THEORY AND SIMULATION OF THE ONSETS OF CONVECTION INDUCED BY LIQUID DIFFUSION IN LIQUID AND INCipient INSTABILITY CAUSED BY LIQUID FLOW PARTICLE BED

SIEW FONG WAH.

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By

SIEW FONG WAH

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia,
in Fulfilment of the Requirements for the Degree of Master of Science

March 2006
To Dad and Mom
Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

THEORY AND SIMULATION OF THE ONSETS OF CONVECTION INDUCED BY LIQUID DIFFUSION IN LIQUID AND INCIPIENT INSTABILITY CAUSED BY LIQUID FLOW IN PARTICLE BED

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March 2006

Chairman: Associate Professor Thomas Choong Shean Yaw, PhD

Faculty: Engineering

The theory of transient instability of gas in aqueous liquid proposed by Tan and Thorpe (1999) was extended to study the onset of convection induced by liquid diffusion in liquid. 2D time-dependent simulations were conducted using a CFD package – FLUENT to verify the analysis of transient instability. The onset of buoyancy convection induced by liquid mass diffusion was simulated for water diffused into 1-butanol, ethyl acetate and 2-butanone at different temperatures. The formation and the development of the convection plumes were successfully simulated. The transient liquid diffusion simulation was verified using Fick’s Law. The average simulated transient Rayleigh number was found to be 1315, which was close to 1404, i.e. the average value of critical Rayleigh number of 1100 and 1708 for boundary conditions with free-solid and solid-solid surfaces. The simulated onset time, the critical diffusion depth and the size of the plumes were found to agree well with values predicted by theory.
By recognizing that the mass transfer effect is very much smaller compared to the effect of momentum diffusion in causing the incipient instability in particle bed, the transient critical Rayleigh number proposed by Tan (2004) was re-defined based on momentum diffusion. The time-dependent simulations for incipient instability caused by liquid flow in particle bed were performed for different particle sizes, flow rates and initial bed heights. The CFD simulations were verified by the variation of particle bed height with superficial velocity. The onset time of incipient instability could be determined by the series contours of volume fraction, velocity vectors and maximum velocity. The simulated critical Rayleigh numbers based on the momentum diffusion were found to be inconsistent. A modified critical Rayleigh number was proposed by incorporating a factor of $z_c/H$ to the critical Rayleigh number. The simulated modified critical Rayleigh number was found to be almost constant, $Ra_c' = 16.8$, which was very close to the theoretical value of 17.7 for buoyancy instability for porous media.
Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

TEORI DAN SIMULASI UNTUK PERMULAAN PEROLOKAN DISEBABKAN OLEH RESAPAN CECAIR DALAM CECAIR DAN INSPIEN KETIDAKSTABILAN DISEBABKAN OLEH ALIRAN CECAIR DALAM LAPISAN BUTIRAN

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Lastly, I would like to thank my friends for their encouragement throughout the course of this project.
I certify that an Examination Committee has met on 23rd March 2006 to conduct the final examination of Siew Fong Wah on his Master of Science thesis entitled "Theory and Simulation of the Onsets of Convection Induced by Liquid Diffusion in Liquid and Incipient Instability Caused by Liquid Flow in Particle Bed" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the candidate be awarded the relevant degree. Members of the Examination Committee are as follows:

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Date: 10 AUG 2006
DECLARATION

I hereby declare that the thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at UPM or other institutions.

Date: 13 July 2006

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LIST OF ABBREVIATIONS/NOMENCLATURE

\( a_p \)  center coefficient
\( a_{nb} \)  influence coefficients for the neighboring cells
\( \bar{a}_c \)  dimensionless critical wavenumber
\( A \)  total area of the cross section of the bed column, m\(^2\)
\( c^* \)  bulk concentration or saturated concentration of solute, kmol/m\(^3\) or kg/m\(^3\)
\( c^0 \)  initial concentration of solute, kmol/m\(^3\) or kg/m\(^3\)
\( c \)  concentration, kmol/m\(^3\) or kg/m\(^3\)
\( C_D \)  drag function
\( D_b \)  bed diameter, m
\( d_p \)  particle diameter, m
\( D \)  diffusivity of mass, m\(^2\)/s
\( E \)  total energy, J
\( \bar{F}_q \)  external body force, N
\( \bar{F}_{\text{lift},q} \)  lift force, N
\( \bar{F}_{\text{v,mq}} \)  virtual mass force, N
\( g \)  gravitational force, m/s\(^2\)
\( h \)  heat transfer coefficient, W/m\(^2\).K
\( H \)  height of the particle bed, m
\( H \)  height of simulation chamber, mm
\( j \)  mass transfer, kg/m\(^2\)
\( j^0 \)  constant mass flux, kg/m\(^2\).s
\( k \)  thermal conductivity, W/m°C
\( k_{\text{eff}} \)  effective conductivity, W/m.K
\( K_{sl} \)  solid-fluid exchange coefficient
\( K_e \)  permeability, m\(^2\)
\( m \)  molality, mol/kg
\( q \)  constant heat flux, W/m\(^2\)
\( MW \)  molecular weight of a component, kg/kmol
\( \Delta p \)  pressure drop across the bed, Pa
\( R^4 \)  residual
\( \vec{R}_{pq} \) interaction force between phases, N

\( t \) time, s

\( \Delta t \) time step

\( u_e \) interstitial velocity, m/s

\( U \) superficial velocity of the mass flow, m/s

\( U_e \) interstitial velocity, m/s

\( U_{mf} \) minimum fluidization velocity, m/s

\( V \) molar volume, \( m^3/kmol \)

\( V^o \) partial molar volume, \( m^3/kmol \)

\( V_\varnothing \) apparent molar volume, \( m^3/kmol \)

\( W \) weight of the solids in the bed, kg

\( W \) width of simulation chamber, mm

\( x \) mass fraction of component, kg/kg

\( y \) mole fraction of component, kmol/kmol

\( z \) effective distance of the mass diffusion, m

Greek Symbols

\( \rho \) density, kg/m\(^3\)

\( k_{sl} \) exchange coefficient

\( \alpha \) molar density coefficient, kg/kmol

\( \beta \) thermal expansion, 1/K

\( \kappa \) thermal diffusivity, m\(^2\)/s

\( \tau_s \) particulate relaxation time, s

\( \varepsilon \) porosity

\( \tau_q \) \( q^{th} \) phase stress-strain, Pa

\( \Psi \) shape factor of particle or sphericity

\( v_{r,s} \) terminal velocity correlation for the solid particle

\( \tau \) tortuosity

\( \mu \) dynamic viscosity, kg/ms

\( \nu \) diffusivity of momentum, kinematic viscosity, \( = \mu / \rho \), m\(^2\)/s

\( \lambda \) wavelength, m
Abbreviations

LSA  Linear Stability Analysis
FSC  Fixed Surface Concentration
CMF  Constant Mass Flux
Bi   Biot number
Gr   Grashof number
Ra   Rayleigh number
Re   Reynolds number
Sc   Schmidt number

Subscripts

c    critical
c    solid particle bed
f    fluid
n    1-butanol
i    interfacial
o    initial state
p    particle
s    solution of solute and solvent
v    voids
w    water
\text{eff}  effective
\text{max}  maximum
CHAPTER 1
INTRODUCTION

Mass transfer may produce density gradient that in turn causes free convection. As an example, imagine that a crystal of salt is held just under the surface of initially stagnant water. The salt dissolves and diffuses away from the crystal, producing a solution near the crystal that is denser than the water itself. This solution flows downward, and the resulting flow increases the rate of mass transfer. Thus, mass transfer and flow couple to accelerate dissolution.

The natural convection in a binary gas-liquid system (Okhotsimskii and Hozowa, 1998; Blair and Quinn, 1969) and Maragoni-unstable interfacial turbulence at liquid-liquid interface (Sterling and Scriven, 1959; Orell and Westwater, 1962; Nakaike et al., 1969) have been extensively studied. The transient instability theory of gas diffusion in liquid that leads to the onset of buoyancy convection has been reported by Tan and Thorpe (1996, 1999). However, the case of a Rayleigh-unstable binary liquid system in which the density-driven plume convection happens has attracted much less attention (Berg and Morig, 1969). A new transient Rayleigh number is needed to study the onset of convection induced by liquid diffusion in liquid. The density gradient developed by transient liquid mass diffusion of a solute in a solvent may contribute to the convection accompanying the mass transfer.
The knowledge of natural convection induced by diffusing liquid into a stagnant liquid is of considerable interest because of its importance in the study of mass transfer of liquid solute in solvent for biological and pharmaceutical use, dispersion of effluent in treatment plants and lakes, and other engineering applications. Convection causes more liquid to be dissolved in the solution compared to diffusion.

Fluidization is a two-phase contact process which relies on keeping up the grains of solid substance in suspension state in the flux flowing up with appropriate velocity. The fluidization plays an essential role in many engineering processes because intensive mixing of phases is conductive to all sorts of diffusion phenomena, such as: intensive exchange of mass and heat, generation of uniform field of temperatures, etc.

The linear stability analysis (LSA) was first employed by Jackson (1963) to study the onset of incipient instability by assuming a convection mechanism. The inertia force of the particle imparted by average fluid velocity was found to contribute to the instability. Past researches considered the increase of voidage as an indicator of the onset of fluidization, although the expansion of voidage is in fact a consequence of fluidization of the bed.

The main cause of incipient fluidization is the momentum of the high incoming flux that overcomes the buoyancy force of the solid particles. Moreover, the incipient
instability is preceded by the flowing of the liquid into the interstices of the particle bed with a mass flux or superficial velocity exceeding the minimum fluidization velocity, $U_{mf}$. Therefore the first abrupt increase of flow velocity should be the indicator of the onset of incipient fluidization when the localized saturation of the interstices is sufficient to lift the particles. The onset of incipient fluidization caused by liquid flow in particle bed is coupled with the immense change of mass fraction of the liquid in the region close to the flow inlet.

The objectives of this work are to study the onset of convection caused by liquid diffusion in liquid and the onset of incipient instability caused by liquid flow in particle bed. A critical Rayleigh number will be derived for the prediction of the onset of buoyancy convection when water diffuses into various partially miscible organic liquids at different temperatures. For the onset of incipient instability of liquid flow in particle, a critical Rayleigh number based on momentum diffusion will be derived. The derivation performed in this study follow closely the principle of transient instability theories advanced by Tan and Thorpe (1996 and 1999) and Tan (2004).

Two-dimensional time-dependent simulations will be conducted to verify the principles of transient instability. The onset of buoyancy convection induced by liquid mass diffusion and the mechanism of incipient instability in a particle bed will be simulated using a CFD package, FLUENT. Parameters such as the onset time of
instability, critical diffusion depth, size of the plume and critical Rayleigh number will be obtained from simulations.

This study predicts the particular mass transfer parameters for liquid diffusion in liquid as well as liquid flow in particle bed without conducting the real experiments. These experiments are hardly to be carried out as advanced technology and equipments are required. The simulated design of experiment in this research may shed some light for the experiment models.
CHAPTER 2
LITERATURE REVIEW

2.1 Introduction
The theory of transient convection and instabilities will first be reviewed. This is followed by the discussion of the experimental studies of convective instability induced by adverse density gradient. The typical convection patterns are presented. Finally, discussions on the instability and flow pattern in liquid fluidized bed are given.

2.2 Theory of Transient Convection and Instabilities
Lord Rayleigh (1916) derived a criterion for the onset of buoyancy convection in a fluid layer bounded by two free surfaces based on an adverse linear temperature gradient typical of steady-state heat transport; the stability criterion is rearranged later to become the well-known Rayleigh number,

\[ Ra = \frac{g \beta d^3 \Delta T}{\nu \kappa} \]  \hspace{1cm} (2.1)

where \( g \) is the gravitational acceleration, \( \beta \) is the thermal expansion, \( d \) is the depth of fluid layer, \( \Delta T \) is the temperature difference of two surfaces, \( \nu \) is the kinematic viscosity and \( \kappa \) is the thermal diffusivity.

However, natural convection is induced by a time-dependent and non-linear temperature profile. Several definitions of time-dependent Rayleigh number were suggested by Howard (1964) and Davenport and King (1972), but none of them has