



# Linear perturbation analysis of density change caused by dissolution of carbon dioxide in saline aqueous phase



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## ABSTRACT

This paper presents a linear perturbation analysis of natural convection in porous media due to density change caused by dissolution of an injected carbon dioxide in saline aqueous phase. The main application is in geological sequestration of CO<sub>2</sub> in deep aquifer. In this study, both density and viscosity of the fluid in the medium are functions of the dissolved substance concentration.

Effects of concentration-dependent viscosity and density on the stability of the system are studied parametrically. By linearizing the system of equations, a set of governing equations for perturbed concentration and velocity is derived as a function of density and viscosity derivatives. Then, the eigenvalue problem is solved numerically to obtain growth rate of waves as a function of wave-number.

From a quantitative analysis on the onset of instability, it is found that the system is more stable when the viscosity increases more rapidly with concentration. In addition, parametric analysis of density–concentration relationship shows that the form of density–concentration relationship affects the prediction of onset of instability. In this way, concavity of density–concentration function decreases the weight of the top layer and lowers the destabilizing effect of the layer. These findings provide a more realistic outlook to recognize the onset of convection and long-term fate of disposed gas in large-scale geological sequestration of CO<sub>2</sub>.

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## 1. Introduction

Natural convection in porous media is a type of density driven flows. This may occur when the fluid on top becomes denser than the rest of the fluid. Two factors which may cause the density change are variation in temperature and composition of the fluid. Thermal natural convection in porous media can be observed vastly in industrial and geological applications.

For a concise survey, the reader can refer to the book of Nield and Bejan [1]. On the other hand, when the composition of a stagnant fluid in porous media changes due to dissolution of a secondary substance, the situation may become unstable and convection may take place. This phenomenon mainly appears in the geological cases.

The present work is focused on cases in which the secondary material on the top is in contact with the fluid in porous media and dissolves due to the diffusion process. If the rich mixture at the top becomes denser than the fresh fluid, it tends to flow downward. Then the instability in the fluid may occur. The onset of convection in this problem is mostly described as a function of non-dimensional group Rayleigh number.

The well-known example of the abovementioned phenomenon is geological sequestration of CO<sub>2</sub> in deep aquifers. Deep aquifers are found as natural field with large capacity for gas storage as a carbon capture and sequestration (CCS) project [2–4].

Dissolution of CO<sub>2</sub> into aquifers and subsequent downward diffusion increases water density at the top of aquifer, which in turn results in a density gradient. As a consequence, convective mixing occurs and dissolution of CO<sub>2</sub> into aquifer is accelerated significantly [5–7]. Convective mixing enhances the rate of dissolution as compared to diffusion by distribution of CO<sub>2</sub> through the whole aquifer volume. This effect reduces the storage time and prevents CO<sub>2</sub> from leaking out of reservoir after injection [6,8,9].

Hydrodynamic instability phenomenon under different conditions in porous media has been studied extensively. Researchers have often studied this phenomenon by focusing on analytical methods. Linear amplification theory, non-linear amplitude method and the energy method have been used for characterizing time-dependent concentration [10] or temperature profiles [11]. Also, amplification theory for predicting the onset of buoyancy-driven flow in porous media has been used in [12–14]. In addition, this method has been applied to investigate the role of anisotropy on the onset of convection [15,16] and to find a scaling relationship for onset of natural convection based on the dominant mode of self-similar diffusion operator and numerical

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simulations [17]. The methodical description and comparison between different approaches to stability theory may be found in Rees et al. [18].

In literature, normally, two assumptions are taken; the viscosity of the fluid is taken to be constant and the variation of the fluid's density due to concentration of dissolved substance (or temperature) assumed to be linear.

Dependency of viscosity to solute concentration can affect the flow pattern for example, reported experimental data [19,20] for changes in water viscosity with dissolved CO<sub>2</sub> show that dissolved CO<sub>2</sub> has a significant impact (up to 13%) on brine viscosity and it may influence the flow regime and onset of instability during CO<sub>2</sub> sequestration. Recently, Meulenbroek et al. [21] examined the effect of variation of brine viscosity due to dissolved CO<sub>2</sub> and showed its effect on onset of instability when viscosity changes linearly with CO<sub>2</sub> concentration by two different slopes. Also, in a similar application, Kim [13] predicted that the onset of buoyancy driven natural convection in carbon dioxide (CO<sub>2</sub>) improved oil recovery assuming that the viscosity of the solution is an exponential function of concentration.

The abovementioned phenomenon should not to be confused with the transient thermal convection of fluid cooled from above. This latter flow is also unstable at high Rayleigh numbers and is vastly studied in the literature, [22–25] with application in the flow in Earth's mantle. Nevertheless, the above concepts and Navier–Stokes equations are not applicable for fluid flow in porous media such as aquifers. Rather, it is necessary to use Darcy's Law or its modifications as the momentum equation. Comparison of scaling relationships for onset of buoyancy driven flow in porous media [8,26–28] with those for bulk fluid [25,29,30] asserts the fact that implementation of different momentum equations leads to different final relationships between the onset of convection and Rayleigh number. This difference is also revealed in the different definitions of Rayleigh number for these two systems.

In the literature, commonly, Boussinesq approximation has been used to simplify the continuity equation, where the coefficient of volume expansion is considered very small, so density was considered constant for all terms in equations of motion, except for one in the body force. Also, previous studies supposed mixture density as a linear function of solute concentration. However, in general, the density of the fluid is not a linear function (for instance see [31,32] for the aqueous solutions of CO<sub>2</sub>).

In this paper, the effect of variation of fluid's density and viscosity on the onset of natural convection in porous media is studied using a quasi-steady state stability analysis. Here, a quadratic density function is applied in Boussinesq approximation and the function's influence is examined using the approach adopted by researchers [33–38]. In this study, it is assumed that the viscosity of the fluid increases linearly with concentration of the dissolved substance. It has been shown in the literature that this effect significantly increases the onset time of instability in the case of CO<sub>2</sub> sequestration in aquifer [21,36–38]. In addition, the effect of viscosity variation is derived in the form of scaling relationship.

In the following, the governing equations and boundary conditions are introduced. Then, the linear stability analysis is applied on the problem and the results of the onset time of instability are discussed for different cases of variable fluid's properties.

## 2. Mathematical model

A two-dimensional, lateral infinite homogeneous and isotropic porous layer of thickness  $H$  saturated with a homogeneous fluid (say brine) is modeled. Symmetry condition at the lateral boundaries and no-flow at the bottom boundary are set for the model at all times. Schematic diagram of the model and its boundary conditions is shown in Fig. 1. The top boundary is in contact with another fluid. Due to thermodynamic equilibrium, it is assumed that the interface concentration of dissolved substance  $C^s$  is constant. Other assumptions made for this model are as follows:

1. Temperature gradient is negligible;
2. Velocity-based dispersion effect is negligible;
3. Geochemical reactions are ignored;
4. The flow is single-phase and incompressible;
5. The dissolved substance is single component.

### 2.1. Governing equations

As a result of diffusion and dissolution of dissolved substance, the saturated fluid at the top becomes denser than resident fluid in lower layers. This may lead to instability in the system under study. In order to determine how viscosity and density variations may affect stability of this system, the set of governing equations with proper initial and boundary conditions needs to be solved. After Boussinesq approximation, the governing equations of this system include equation of continuity, equation of motion (Darcy's equation) and the convection–diffusion equation for conservation of dissolved substance (Eqs. (1) to (3)):

$$\nabla \cdot \vec{V} = 0, \quad (1)$$

$$\vec{V} = -\frac{K}{\mu}(\nabla P - \rho g \nabla z), \quad (2)$$

$$D \nabla^2 C - \vec{V} \cdot \nabla C = \varnothing \frac{\partial C}{\partial t}. \quad (3)$$

Viscosity and density are generally functions of solute concentration, i.e.  $\mu = \mu(C)$  and  $\rho = \rho(C)$ . Generally, these functions are complicated and depend on the fluid and model characteristics.

Here, a linear stability analysis is performed to explore the impact of concentration dependency of fluid viscosity and density to the onset of instability and mixing mechanism. For convenience, a linear viscosity–concentration and quadratic density–concentration relationships were adopted to apply into perturbation equation form.

In the following sections, two parametric studies are presented to show how viscosity and density profile can affect the flow regimes as acid gas trapped into brine layers. Before that, solution of the base state should be obtained.

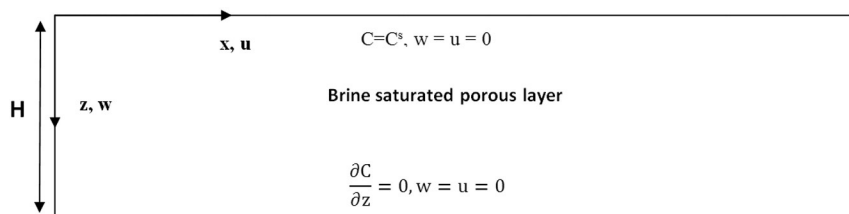


Fig. 1. Geometry and boundary conditions used in this study.

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