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Experimental and numerical analysis of buoyancy-induced instability during CO_2 absorption in $NaHCO_3$–Na_2CO_3 aqueous solutions

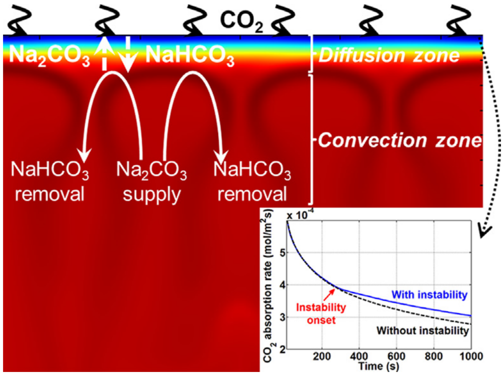
C. Wylock*, A. Rednikov, P. Colinet, B. Haut

Transfers, Interfaces and Processes, Université Libre de Bruxelles, 50 av. F.D. Roosevelt CP 165/67, 1050 Brussels, Belgium

HIGHLIGHTS

- CO_2 absorption in aqueous solutions of $NaHCO_3$ and Na_2CO_3 in a Hele-Shaw cell.
- Visualization of density variations resulting to unusual buoyancy-induced instability.
- Numerical simulation results in qualitative agreement with the experiments.
- Simulation analysis enables the proposition of mechanism for the observed dynamics.
- The absorption rate is not significantly enhanced by this particular dynamics.

GRAPHICAL ABSTRACT



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ABSTRACT

This work deals with the experimental analysis and the mathematical modelling of the CO_2 absorption in an initially quiescent aqueous solution of $NaHCO_3$ and Na_2CO_3 inside a Hele-Shaw cell. This absorption, initially driven by the coupling between diffusion and chemical reactions, eventually leads to the apparition of a peculiar gravitational instability with non-monotonic dynamics, and liquid plumes generated at some distance from the gas-liquid interface (Wylock et al., 2014). Considering that this instability is triggered by a Rayleigh–Taylor like mechanism, a two-dimensional model, coupling diffusion, chemical reaction and convection, is proposed to simulate the onset and the dynamics of such an instability. It is observed that the simulated instability dynamics agree qualitatively with the experimental observations and that the order of magnitude of the onset time is well estimated. Thanks to the simulation, the interaction between the various phenomena after the instability onset is further investigated and a mechanism is proposed to explain the unusual dynamics of the studied system. It is notably shown that this dynamics is due to the particular non-monotonic liquid density variations with the depth, induced by the absorption. A criterion to obtain such type of density profile is presented. In addition, the simulation enables to assess the influence of the instability on the CO_2 absorption rate and it is observed that the generated flow pattern does not lead to a significant enhancement of the gas–liquid absorption rate. This result is of significant importance for optimizing chemisorption (e.g. for CO_2 capture or sequestration) processes.

1. Introduction

Numerous chemical engineering operations are based on the absorption of a gaseous component into a liquid phase. It is commonly admitted that the global gas–liquid mass transfer rate

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* Corresponding author.
E-mail address: cwylock@ulb.ac.be (C. Wylock).

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Nomenclature

Latin

$[X]$	concentration of species X, mol/m^3
d	computational domain depth, m
D	diffusion coefficient, m^2/s
$\tilde{D}$	ratio of the diffusion coefficient of C to the diffusion coefficient of B
DV	density variation
e	distance between the two parallel plates of the Hele-Shaw cell, m
g	gravity acceleration, m/s^2
h	solubility coefficient
k	kinetic constant
K	equilibrium constant
MZI	Mach–Zehnder interferometer
N	averaged interfacial gas–liquid transfer rate, mol/(m^{2}s)
NSD	Navier–Stokes–Darcy
p	pressure, Pa
r	reaction rate, mol/(m^{3}s)
R	ideal gas constant, J/(mol K)
RI	refractive index
RT	Rayleigh–Taylor
t	time, s
T	absolute temperature, K
u	horizontal component of the velocity, m/s
v	vertical component of the velocity, m/s
w	computational domain width, m
x	horizontal coordinate, m
y	vertical coordinate, m
$\tilde{y}$	dimensionless vertical coordinate

Greek

$\tilde{\delta}$	ratio of the density contribution of C to the density contribution of B
Δ	variation
μ	dynamic viscosity, kg/(m s)
ν	stoichiometric coefficient
$\tilde{\nu}$	ratio of the stoichiometric coefficient of C to the stoichiometric coefficient of B
ρ	density, kg/m^3

Superscript

d	diffusion case
dr	diffusion–reaction case
drc	diffusion–reaction–convection case
$dr-sl$	diffusion reaction in stagnant layer case
exp	experimental
sim	simulated

Subscript

0	at initial time
onset	at instability onset
w	water

in these operations is controlled by phenomena occurring in layers close to the gas–liquid interface (Bird et al., 2002; Coulson and Richardson, 1999; Danckwerts, 1970; Trambouze and Euzen, 2004), such as the diffusive transport of the involved species and the chemical reactions. A good understanding of these phenomena is thus of fundamental interest for the modelling of these operations.

Recently, the carbon dioxide (CO_2) absorption in aqueous alkaline solutions inside a Hele-Shaw cell became more and more studied in the framework of the CO_2 sequestration in deep saline aquifers (Loodts et al. 2014a,b; Javaheri et al., 2010; Kim and Choi, 2012; Kim, 2015; Riaz et al., 2006; Rongy et al., 2012; Wylock et al., 2008, 2011, 2013, 2014). Using these Hele-Shaw cells, many of these studies have indeed highlighted the occurrence of a Rayleigh–Taylor (RT) instability when CO_2 is absorbed in alkaline solutions, resulting from an unstable liquid density stratification during the absorption. Indeed, the chemical reactions involving CO_2 that take place in such solutions may modify locally the liquid density in the reaction zone, leading to the appearance of natural convection when heavier liquid lies upon lighter liquid.

The parallel between CO_2 absorption in a liquid in a Hele-Shaw cell and CO_2 sequestration in porous media such as saline aquifers lies in the fact that the only possible mixing mechanism to add up to the effect of chemical reactions in enhancing the CO_2 transfer is buoyancy-induced convection. The analysis of RT instability taking place in a Hele-Shaw cell is therefore of significant importance for further development of CO_2 sequestration processes, especially in order to identify the conditions favoring the overall transfer efficiency.

In previous works, a Hele-Shaw cell has been used to investigate the CO_2 absorption in aqueous solutions of sodium bicarbonate ($NaHCO_3$) and carbonate (Na_2CO_3) (Wylock et al., 2008, 2011). This absorption is indeed met in several industrial processes (e.g. Solvay process for refined $NaHCO_3$ production) and it is an interesting model to analyze the CO_2 absorption in saline aquifers. Using a Mach–Zehnder interferometer (MZI, see Fig. 2), the local density variations (DV) resulting from the CO_2 absorption in the liquid are visualized thanks to the refractive index (RI) variations caused by the DV. Full details of this device and of the associated image processing is given in Wylock et al. (2011).

During experiments, the appearance of RT instability has been systematically observed (see the videos in supplementary material). A typical time evolution of the DV field is presented in Fig. 1. During the first stage of absorption, the DV field is homogeneous on any horizontal line. At this stage, the absorption is controlled only by diffusion and reaction. The DV appears to be negative at the gas–liquid interface and to present a positive maximum at some distance from the interface (as illustrated in Fig. 4). After some time, the onset of a RT instability is observed. Some plumes are generated at a small distance from the interface and fall towards the bottom of the cell. These plumes may sometimes show an oscillating behavior (as in Fig. 1). In such cases, it is observed that the plumes tend to slow down at some depth, soften, and almost vanish. Meanwhile, a second generation of plumes appears in between the primary ones, and the same scenario is possibly repeated several times. In other cases, the plumes do not vanish when they sink but the location of their origin is varying.

The dynamics of the RT instability observed in this system is rather unusual. Indeed, most of the referenced RT instabilities occurring during acid–base reactions correspond to plumes originating at the fluid interface, and penetrating the bulk without any oscillating behavior. Therefore, their effect on the overall gas–liquid mass transfer may be significantly different from those of a classical RT instability.

The objectives of this work are therefore: