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Effect of cross-diffusion on the gravitational instability in a ternary mixture: Asymptotic and linear analyses

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HIGHLIGHTS

- Effects of cross diffusion on the onset of gravitational instability are analyzed theoretically.
- New stability limits are obtained by considering a non-monotonic density profile.
- Instabilities are possible without the density inversion.
- Stability conditions from the asymptotic and the linear stability analyses are in good agreement.

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ABSTRACT

To consider the effect of cross diffusion more rigorously, the onset and the growth of the gravitational instabilities in a Hele-Shaw cell saturated with ternary solution are analyzed by considering all cross diffusion coefficients. Through the asymptotic analysis, we identify the double-diffusive (DD)-, diffusive-layer convection (DLC)- and extended double diffusive (EDD)-type instability regimes. To support the asymptotic stability analysis, new linear stability equations are derived in the global domain and then transformed into the similar domain. In the similar domain, we prove that initially the system is unconditionally stable. For transient stability analysis, the linear stability equations are solved by employing quasi-steady state approximations (QSSA's). To avoid the limit of the conventional QSSAz, we obtain the critical time for the onset of instability motion using the QSSA in the similar domain (QSSAζ).

1. Introduction

Cross-diffusion, whereby a diffusive flux of a given species is driven by the concentration gradients of other species, is inevitable in multicomponent systems where unusual chemical interactions are involved. These unusual interactions are common in the systems of (i) solutes of very different size, (ii) solutes in a highly nonideal solution, (iii) concentrated electrolytes and (iv) concentrated alloys (Cussler, 2009). When cross-diffusion effects are combined with linear or nonlinear reactions, it plays an important role in both self-assembly and self-organization in many multicomponent systems (Vanag and Epstein, 2009). Recently, to understand the protein crystallization in various aqueous solutions, much experimental work has been conducted to determine the diffusion coefficients matrix in multicomponent systems (Capuano et al., 2011).

In the gravitational field, cross-diffusion effects can induce buoyancy-driven convective instabilities even in an initially stably-stratified system. In the polyvinylpyrrolidone (PVP)-dextran-water system, Laurent et al. (1979) observed that the diffusion rate of one macromolecule can be paradoxically enhanced by adding a sufficient amount of a suitable second macromolecular component (dextran). Later, in the PVP-dextran-water system, Preston et al. (1980) found that adding a second component in a polymer solution can induce ordered motion near the interface and therefore, can increase the transport rate of a polymer component. However, this interesting phenomenon was not observed in the sorbitol-dextran-water system. Later, in the $SrCl_2$-NaCl-H_2O system, Miller and Vitagliano (1986) experimentally found that the instabilities far from the interface (overstability) as well as near the interface (fingering) are possible.

Recently, a non-reactive sodium bis(2ethylhexyl)sulfosuccinate Aerosol OT(AOT) microemulsion(ME) system has attracted many researchers' interest in context with the onset of Turing instability in the Belousov-Zhabotinsky(BZ)-reaction dispersed in

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a water-in-oil AOT-ME (BZ-AOT) system (Vanag and Epstein, 2009; Rossi et al., 2010; Rossi et al., 2011). Also, much attention has been given to the ternary mixture of 1,2,3,4-tetrahydronaphthalene (THN), iso-butylbenzene (IBB) and n-dodecane (nC_{12}) (Larrañaga et al., 2014; Sechenyh et al., 2016; Pallares et al., 2017). This ternary mixture is used as a representative model of the different molecular families in the petroleum industry. Specifically, the polycyclic, aromatic and aliphatic compounds are associated with the THN, IBB and nC_{12}, respectively. For the AOT-ME in water system, Budroni et al. (2015a,b) experimentally found gravitational instability growing near the initial interface region symmetrically across the interface between two identical stratified AOT-ME in a Hele-Shaw cell. They showed that double diffusive (DD) or diffusive layer convection (DLC) hydrodynamic instabilities (Trevelyan et al., 2011) are possible by changing the initial composition of the MEs along the gravitational axis. Their instability scenario is similar to that observed in the PVP-dextran-water system (Preston et al., 1980). Also, Budroni (2015) analyzed the gravitational instability of an AOT-ME system using a non-linear numerical simulation. He showed that according to the sign of a cross-diffusion coefficient, two types of non-monotonic density profiles can be developed. If the density gradient at the interface is positive, a local maximum overlies a minimum across the initial interface. This non-monotonic density profile corresponds to double diffusive (DD) instability condition. He also showed that a diffusive layer convection (DLC)-type instability is possible if the density gradient at the interface is negative. Kim and Song (2016) conducted a systematic linear stability analysis for the AOT-ME system and corrected Budroni (2015) minor error. However, they assumed that one of the cross-diffusion coefficients, D_{12} is very small with respect to the normal diffusion coefficient, D_{11}. Very recently, taking all the cross-diffusion effects into account, Pallares et al. (2017) studied the onset of cross-diffusion driven gravitational instability theoretically and numerically. They showed that the instability is possible even in the stably-stratified system. However, Pallares et al. (2017) committed a minor error and an oversimplification, and therefore, more refined work is necessary to understand the cross-diffusion effects on the buoyancy-driven convection in multi-component systems.

In the present study, we analyze the effect of a cross-diffusion on the onset and the growth of gravitational instability of a miscible interface. Here, we will extend Budroni (2015) and Kim and Song (2016) analyses where one of the cross-diffusion coefficients, D_{12}, was neglected, i.e., we will consider the effect of cross-diffusion more rigorously. In addition, we will identify three different types of an unstable density profile and suggest the asymptotic stability without a detailed stability analysis. Then, in the similar domain, the new stability equations are to be derived and solved with the quasi-steady state approximation (QSSA). Based on the present asymptotic and linear analyses, we will clearly show that in multicomponent systems, gravitational instability is possible without the adverse density gradient. This interesting feature was not considered by Budroni (2015) and Kim and Song (2016). Because we correct some mathematical errors, generalize the previous work and consider a new instability regime which has been ignored, the present work provides a robust theoretical basis to understand gravitational instability in multi-component systems.

2. Governing equations and base states

Recently, Pallares et al. (2017) conducted stability analysis for the fluid system in Mialdun et al. (2013) counter flow cell (CFC) to determine the diffusivities of the THN, IBB and nC_{12} mixture. In addition, Budroni et al. (2015a,b) conducted systematic experiences on the onset of gravitational instability in a Hele-Shaw cell where two ternary miscible solutions are placed in contact inside a vertically orientated configuration such as that schematized in Fig. 1. The initial weight fraction of component "i" are $C_{i,L}$ and $C_{i,U}$, here "U" and "L" represent the upper and the lower layer conditions. If $\rho_L > \rho_U$, the system is initially gravitationally stable, where ρ is density. Under the Hele-Shaw approximation, i.e., $h \ll (d, H)$, the governing equations for the ternary system can be written as (Pallares et al., 2017; Kim and Song, 2016)

$$\nabla \cdot \mathbf{U} = 0, \tag{1}$$

$$\frac{\mu}{K}\mathbf{U} = -\nabla P + \rho\mathbf{g}, \tag{2}$$

$$\frac{\partial C_1}{\partial t} + \mathbf{U} \cdot \nabla C_1 = D_{11}\nabla^2 C_1 + D_{12}\nabla^2 C_2, \tag{3}$$

$$\frac{\partial C_2}{\partial t} + \mathbf{U} \cdot \nabla C_2 = D_{22}\nabla^2 C_2 + D_{21}\nabla^2 C_1. \tag{4}$$

Here, $\mathbf{U}$ is the velocity vector, and Eqs. (1) and (2) are the continuity equation and the Darcy's law to describe the motion of Newtonian fluid in a porous medium and in a Hele-Shaw cell, respectively. Pallares et al. (2017) employed the Navier-Stokes equation rather than the Darcy's law in their analysis. In a Hele-Shaw cell, μ is the viscosity of the fluid, $K(= h^2/12)$ is the permeability in the Darcy's law, and D_{12} and D_{21} are the cross diffusion coefficients. Using the relation of $\sum_{i=1}^{3} C_i = 1$, the mass transfer equation for the third dependent component, i.e., the common solvent, can be written as

$$\frac{\partial C_3}{\partial t} + \mathbf{U} \cdot \nabla C_3 = -(D_{11} + D_{21})\nabla^2 C_1 - (D_{12} + D_{22})\nabla^2 C_2. \tag{5}$$

Thermodynamics imposed the following constraints on the diffusion coefficients, in order to have a stable diffusion process (Taylor and Krishna, 1993):

$$D_{11} > 0 \text{ and } D_{22} > 0, \tag{6a}$$

$$D_{11}D_{22} - D_{12}D_{21} > 0, \tag{6b}$$

and

$$(D_{11} - D_{22})^2 + 4D_{12}D_{21} \geqslant 0. \tag{6c}$$

The above thermodynamic constraints (6b) and (6c) are naturally satisfied in Budroni (2015) and Kim and Song (2016) analyses,

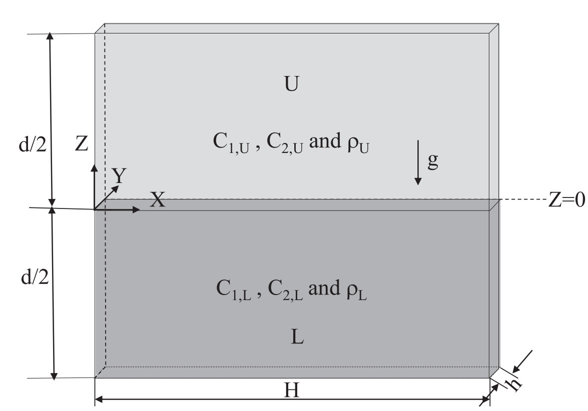

Fig. 1. Schematic diagram of the initial distribution of solutes A and B. Z = 0 represents the initial contact plane. In the case of $\rho_L > \rho_U$, the present initially stably-stratified system can be unstable due to the effect of multicomponent diffusions. The Hele-Shaw approximation can be introduced in the case of $h \ll (d, H)$.