign is an aqueous solution of monoethanolamine (MEA) at 30% in weight, which is commonly adopted in absorption applications. The corresponding physical properties are described in [3]. Two different counter-current gases are used in the experiments: pure CO_2 and a mixture of CO_2 (at 16% mol) and nitrogen. When the two phases interact within the channel, CO_2 is chemically absorbed from the gas to the liquid mainly thanks to the chemical reactions occurring at the interface between the MEA and the gas flow. The absorption rate is obtained by getting a liquid sample at the bottom of the test-cell, and then by titration of the same sample. This technique provides the composition of water, CO_2 and MEA, and is averaged among several measures for the same sample in order to reduce the measurement error.

Figure 1: Sketch of the experimental set-up: a liquid film flows down the inclined channel and interacts with a counter-current gas flow.

3. Main experimental results

The aim of our experimental work is to measure the absorption rate and investigate independently the effect of liquid load, gas load, gas composition and pressure. To begin with, the liquid load is varied during the experimental run and the absorption of CO_2 has been measured with five different liquid loads, ranging from $0.2\ m^3/m/h$ to $0.4\ m^3/m/h$. Fig. 2 shows that the absorption rate, measured as percentage in weight of CO_2 in the liquid phase, decreases when the liquid load is increased. This interesting result can be explained as follow: although an increase of the liquid load returns a more disturbed and wavy interface intensifying the liquid-gas mass transfer, meanwhile the interaction time between the two phases decreases (the liquid flows faster in the channel) causing that the absorption drops down.

The second set of measures has consisted in varying the gas load, which is reported using the F-factor $F = U_G\sqrt{\rho_G}$, where U_G and ρ_G are the superficial gas velocity and gas density, respectively. By comparing the red and the green lines in Fig. 2, one notices that increasing the F-factor leads to an increase of the absorption rate of CO_2.

Subsequently, the pressure within the test-cell has also been varied, revealing that absorption increases at higher pressure although with a modest impact. On the contrary, we show that the gas composition plays a great role on the gas-liquid absorption. Indeed, when the counter-current gas consists of pure CO_2, the measurements of carbon dioxide within the liquid have provided greater absorption rates than those obtained with a counter-current gas mixture of CO_2 (at 16% mol) and nitrogen. This is shown on the left hand side of Fig. 3.
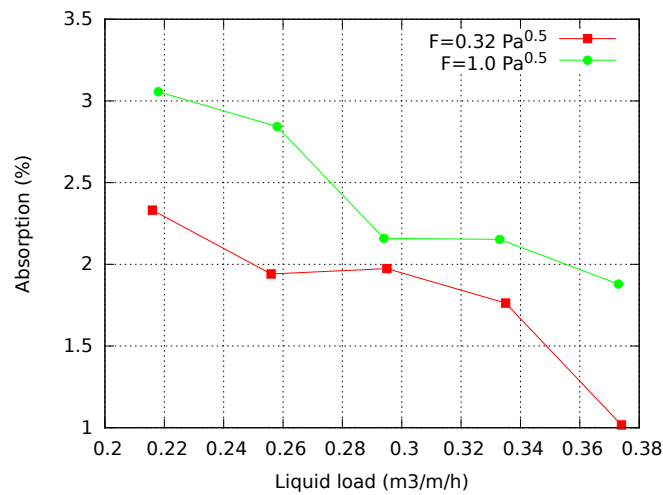
Figure 2: Absorption rate of CO_2 as function of liquid load for two F-factors, i.e. the gas load.
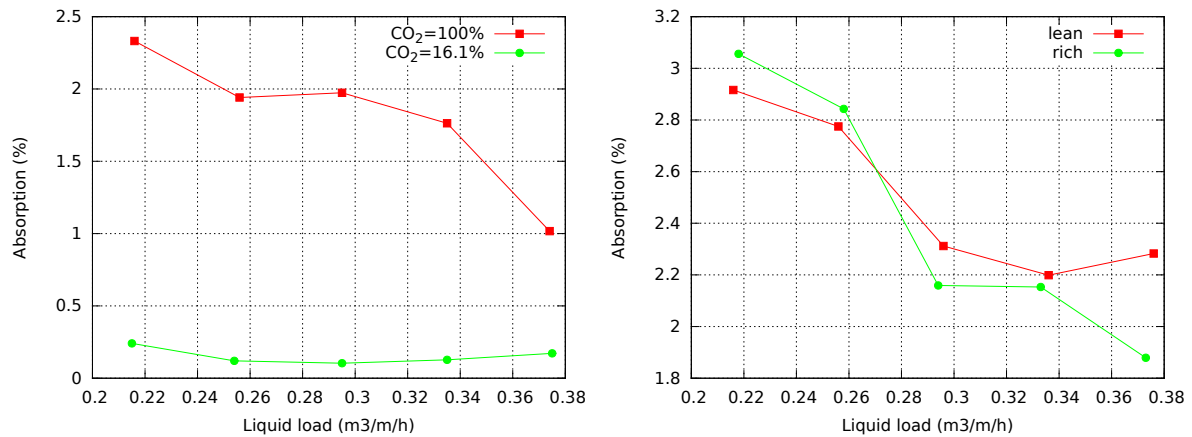

Figure 3: Absorption rate of CO_2 as function of liquid load: effect of varying the gas mixture (left hand panel) and relative effect of the starting CO_2 liquid concentration (right hand panel).

Finally, another experimental campaign has been performed by changing the starting concentration of CO_2 in the liquid phase. Instead of disposing of the "rich solvent", i.e. the solvent loaded with CO_2 issued from previous runs, we have recycled it, aiming to investigate how a solvent rich of carbon dioxide can still absorb from a counter-current gas. We have thus compared the absorption rate obtained through a lean solvent (with no CO_2 at the starting point) against a rich solvent loaded with 2.5% of CO_2 in weight. Noteworthy is that no remarkable variations are found in the absorption rate, meaning that the rich solvent is still capable to absorb the same quantity of carbon dioxide as the lean solvent (under the same configurations of liquid load, gas load and pressure). The right hand side of Fig. 3 shows that both the solvents give the same relative absorption (note that the absolute concentration of CO_2 for the rich solvent is obtained by adding 2.5% to the green-line data). The absorption of carbon dioxide starting with a rich solvent is supposed to vanish when saturation of CO_2 is reached in the liquid phase.

4. Conclusions

The series of experimental data shown here will complement the validation of our numerical tool. This is an open-source 3D solver optimised for the Archer UK Supercomputer and based on a level-set DNS approach, see [4]. Velocities and pressure are discretized on a marker-and-cell (MAC) grid, through second- and third-order space discretization. Diffusion mechanism is governed by the advective-diffusion equation, which is implemented in the level-set formalism. Boundary conditions at the interface satisfy the equilibrium concentration through the Henry's law. The mass-transfer module is to be validated first by comparing the purely diffusive case (without convection) to the analytical solution. The following step will consist in validating the absorption rate with hydrodynamics between two non-reactive flows. The final validation will address instead the comparison of absorption of CO_2 with chemical reactions against the experimental results presented above. This thorough validation will provide a novel and efficient numerical module to take into account the liquid-gas interfacial mass-transfer in presence of chemical reactions, aiming to extend this work to more complex geometries, such as the 3D elementary cell of typical structured packings used in carbon capture absorption units.

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