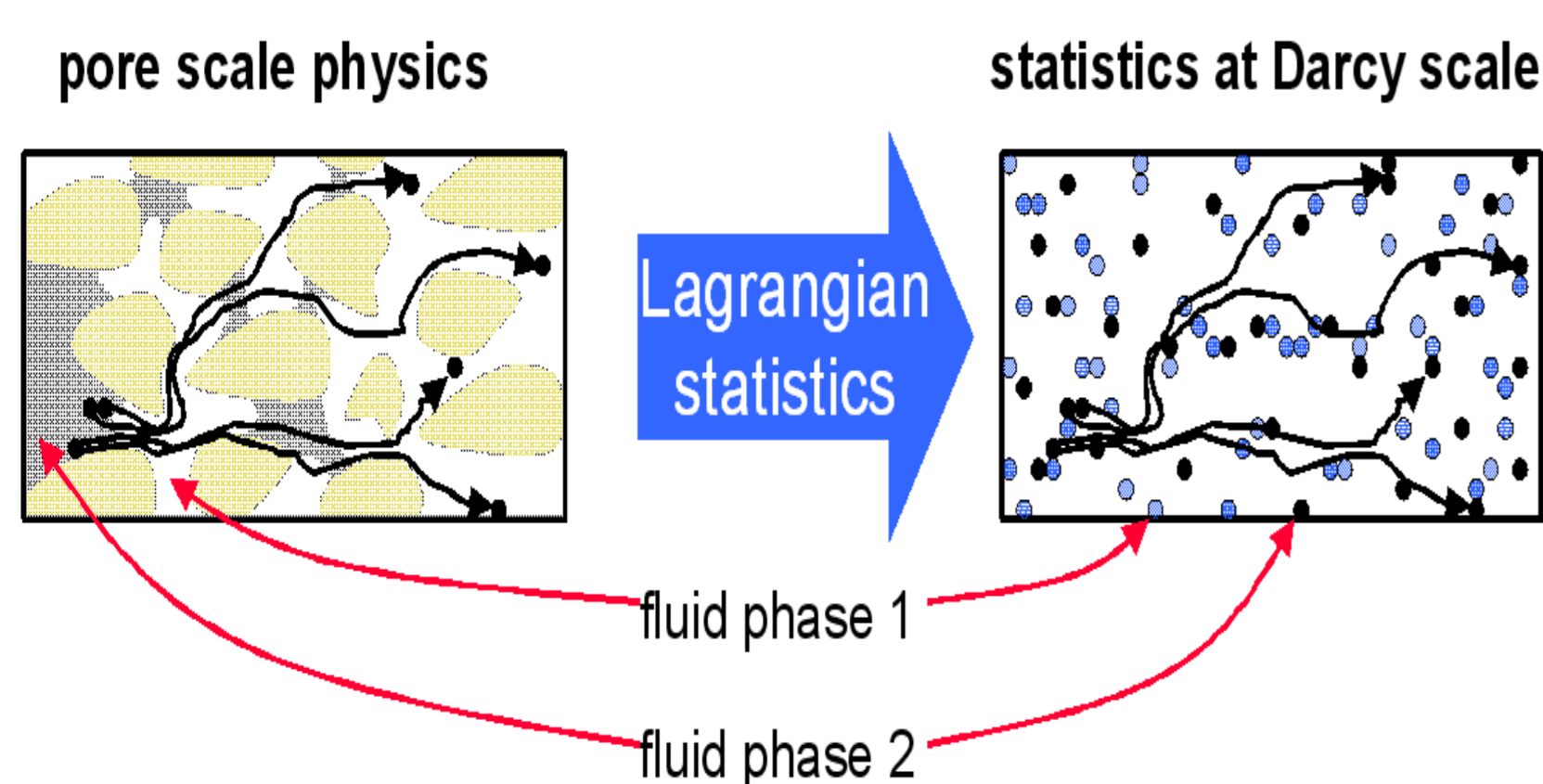


# PDF Framework for Modeling Non-Equilibrium Flows during CO<sub>2</sub> Storage

## 1. Introduction

Many of the complex physical processes relevant for compositional multi-phase flow in porous media are well understood at the pore-scale level. In order to study CO<sub>2</sub> storage in subsurface formations, however, it is not feasible to perform simulations at these small scales directly and effective models for multi-phase flow description at Darcy scale are needed. Unfortunately, in many cases it is not clear how the micro-scale knowledge can rigorously be translated into consistent macroscopic equations. Here, we present a new methodology, which provides a link between Lagrangian statistics of phase particle evolution and Darcy scale dynamics. Unlike in finite-volume methods, the evolution of Lagrangian particles representing small fluid phase volumes is modeled. Each particle has a state vector consisting of its position, velocity, fluid phase information and possibly other properties like phase composition. While the particles are transported through the computational domain according to their individual velocities, the properties are modeled via-stochastic processes honoring specified Lagrangian statistics.



## 2. Stochastic Particle Method (SPM)

Consider a stochastic particle representing a fluid volume of a phase having velocity  $\mathbf{u}^*$ , mobility  $\lambda^*$  and density  $\rho^*$ . The dynamics of particle is governed by the following Darcy like equation:

$$\mathbf{u}^* = -\frac{\lambda^*}{\phi}(\nabla p + \rho^* g \mathbf{e}_z), \quad (1)$$

where  $p$  is the average pressure. Each particle belongs to a phase, which is denoted by the random variable  $a^* \in \{1, 2, \dots, n\}$ . If  $m^*$  and  $v^*$  be the particle mass and particle volume, then, the phase saturation and density are defined as

$$S_\alpha = \frac{\langle v^* \delta(a^* - \alpha) \rangle}{\langle v^* \rangle} \quad \rho_\alpha = \frac{\langle m^* \delta(a^* - \alpha) \rangle}{\langle v^* \delta(a^* - \alpha) \rangle}$$

The particles evolve in physical space according to the following stochastic differential equation

$$d\mathbf{x} = \mathbf{u}^* dt + \sqrt{2D} d\mathbf{W} + \nabla D dt, \quad (2)$$

where  $d\mathbf{W}$  is the Wiener process and  $D$  the dispersion coefficient. By applying the average mass conservation the following pressure equation is obtained

$$\nabla \cdot \left( \sum_\alpha \rho_\alpha S_\alpha (\lambda^* \rho^*)|_{a^*=\alpha} \nabla p \right) = \frac{\partial \sum_\alpha \rho_\alpha S_\alpha}{\partial t} - g \mathbf{e}_z \cdot \nabla \sum_\alpha \rho_\alpha S_\alpha (\lambda^* \rho^*)|_{a^*=\alpha}, \quad (3)$$

where the Favre average of a quantity  $\psi$  conditioned on  $a^* = \alpha$  is defined as

$$\overline{\psi^*|_{a^*=\alpha}} = \frac{\langle m^* \delta(a^* - \alpha) \psi^* \rangle}{\langle m^* \delta(a^* - \alpha) \rangle}. \quad (4)$$

## 3. 2D Validation

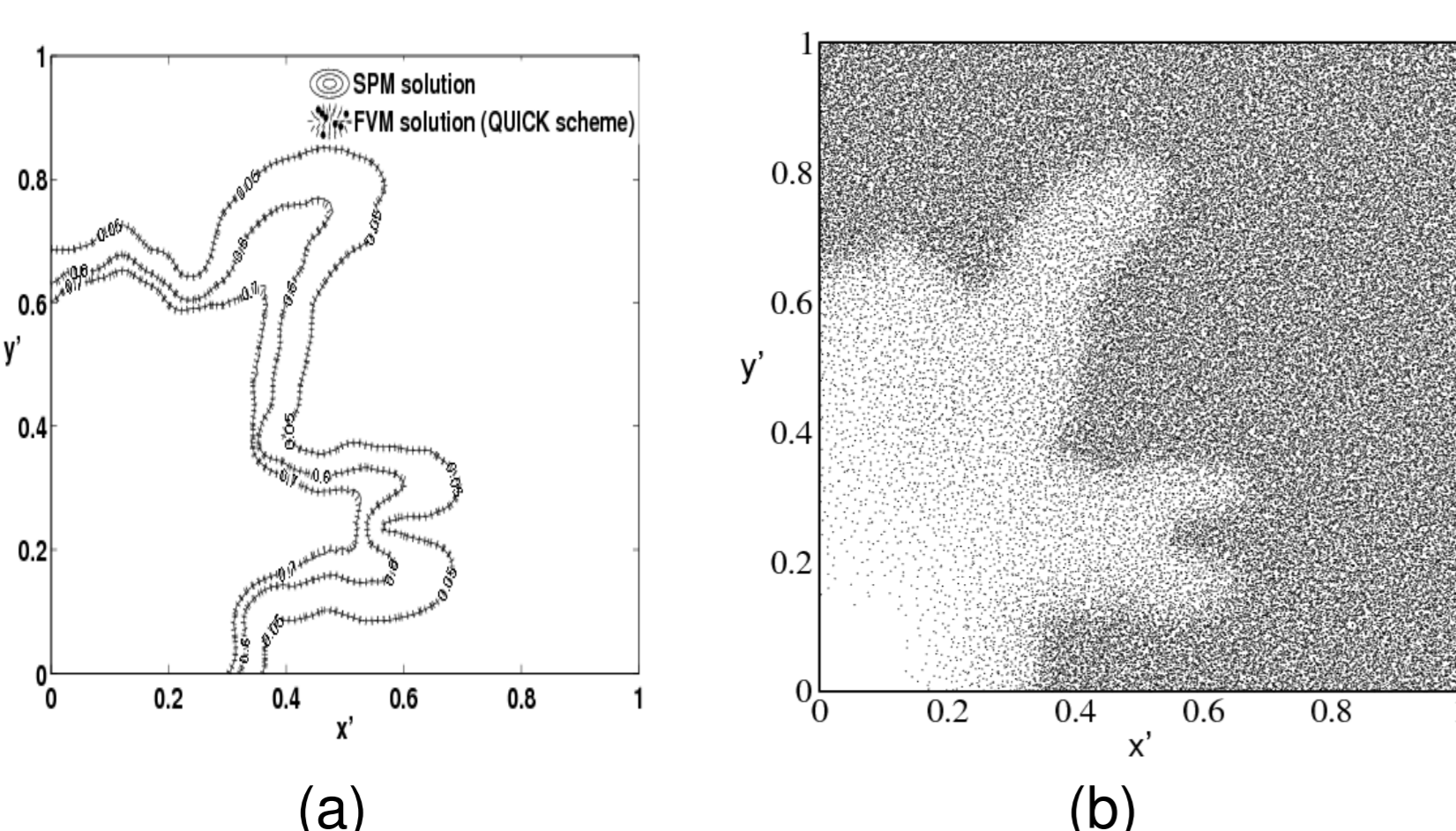


Figure 2: Simulation results of quarter-five-spot problem in heterogeneous medium (a): injected phase saturation contours; (b): injected phase particle distribution.

## 4. Stochastic Model for Dissolution and Mixing in Two-Phase Flow

Now we consider two-phase flow of brine ( $a^* = 1$ ) and CO<sub>2</sub> ( $a^* = 2$ ) with CO<sub>2</sub> dissolving into the brine phase. Let  $c^*$  be the concentration of dissolved CO<sub>2</sub> in brine, then its evolution is governed by the following non-equilibrium kinetic equation

$$\frac{dc^*}{dt} = -\underbrace{\frac{1}{\tau_d}(c^* - c^{eq})}_{\text{dissolution}} - \underbrace{\frac{1}{\tau_m}(c^* - \overline{c^*|_{a^*=1}})}_{\text{mixing}}, \quad (5)$$

where  $c^{eq}$ ,  $\tau_d$  and  $\tau_m$  are the equilibrium concentration, dissolution time and mixing time, respectively. The brine density is assumed to vary linearly with the dissolved CO<sub>2</sub> concentration, i.e.

$$\rho^*|_{a^*=1} = \rho_1 + \Delta \rho c^* \quad (6)$$

The brine density variation due to the CO<sub>2</sub> dissolution gives rise to gravity currents in the brine phase and introduces long correlation time in the flow dynamics. In order to simplify the problem, we employ the Bussinesq approximation, such that the effect of brine density variation is considered only in the brine velocity, otherwise the flow is incompressible. Let  $f(c; \mathbf{x}, t)$  be the brine phase mass density function (MDF) for the random variable  $\hat{c}$  in the concentration space  $c$ . Its evolution is governed by the following high-dimensional PDE

$$\frac{\partial f}{\partial t} + \nabla \cdot \left\{ \left\langle \frac{d\hat{\mathbf{x}}}{dt} | c; \mathbf{x}, t \right\rangle f \right\} + \frac{\partial}{\partial c} \left\{ \left\langle \frac{d\hat{c}}{dt} | c; \mathbf{x}, t \right\rangle f \right\} = 0 \quad (7)$$

The MDF has the following properties

$$\int f dc = \phi S_1, \quad \int f c dc = \phi S_1 \overline{\hat{c}|_{a=1}}, \quad \dots \quad (8)$$

The evolution equations for the average quantities can be obtained by multiplying Eq. (7) with  $1, c, c^2, \dots$  and integrating over the  $c$ -space, i.e.

$$\phi \frac{\partial S_1}{\partial t} + \nabla \cdot \{ \lambda_1 S_1 (\nabla p + \overline{\hat{\rho}|_{a=1}} g \mathbf{e}_z) \} = 0. \quad (9)$$

$$\phi \frac{\partial \overline{\hat{c}|_{a=1}} S_1}{\partial t} + \nabla \cdot \{ \lambda_1 S_1 (\overline{\hat{c}|_{a=1}} \nabla p + \overline{\hat{\rho} \hat{c}|_{a=1}} g \mathbf{e}_z) \} + \frac{\phi S_1}{\tau_d} (\overline{\hat{c}|_{a=1}} - c^{eq}) = 0. \quad (10)$$

$$\phi \frac{\partial \overline{\hat{c}^2|_{a=1}} S_1}{\partial t} + \nabla \cdot \{ \lambda_1 S_1 (\overline{\hat{c}^2|_{a=1}} \nabla p + \overline{\hat{\rho} \hat{c}^2|_{a=1}} g \mathbf{e}_z) \} + \frac{2\phi S_1}{\tau_d} (\overline{\hat{c}^2|_{a=1}} - \overline{\hat{c}|_{a=1}} c^{eq}) + \frac{2\phi S_1}{\tau_m} (\overline{\hat{c}^2|_{a=1}} - \overline{\hat{c}|_{a=1}}^2) = 0. \quad (11)$$

## 5. Simulation Results of CO<sub>2</sub> Plume Rising in Brine

Here we present 2D simulation results of a CO<sub>2</sub> plume rising due to buoyancy in a brine aquifer. The geometrical details of the problem are given in Figure 3. In order to reduce the number of independent variables, we introduce the following buoyancy time scale  $\tau_g$  and the ratio  $\gamma$

$$\tau_g = \frac{\phi H \mu_1}{K(\rho_1 - \rho_2)g}, \quad \gamma = \frac{\Delta \rho c^{eq}}{\rho_1 - \rho_2}. \quad (12)$$

In the following results, the values of the flow parameters and variables are:  $\lambda_1 = S_1 K / \mu_1$ ,  $\lambda_2 = S_2 K / \mu_2$ ,  $\mu_2 / \mu_1 = 1$ ,  $L/H = 1$ ,  $\tau_d = 0.1 \tau_g$ ,  $\tau_m = 0$  and  $\gamma = 1$ .

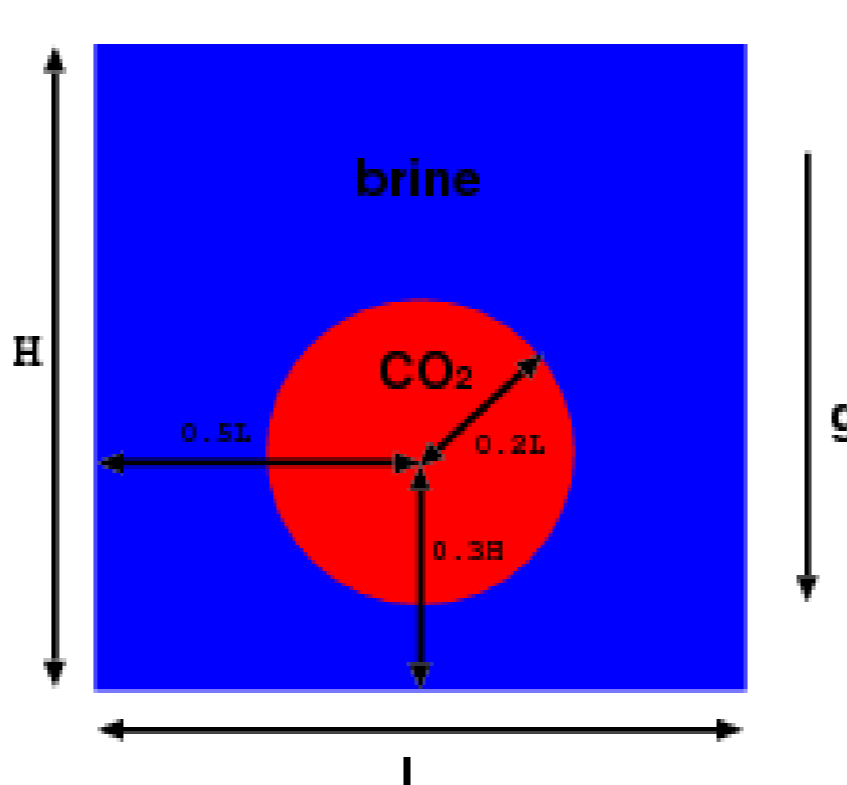


Figure 3: Initial configuration of CO<sub>2</sub> plume in brine.

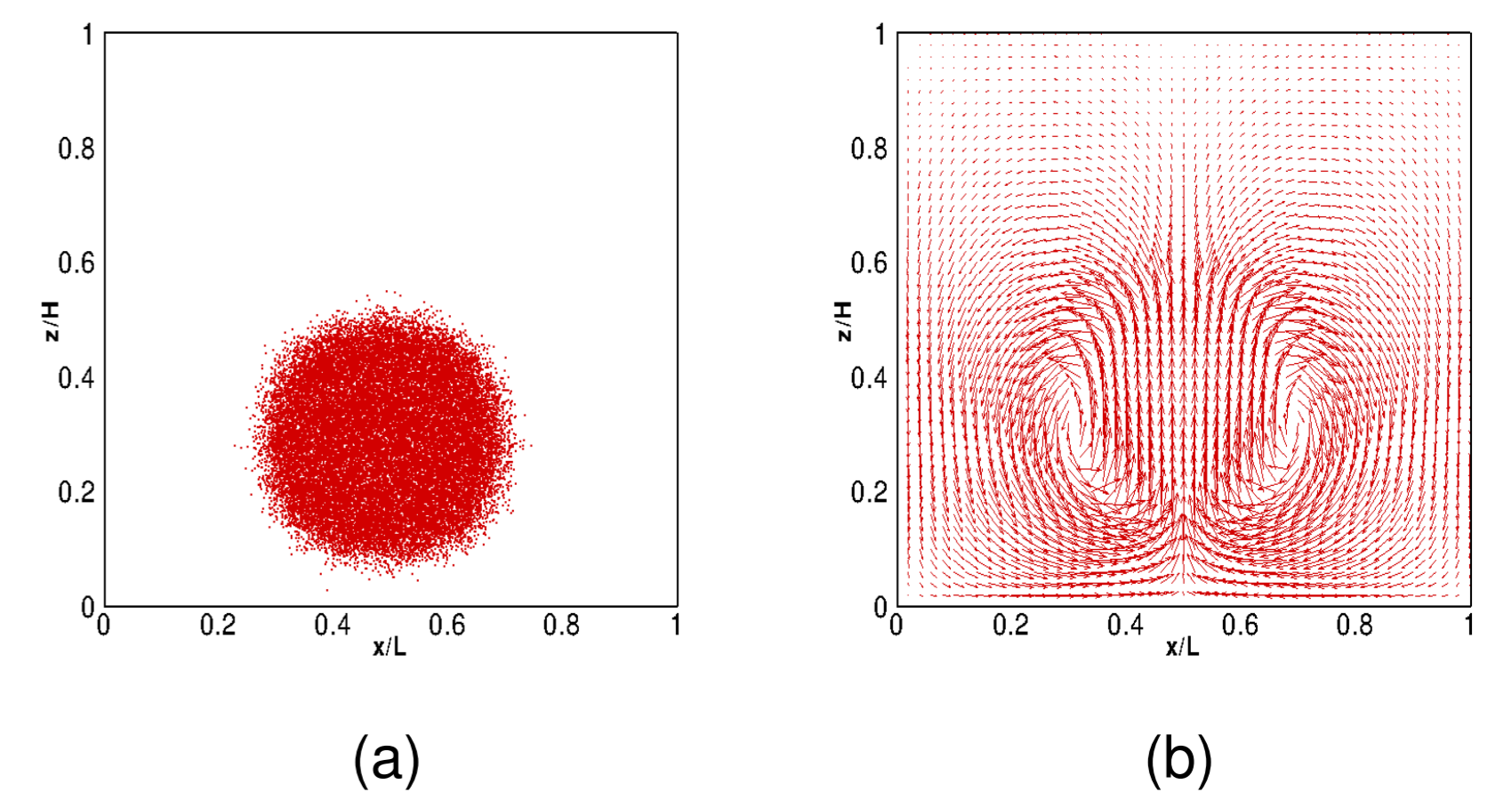


Figure 4: Simulation results at  $t/\tau_g = 0.05$  (a): CO<sub>2</sub> particles; (b): total flux.

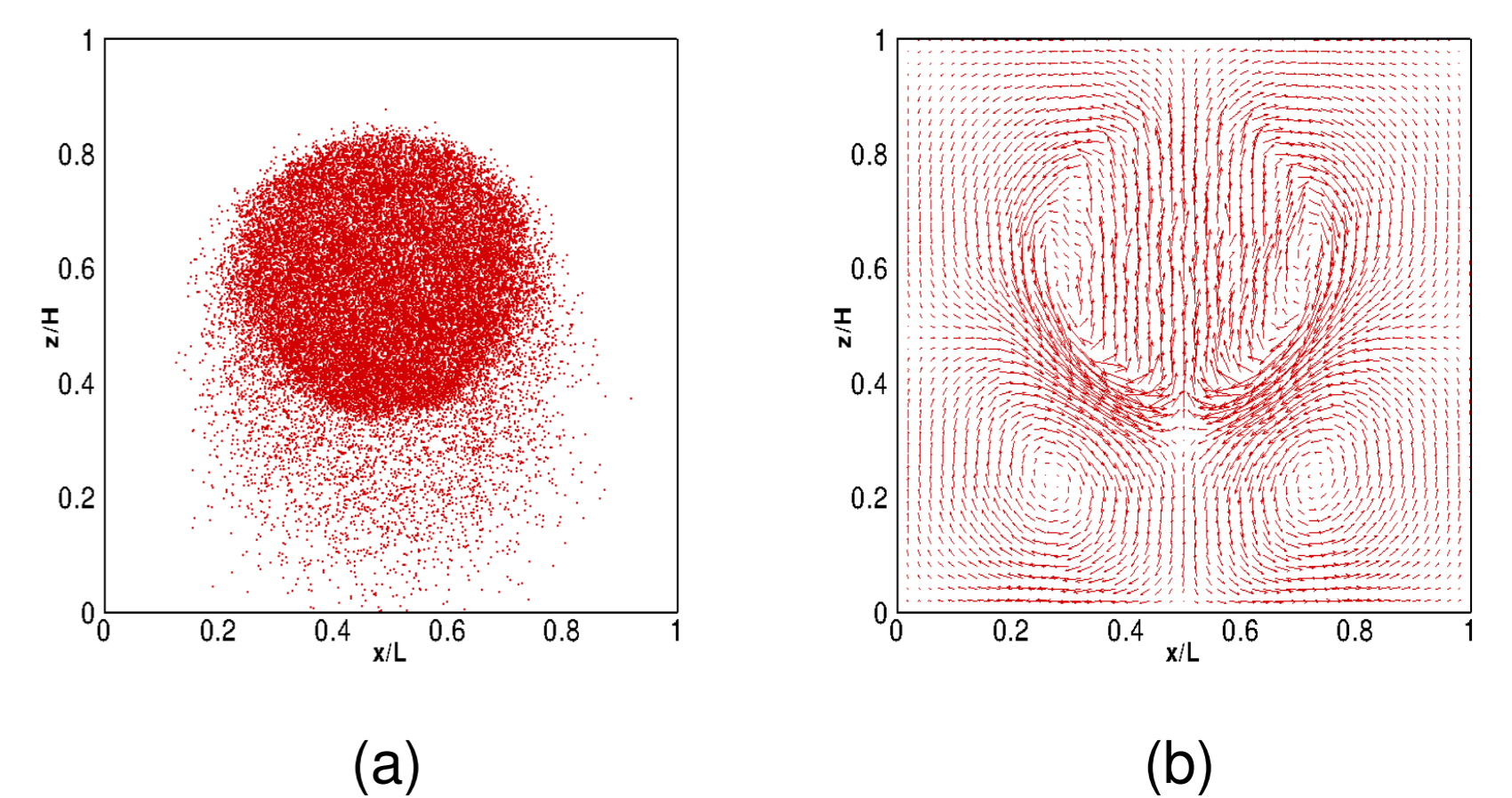


Figure 5: Simulation results at  $t/\tau_g = 0.75$  (a): CO<sub>2</sub> particles; (b): total flux.

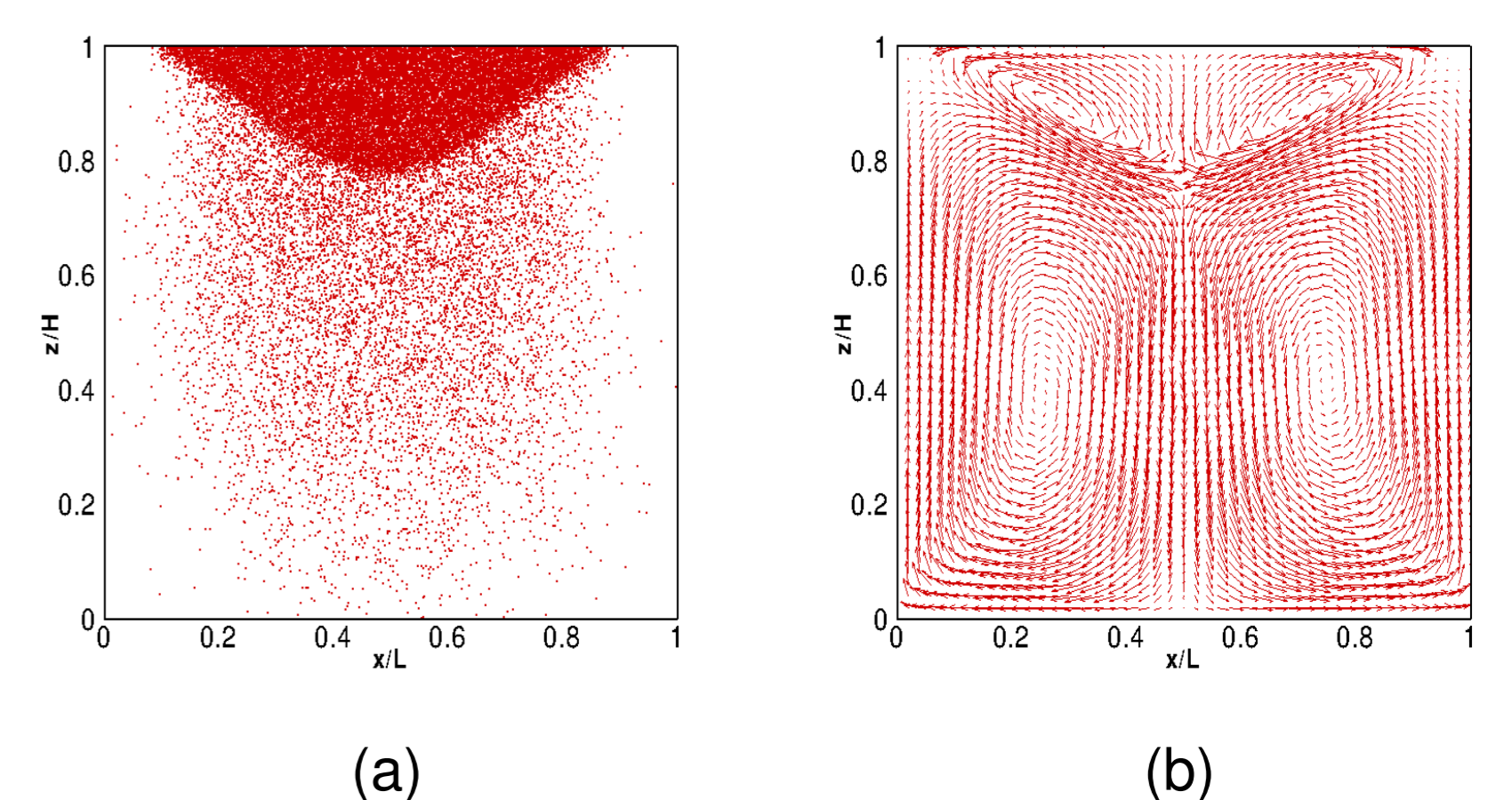


Figure 6: Simulation results at  $t/\tau_g = 1.75$  (a): CO<sub>2</sub> particles; (b): total flux.

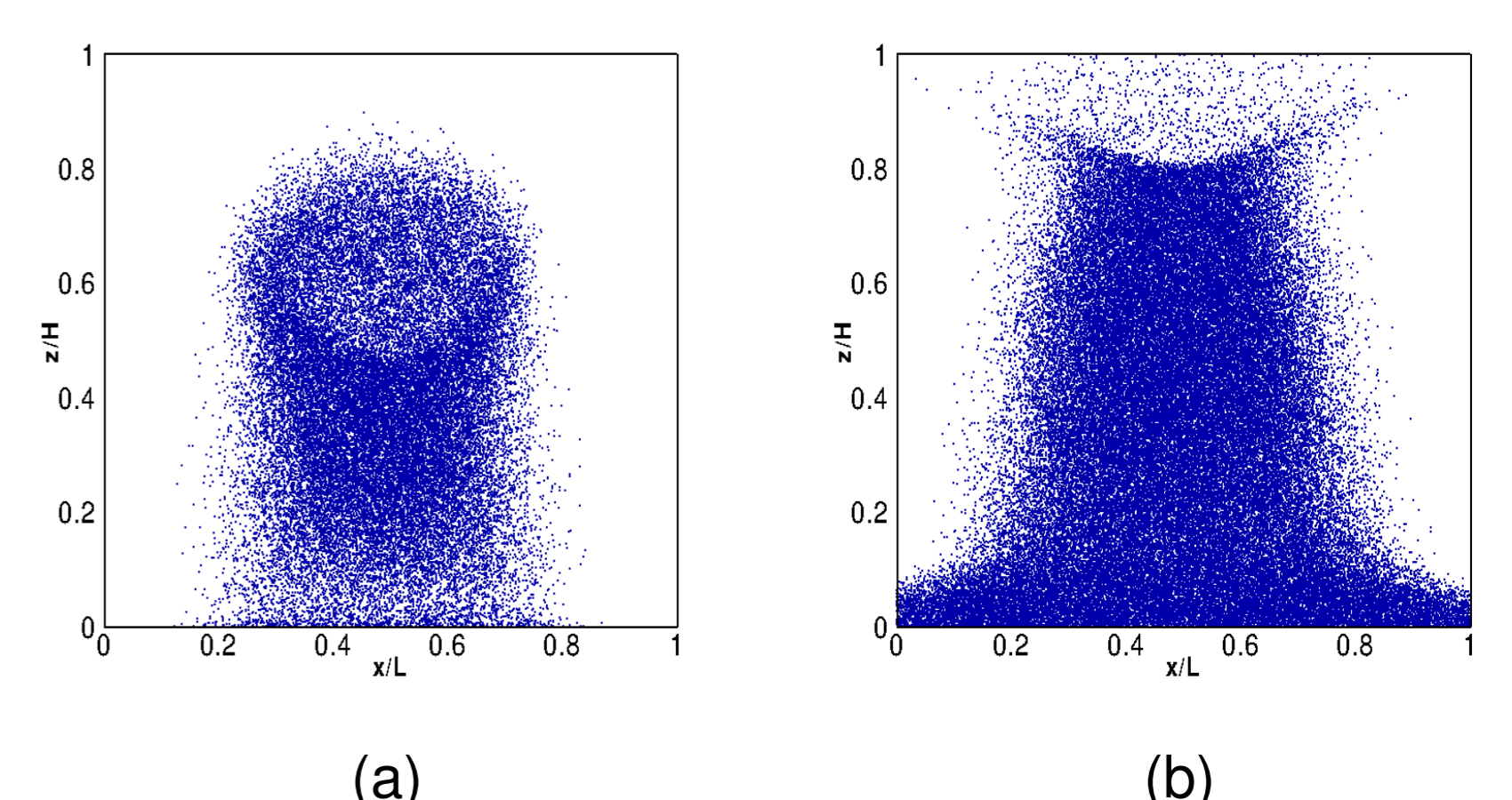


Figure 7: Brine phase particles with  $c/c^{eq} > 0.5$  at (a):  $t/\tau_g = 0.75$  and (b):  $t/\tau_g = 1.75$ .

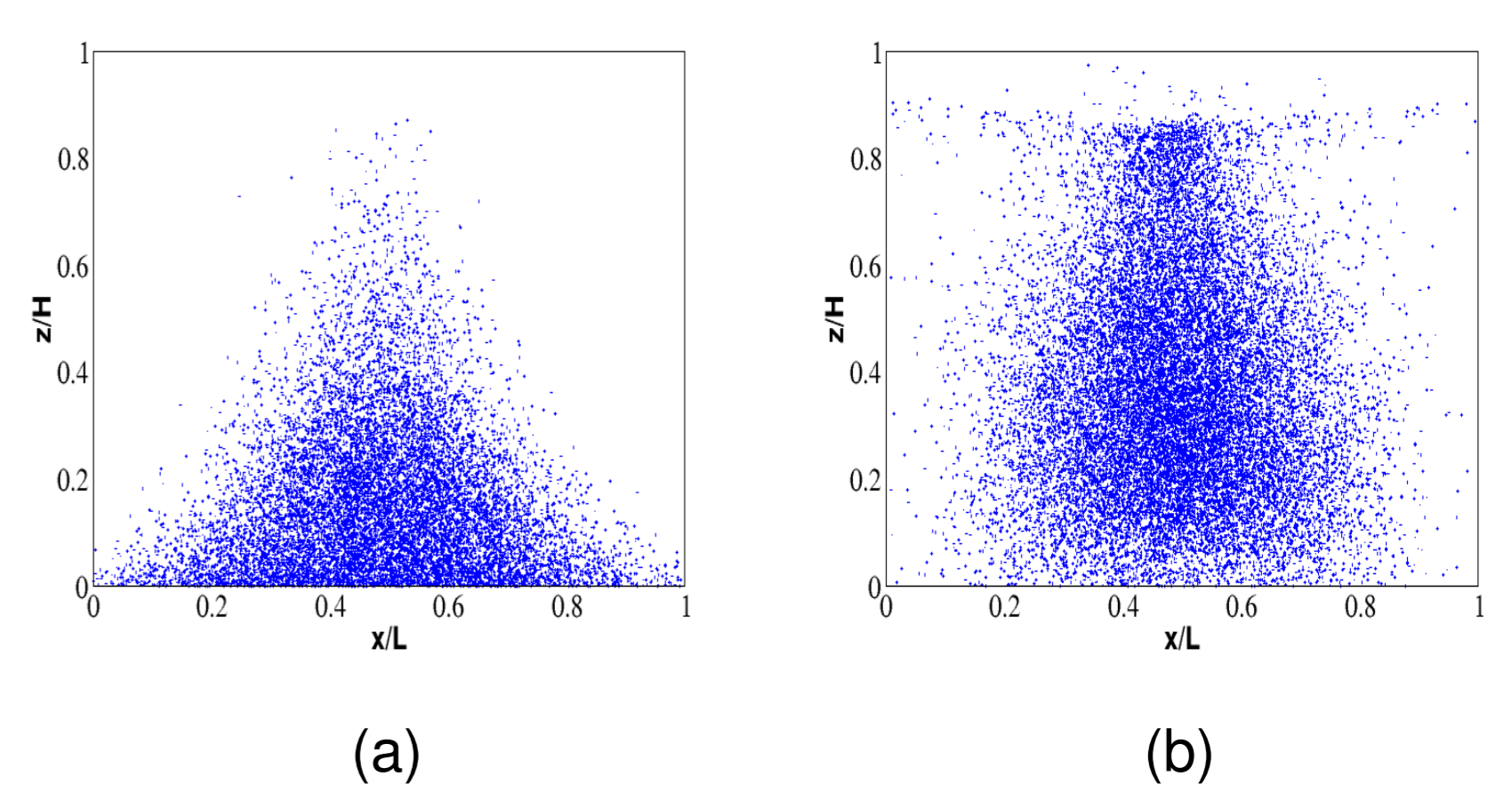


Figure 8: Brine phase particles with  $c/c^{eq} > 0.5$  and  $\tau_d = 0.5 \tau_g$  at  $t/\tau_g = 5$  (a): simulation with correlation and (b): simulation without correlation,  $\hat{\rho} = \langle \hat{\rho} |_{a=1} \rangle$ .