Chemical Engineering Science 104 (2013) 10–16

Contents lists available at ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

Modeling of CO_2 dissolution by static mixers using back flow mixing approach with application to geological storage

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HIGHLIGHTS

- Back flow cell model (BFCM) is used to simulate mass transfer of CO_2 in water in static mixer.
- Residence time distribution experiments are conducted to find BFCM parameters.
- Optimal Reynolds number of CO_2 and water are obtained using BFCM.
- Proper length of static mixer is obtained for optimal Reynolds numbers.

ARTICLE INFO

Article history:
Received 18 May 2013
Received in revised form
13 August 2013
Accepted 28 August 2013
Available online 5 September 2013

Keywords:
CO_2 dissolution
CO_2 storage
Geological storage
Static mixer
Back flow mixing
Back flow cell model (BFCM)

ABSTRACT

We propose the application of a back flow cell model for the simulation of supercritical CO_2 dissolution in water in static mixers at pressures and temperatures pertinent to geological CO_2 storage. Using step tracer injection experiments, the required parameters for calibration of the back flow cell model, such as the number of the cells and the back flow ratio, were obtained. The calibrated model was then verified with the experimental mass transfer measurements at supercritical conditions, and an acceptable agreement was observed. Experimental data of CO_2 dissolution in water were regenerated with an average absolute relative deviation of less than 5%. Using the developed model, the optimal operating conditions were determined. The developed model will be useful for the engineering design of CO_2 dissolution systems and simulation of supercritical CO_2 dissolution in water for geological CO_2 storage.

1. Introduction

Human activities are believed to be responsible for the growth of global greenhouse gas (GHG) emissions (Metz et al., 2007; IPCC, 2005). The primary source of carbon dioxide (CO_2) emissions, as the major GHG emitted by human activities, is the burning of fossil fuels.

One of the mitigation actions for the stabilization of atmospheric CO_2 concentrations is CO_2 capture and storage, which involves capturing the CO_2 produced from the combustion of fossil fuels and storing it in underground subsurface formations (Bachu and Adams, 2003). The capture of CO_2 can be applied to large emission sources, such as power plants, and then be stored in large-scale storage sites, such as geological formations, oceans and mineral carbonates, or for use in industrial processes (IPCC, 2005; Saito et al., 2000; Haugan and Drange, 1992). The storage of CO_2 in geological formations, particularly in deep saline aquifers, has become the most important mitigation option, because CO_2 storage in geological formations is seemingly a self-contained and volumetrically efficient process.

Deep saline aquifers are sedimentary formations saturated with brines containing high concentrations of dissolved salts that are not suitable for agriculture or drinking. These formations are distributed widely and, in many cases, are located close to sources of high CO_2 emissions. Deep saline aquifer formations often have large storage capacities, making them a suitable option for the geological storage of CO_2 (Gale, 2004). However, the upward migration of CO_2 due to the density contrast between the injected CO_2 and the host brine increases the risk of leakage of CO_2 from aquifers. This risk can be decreased when CO_2 is dissolved into the formation brine, resulting in higher security of the CO_2 injection process (Gilfillan et al., 2009; Hassanzadeh et al., 2009; Shafaei et al., 2012; Zendehboudi et al., 2011, 2012). CO_2 dissolution is also the dominant mechanism of CO_2 injection into shallow ocean locations (Saito and Kajishima, 2000; Haugan and Drange, 1992).

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0009-2509/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.ces.2013.08.056

Many techniques have been developed and studied to accelerate CO_2 dissolution (Hassanzadeh et al., 2009; Shafaei et al., 2012; Burton and Bryant, 2009; Zendehboudi et al., 2011, 2012; Leonenko and Keith, 2008; Bergmo et al., 2011; Saito et al., 1999, 2000, 2004, 2005; Kosugi et al., 2001; Niwa et al., 2005; Kajishima et al., 1995; Kajishima and Saito, 1997). In most of these processes, the major equipment in the injection system is exposed to corrosive CO_2–water mixtures, greatly increasing the capital and operational costs of the storage process.

The application of static mixers has recently been proposed as the mixing and mass transfer device to mix CO_2 and water on the surface (Eke et al., 2011) and for downhole dissolution (Zirrahi et al., 2013) and ocean submersion (Tajima et al., 2004, 2005, 2006, 2010). Using the static mixer not only reduces the exposure of surface and subsurface injection facilities to the corrosive mixture of CO_2 and brine, but also provides an efficient mixing and mass transfer process. Therefore, the investigation of the mass transfer and hydrodynamics of the static mixer is necessary, in order to obtain the optimal engineering design for the CO_2 dissolution process.

There are several studies on the mass transfer and hydrodynamics of the static mixers in the literature (Tizaoui and Zhang, 2010; Taweel and Walker, 1983; Cavatorta et al., 1999; Gotoa and Gaspillo, 1992; Song and Han, 2005; Heyouni et al., 2002; Turunen and Haario, 1994). Recently, Blyton and Bryant (2013) reported experiments to study the effect of temperature, CO_2 density and water salinity on the mass transfer of CO_2 into brine. However, there is still no accurate model for the mass transfer of supercritical CO_2 in water for CO_2 injection under subsurface conditions using static mixers. In this work, we apply a back flow cell model (BFCM) to model CO_2 mass transfer in water at geological storage temperature and pressure conditions. The BFCM is an accurate model dating back to the 1960s, which has recently gained interest for the modeling of mass transfer phenomenon in static mixers (Tizaoui and Zhang, 2010).

We have carried out experimental measurements of the residence time distribution (RTD) of the static mixer to find the required parameters for application of the BFCM. After calibration of the BFCM, the results were compared with the experimental data of a CO_2–water system at geological storage conditions.

The rest of this paper is organized as follows. First, we describe the BFCM for the modeling of supercritical CO_2 mass transfer in water. We then present the details of our experiments. Results and discussion are presented, followed by conclusions.

2. Model development

The BFCM is a modification of an ordinary tanks-in-series model (TSM) and considers a recirculation or back flow of a fraction of the feed flow rate, α, from each cell back to the preceding cell. The variation of α from zero to infinity results in the performance change of the BFCM from an ordinary tanks-in-series model to a single mixed tank (Roemer and Durbin, 1967; Tizaoui and Zhang, 2010). Fig. 1 shows the schematic diagram of the BFCM for the static mixer. The first (zero) and last $(N+1)$ cells represent image cells with negligible gas and liquid holdup (Roemer and Durbin, 1967).

The composition of the CO_2 phase could be assumed to be constant, because the water content of supercritical CO_2 at the conditions of interest is below 1% (Zirrahi et al., 2012). The back flow mixing of the gas phase was neglected (Tizaoui and Zhang, 2010), and the volumetric mass transfer coefficient, k_la, was assumed to be constant along the static mixer.

Writing a mass balance for the liquid phase under steady-state conditions results in the following set of equations:

$$i=0 \quad (1+\alpha)x_0-\alpha x_1=x_{in} \tag{1}$$

$$1\le i\le N \quad (1+\alpha)x_{i-1}-\left(1+2\alpha+k_la\frac{V_c}{L}\right)x_i+\alpha x_{i+1}=-k_la\frac{V_c}{L}x_i^* \tag{2}$$

$$i=N+1 \quad x_{N+1}=x_N=x_{out} \tag{3}$$

where x is the mole fraction of CO_2 in the aqueous phase; V_C and L denote the volume of each cell and the injected water volumetric flow rate, respectively; and x^* shows the equilibrium mole fraction of CO_2 in water at the experimental temperatures and pressures.

The volumetric mass transfer coefficient, k_la with dimension of (1/s), is obtained using (Tizaoui and Zhang, 2010):

$$k_la=\frac{L}{V_L}\frac{x_{in}-x_{out}}{\Delta x_{lm}} \tag{4}$$

where V_L is the volume of the liquid phase, and Δx_{lm} is the logarithmic mole fraction difference as follows:

$$\Delta x_{lm}=\frac{(x_{in}^*-x_{in})-(x_{out}^*-x_{out})}{\ln\left(\frac{x_{in}^*-x_{in}}{x_{out}^*-x_{out}}\right)} \tag{5}$$

The remaining parameters for the application of the BFCM are the number of cells, N, and the back flow ratio, α. Roemer and Durbin (1967) reported the analytical solution of the step and impulse response of the BFCM subject to an inlet tracer input. N and α for the BFCM of the static mixer can be calculated by matching the experimental response of the static mixer to a step or impulse inlet tracer input with the Roemer and Durbin (1967) model. According to the Roemer and Durbin model, the response of a step input is obtained as follows:

$$F(\theta)=1+\sum_{i=1}^{N}\left(\frac{A_i}{s_i}\right)\exp(s_i\theta) \tag{6}$$

where θ is the dimensionless time defined as t/τ, where τ is the residence time defined as $\tau=V_L/L$.

In Eq. (6), A_i and s_i are calculated as follows:

$$A_i=(-2N\gamma^{-N/2})\left(\frac{\sin^2(\varphi_i)}{D'(\varphi_i)}\right) \tag{7}$$

$$s_i=\left(\frac{N}{1-\gamma}\right)[2\gamma^{0.5}\cos(\varphi_i)-(1+\gamma)] \tag{8}$$

$$D(\varphi)=\gamma^{-0.5}\sin[(N+1)\varphi]-2\sin(N\varphi)+\gamma^{0.5}\sin[(N-1)\varphi] \tag{9}$$

where φ is the root of Eq. (9) in the interval of $0<\varphi<\pi$, and $D'(\varphi)$ denotes the derivative of D at φ; γ is the total flow ratio defined as $\alpha/(1+\alpha)$.

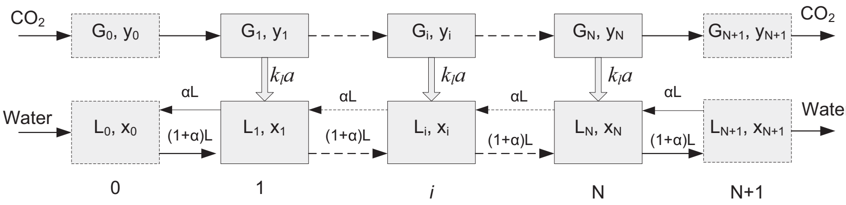

Fig. 1. Schematic diagram of the BFCM for the static mixer.