Consequence Study of CO₂ Leakage from Ocean Storage

Loi Hoang Huy Phuoc Pham, Risza Rusli, Lau Kok Keong

Center Advance of Process Safety, Chemical Engineering Department, Universiti Teknologi PETRONAS, Bandar Seri Iskandar and 32610, Perak, Malaysia
Research Center for CO₂ Capture, Chemical Engineering Department, Universiti Teknologi PETRONAS, Bandar Seri Iskandar and 32610, Perak, Malaysia.

Abstract

Carbon capture and storage (CCS) technology is considered as a viable alternative for reducing a large amount of CO₂ gas discharged from power plants and steel production plants. CO₂ storage is a part of the CCS to keep the discharged CO₂ at deep sub-seabed geological areas. A leakage of this stored CO₂ may result in a CO₂ bubble plume which causes a pH reduction and an increase in pCO₂ (partial pressure of CO₂) of the ocean environment due to the ocean acidification between the leaked CO₂ and the seawater. Consequently, this leads to the environmental influence on the marine life. The aim of this article is to propose an integrated model of computational fluid dynamics and population balance model to account for the dynamical behaviour of the leaked CO₂ bubble plume via the man-made ocean CO₂ storage against a recently published experiment. The integrated model was used to simulate the processes of the breakup, coalescence and mass-transfer between the CO₂ bubbles and the seawater, and the momentum of the CO₂ bubble plume. Thus, the changes in the pH and pCO₂ of the seawater during the occurrence of the CO₂ leakage, and the rising velocity and size distribution of the CO₂ bubbles in the plume were predicted. It was found that the predictions had a good agreement compared to the published experimental data. In addition, the predicted profiles were used to analyse the consequences of the CO₂ leakage from the small-scale ocean storage.

Keywords: CO₂ bubble; Eulerian model; realizable k-ε turbulent model; population balance model; breakup; coalescence; mass transfer.

1. Introduction

In recent years carbon capture and storage (CCS) technology has been developed to mitigate the large volume of CO₂ gas discharged from fossil fuel power plants, oil refinery plants and cement plants [1]. The deployment of the CCS plants was believed to have a 14% reduction in the CO₂ discharge by 2050 [2]. The CCS system consists of three main processes [3]: (1) capturing CO₂ from the large point sources of emission; (2) compressing and transporting the captured CO₂; and (3) injecting and storing the CO₂ under storage sites. The captured CO₂ can be stored in onshore and offshore geological formations [4]. Global potential storage capacity for CO₂ was estimated by Beck [5]. Hence, the offshore storage can be contributed by depleted gas and oil fields at 690 GtCO₂.

* Corresponding author. Tel.: +605-3687567.
E-mail address: risza@petronas.com.my

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On August 21, 1986, a large volume of CO$_2$-$\text{H}_2\text{O}$ mixture leaked from Lake Nyos in the northwest area of Cameroon, West Africa and killed at least 1700 people and 3000 livestock near the lake and within 14 km radius from the area [6]. This tragic disaster is not associated with the failures of the man-made CO$_2$ storage, however, it should be considered as an evidence to conduct consequence studies for accidental leakages from the geological storage for the CCS performances. Therefore, the IPCC Special Report on Carbon Capture and Storage [3] highlighted that ‘the local health, safety, and environment risks of geological storage would be comparable to the risks of current activities such as natural gas storage, EOR, and deep underground disposal of acid gas.’

There are two main leakage scenarios of the geological CO$_2$ storage [7] including ‘(1) abrupt leakage, through injection well failure or leakage up and abandoned well; and (2) gradual leakage, through undetected faults, fractures or wells.’ In the case of the ocean CO$_2$ storage, the leakages may generate individual rising CO$_2$ droplets in the depth of the marine water between ~3000 and 500 m, and CO$_2$ bubbles if shallower than ~500 m [8]. The leaked CO$_2$ droplets/bubbles travel far from the leakage point to form a plume and it may be diluted by the entrainment of the seawater [9]. This leads to the change of the seawater quality due to the ocean acidification between the leaked CO$_2$ and the seawater [10]. The dissolution rate of the leaked CO$_2$ droplet/bubble depends on its size and rising velocity [11][12]. Particularly, the smaller bubble travels in a nearer distance in the seawater column and it takes a short time to dissolve [11].

Currently, a small-scale leakage experiment named the Quantifying and Monitoring Potential Ecosystem Impacts of Geological Carbon Storage (QICS) [11] was conducted to study the dynamics of CO$_2$ bubbles leaked in the Scottish sea at Ardmucknish Bay. In this experiment, the data profiles including velocity and sizes of the bubbles were observed up to 30 cm from the sea floor when they leaked through the sediment into the seawater. Furthermore, the video filmed during the QICS experiment noted interactions between the CO$_2$ bubbles. Thus, the mechanism of the interactions involved ‘breakup of some of the large CO$_2$ bubbles which reduce their size and, therefore, velocity or coalescence between two CO$_2$ bubbles to give birth to a larger CO$_2$ bubble with higher velocity.’ The QICS experiment also measured the chances in pH and pCO$_2$ of the seawater during the rise of the leaked CO$_2$ bubbles.

Dewar et al. [12] presented an oceanic two-phase plume model to simulate the CO$_2$ bubble plume and the CO$_2$ solution dynamics observed from the QICS field experiment. Thus, the modelling for the behaviours of the CO$_2$ bubble plume included (a) momentum (i.e. rising velocity of the CO$_2$ bubble) and mass transfer (i.e. CO$_2$ dissolution in the seawater); and (b) interactions (i.e. breakup and coalescence) between the bubbles. Dewar found that the simulation from the integration of the mathematical models (a) and (b) performed bubble rise heights which were closer to the QICS experimental data than the single simulation from the models (a). However, the integration of the models has under-predicted the maximum bubble size and the maximum pCO$_2$ peak. Respectively, it was values of 9.8 mm and 713 µatm from the simulation, while the observed data from the QICS experiment were at 12 mm and 1500 µatm.

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**Nomenclature**

| Symbol | Description |
|--------|-------------|
| b | bubble |
| l | liquid |
| Re | Reynolds number |
| g | gravitational acceleration (m/s$^2$) |
| $\alpha_b$ | volume fraction of bubble phase |
| p | pressure shared by all phases (Pa) |
| m | mass transfer rate (kg/m$^2$/s) |
| $B_b$, $B_c$ | birth due to breakup and coalescence of bubbles |
| $D_b$, $D_c$ | death due to breakup and coalescence of bubbles |
| n(v) | number density of bubble (particles/m$^3$) |
| f_i | volume fraction of bubbles of size i |
| V_i | volume of a bubble of size i |
| $\tau_{li}$ | phase stress-strain tensor |
| v | velocity (m/s) |
| $\rho$ | density (kg/m$^3$) |
| $C_o$ | drag coefficient |
| $C_l$ | lift coefficient |
| $C_{mn}$ | user-modifiable constant ($C_{mn}=0.1$) |
| k | turbulent kinetic energy (m$^2$/s$^2$) |
| $\varepsilon$ | turbulent kinetic energy dissipation rate (m$^2$/s$^3$) |
| $\mu_i$ | bulk viscosity for qth phase (kg/ms) |
Computational fluid dynamics (CFD) codes were widely used to simulate the bubble dynamics in the water column such as FLUENT version 14.0 [13], CFX version 12.0 [14], and OpenFOAM [15]. These studies successfully implemented the multi-phase Euler-Euler model to estimate the dynamical behaviour of the bubbly flow by using various equations of the body forces (e.g. drag, lift and turbulent dispersion). However, to account the size distribution and the interactions of bubbles, population balance model (PBM) was integrated into their calculations.

This article aims to present the implementation of the models developed for the bubbly flow in the water column to accurately account for the dynamical behaviour of the leaked CO$_2$ bubble plume in the seawater. Thus, an integrated model of CFD and PBM was used to simulate a two-phase flow of CO$_2$ bubble and seawater. Particularly, the PBM model [16] was integrated with the Eulerian and realizable k-ε turbulent models in FLUENT version 15.0 [17]. The brief of these models was described in the following section.

2. Mathematical Modelling

2.1. Eulerian model

FLUENT [17] includes the Eulerian model allowing for modelling of bubble-liquid flow. This model is based on the Euler-Euler approach which treats mathematically either continuous (primary) or discrete (secondary) phases as continua [17]. For the bubble-liquid flow, the Eulerian model calculates volume fractions for each phase and mechanism for the exchange of momentum and mass between the phases against the equations below:

2.1.1. Volume Fraction Equation

Volume of each phase is defined by

$$ V_b = \int \alpha_b \, dV $$

where

$$ \alpha_b + \alpha_l = 1 $$

2.1.2. Momentum equation

The momentum balance for bubble phase is defined by

$$ \frac{\partial}{\partial t} (\alpha_b \rho_b \vec{v}_b) + \nabla \cdot (\alpha_b \rho_b \vec{v}_b \vec{v}_l) = -\alpha_b \nabla p + \nabla \cdot \tau_b + \alpha_b \rho_b g + \bar{R}_b + \bar{m}_b \vec{v}_b - \bar{m}_l \vec{v}_l + \bar{F}_{b,l} + \bar{F}_{b,s} $$

Equation (2) included the term $\bar{R}_b$ which represents the drag force between the phase ‘b’ and ‘l’. This force is performed by (3):

$$ \bar{R}_b = \frac{3 \alpha_b \alpha_l C_D \rho \mu \vec{v}_l}{4 \delta} (\vec{v}_l - \vec{v}_b) $$

Equation (4) is used to perform the lift force ($\bar{F}_{b,l}$) acting in (2). It is defined by Drew and Lahey [18] and is given as follows:

$$ \bar{F}_{b,l} = -C_l \rho \alpha_b (\vec{v}_l - \vec{v}_b \times (\nabla \times \vec{v}_l)) $$

A model of drag coefficient proposed by Schiller and Naumann [19] and a 0.5 constant of lift coefficient proposed by Auton [20] were used to calculate the impact of the drag and lift forces on the rise of the CO$_2$ bubbles in the seawater.

The term $\bar{F}_{b,s}$ in (2) performs turbulent dispersion force which accounts for the dispersion of bubbles due to transport through eddies. The mathematical model for the turbulent dispersion force used in this study is defined by Bertodano [21]:

$$ \bar{F}_{b,s} = C_{\tau_D} \rho k \nabla \alpha_b $$

2.1.3. Mass Equation

The continuity equation for phase b is defined by (6):

$$ \frac{\partial}{\partial t} (\alpha_b \rho_b) + \nabla \cdot (\alpha_b \rho_b \vec{v}_b) = \bar{m}_b - \bar{m}_l $$
2.2. Realizable k-ε turbulent model

The realizable k-ε turbulent model proposed by Shih et al. [22] was indicated that ‘the realizable model provides the best performance of all the k-ε model versions for several validations of separated flows and flows with complex secondary flow features’ [17]. This model was used to investigate turbulence influence on the mixture of the seawater and CO₂ bubble phases. The turbulence may be caused by velocity fluctuation of each phase [17].

2.3. Population Balance Model

The Eulerian model treated the secondary phase as a continuum with an assumption of a constant of bubble diameter (Eq. (3)). Therefore, in order to model a flow of bubble-liquid in which the bubble phase dispersed in a wide range of size groups, the PBM can be used to integrate with the CFD models. This model can account for the size distribution of the dispersed bubble; the impacts of breakup and coalescence on the bubbly flows; and the dissolution of the bubbles by mass transfer. The PBM method developed by Ramkrishna [16] was used to calculate the size distribution of the leaked CO₂ bubbles. The below equation describes a common formula of population balance equation:

\[
\frac{\partial}{\partial t} (n(v)) + \nabla \cdot (\bar{v}_b n(v)) = B_n + B_c - D_n - D_c
\]  

where the bubble number density, \( n(v) \) (particles per m³), is related to the gas volume fraction \( \alpha_g \) by

\[
\alpha_g f_i = n(v) \tilde{W}_i
\]

3. Case Study

The QICS experiment [11][23] was conducted to study the dynamics of a CO₂ bubble plume leaked into the seawater from the sediment through 35 pockmarks at a 9-m depth of the ocean. In this experiment, the physical properties of the seawater and the CO₂ bubbles measured at 10°C on day 33 at a maximum injection rate of 208 kg/day are shown in Table 1. It was observed that the size distribution of the bubbles was in a range of 2 ÷ 12 mm and the rising velocity differed from 20 cm/s to 45 cm/s up to 30 cm from the sea floor. The sizes of the bubbles were measured as an equivalent spherical diameter. A pH reduction was measured from -1.5 to -2.2 pH during the CO₂ leakage. A pCO₂ increase was measured at 3 cm, 30cm and 9m above the seabed in the ranges of 640÷1140 μatm, 30 μatm and 20~25 μatm, respectively. The measurements were compared with a background of 360 μatm and 8.2 pH. In addition, it was observed that some leaked bubbles reached the sea surface. However, the size distribution of the reached bubbles was not measured.

| Properties                         | Unit  | Seawater | CO₂ bubble |
|------------------------------------|-------|----------|------------|
| Dynamic viscosity                  | mPas  | 1.4      | 14.2       |
| Interfacial tension                | N/m   | 7.37×10^-2 | --         |
| Density                            | Kg/m³ | 1027     | 1.9        |
| Measured salinity                  | ppt   | 33.7     | --         |
| Velocity                           | m/s   | 0.05     | --         |
| Mass release rate                  | Kg/day| 31.2     |            |

3.1. Simulation setup

The leakage experiment of the CO₂ bubbles mentioned above was simulated by using the PBM model integrated with the Eulerian and realizable k-ε turbulent models in FLUENT version 15.0. The 35 pockmarks were assumed as circle holes with the same diameters of 50 cm. The locations of these pockmarks were determined from the previous work of Dewar et al. [12]. Thus, they are formed in an area of 5 m × 15 m when the CO₂ bubbles leaked through the sediment seabed (Fig. 1b). To simulate the dynamics of the CO₂ bubble plume, the computational domain was a box including the 35 pockmarks and its dimension is greater than the region dimension of the pockmarks. The domain spreading 25 m in the Z-axis, 80 m in the X-axis and 9 m in the Y-axis was generated with Design Modeller-ANSYS Workbench (Fig. 1a). This domain was defined with the boundary conditions such as the velocity inlet for
the seawater flow, the mass flow inlet for the CO\(_2\) bubble flow, the outflow for the sea surface, the wall for the seafloor and the symmetry for the remain boundaries. All simulations were run with a mesh generation of the domain at 329,483 cells.

A steady-state simulation of the seawater flow without CO\(_2\) was firstly run to assume for the initial conditions of 360 µatm pCO\(_2\) and 8.02 pH before CO\(_2\) leaked. After the steady-state simulation converged at 248 iterations, the CO\(_2\) bubbles were set to leak into the domain by changing the boundary condition from “wall” to “mass flow rate”. Thus, the simulation of this study was to perform for the pCO\(_2\) increase and the pH reduction. According to the QICS experiment, a transient simulation of the CO\(_2\) bubble leakage was set from the beginning leakage up to 300 s. The time step size of the simulation was set at 0.1 s. Initial diameter of the bubbles was set at a range from 2 mm÷12 mm observed by Sellami et al. [11].

4. Simulation Results and Discussion

4.1. Predictions of ΔpH and ΔpCO\(_2\)

In this study, the integrated model of CFD and PBM only predicted the volume fraction (\(\alpha_b\)) of the leaked CO\(_2\) bubbles dissolved in the seawater. Therefore, the pCO\(_2\) and pH values were calculated based on the predicted profile of the CO\(_2\) bubble volume fraction by using mathematical models developed in the studies of Weiss [24], Lewis and Wallace [25] and Someya et al. [26]. Thus, the pH reduction was predicted in a range of -1.7 to -2.7 pH. The lowest reduction of the pH was predicted with a value of -2.7 pH at 3 cm from the seabed. It was compared that the predicted results of the pH reduction were higher than the published data measured at -1.5 to -2.2 pH.

The pCO\(_2\) increases were predicted as a function of time at 3 cm from 0 to 1184 µatm (Fig. 2a), at 30 cm from 0 to 203 µatm (Fig. 2b), and at 9 m between 0 µatm and 50 µatm (Fig. 2c). It was noted that the simulation results of the pCO\(_2\) increase are higher than the data observed from the QICS experiment which mentioned in section 3. Additionally, the pCO\(_2\) increases were predicted in the maintaining values of 1149 µatm at 3 cm; 39 µatm at 30 cm; and 16 µatm at 9 m.

Both the simulation and the published data indicated that the pCO\(_2\) of the seawater highly increased near to the pockmark area. Then, it decreased along the height of the seawater column. Particularly, the highest peak of pCO\(_2\) at 3 cm was a value of 1544 µatm from the simulation and at 1500 µatm from the experiment for a short period. However, it decreased at the sea surface at 410 µatm from the simulation and at 385 µatm from the experiment.

Gibson et al. [27] presented that the pCO\(_2\) of 1000 µatm in seawater may cause an increase in death rate of marine larvae, embryos, and juveniles. This increase depends on the species, especially in smaller invertebrates. Hence, it was suggested that there is no effect of the leaked CO\(_2\) on the marine life against the simulation results and the published experiment.
4.2. Prediction of velocity distribution

Fig. 3 shows the velocity distribution of leaked CO₂ bubbles from the simulation and the experiment. Thus, the simulation results had a good agreement compared to the experimental data in which most of the bubbles were observed with a rising velocity between 25 cm/s and 45 cm/s.

The rising velocity distributions were estimated on 354 bubbles which leaked through the pockmarks into the seawater column for both the experiment and the simulation. The simulation predicted that the velocity increased up to time. The increase was predicted by acting the breakup and coalescence models. Sellami et al. [11] observed the interaction between the CO₂ bubbles based on videos filmed during the QICS experiment. From there, it was confirmed that a large CO₂ bubble has broken up into two smaller bubbles, they tended to coalesce with another bubble to give birth to a larger bubble with higher velocity.

![Fig. 3. CO₂ bubble velocity distributions from the simulations at: (a) 10 s; (b) 60 s; (c) 100 s; (d) 250 s; (e) 300 s; and (f) from the QICS experiment.](image)

4.3. Prediction of leaked bubble size

The initial size of the CO₂ bubbles was set to leak into the domain based on the size distribution observed by Sellami et al. [11]. This observed size distribution was also used to set for the initial bubble sizes in the modelling development of Dewar et al. [12]. Sellami et al. [11] observed the size of the leaked CO₂ bubbles in a range of 2–12 mm up to 30 cm above the seabed. Fig. 4 shows the prediction of the number density (number bubbles per m³) of the leaked CO₂ bubbles at 300 s after the beginning of the leakage in
the present study. Thus, the maximum diameter of the CO$_2$ bubble leaked into the seawater column was predicted at 12 mm, while it was at 9.8 mm from the prediction of Dewar et al. [12].

![Graph showing number density of leaked CO$_2$ bubble](image)

**Fig. 4.** Prediction of the number density of leaked CO$_2$ bubble of size $i$ in the seawater column.

The simulation predicted that the leaked CO$_2$ bubbles reached the sea surface (at 9 m) with various sizes from 5 mm to 9.5 mm. Fig. 5 shows the contour of the fraction of CO$_2$ bubble volume fraction at the sea surface. Thus, it was predicted that 15~25 % of the bubble volume fraction had a diameter from 5 mm to 6.25 mm; 12~40 % of the bubble volume fraction ranged in diameters of 6.5÷7.75 mm; and 25~36 % had a diameter varying between 8 mm and 9.5 mm.

![Contour map of CO$_2$ bubble volume fraction](image)

**Fig. 5.** Contours of the volume fraction ($f_j$) of bubbles of size $i$ which reached the sea surface at 300 s after the beginning of the leakage: (a) 5 mm; (b) 6 mm; (c) 6.25 mm; (d) 6.5 mm; (e) 7.25 mm; (f) 7.75 mm; (g) 8 mm; (h) 9 mm; (i) 9.25 mm; (j) 9.5 mm.

According to the experiment and the simulation, it confirmed that the leaked CO$_2$ bubbles reached the sea surface. The reached bubbles can escape into the atmosphere.
5. Conclusions

An integrated model of CFD and PBM has been proposed to predict the dynamics of CO₂ bubbles leaked from the sediment into the seawater. The proposed model can account for the changes in the pH and pCO₂ of the seawater during the leakage event, and the rising velocity and size distribution of the bubbles. It was found that the predictions and the published experiment had a good agreement. Besides, the consequences of the small-scale CO₂ leakage from the man-made ocean storage were analysed against the predicted results. It was investigated that the leaked CO₂ bubble plume had an insignificant influence on the marine life.

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