Onset of buoyancy-driven motion with laminar forced convection flows in a horizontal porous channel

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Abstract
Bouyancy-driven motion in the laminar forced convection flow has been investigated in a horizontal porous channel. The stability equations including the inertia and the dispersion effects have been solved analytically under the linear theory and also the principle of exchanges of stabilities. The resulting critical position for the manifest convection, i.e. the undershoot distance, has been examined when the Reynolds number increases. The critical position becomes larger with increasing the Reynolds number, Re, considering the permeability. This means that the more inertia and dispersion make the system more stable.

Keywords:
Buoyancy-driven instability
Porous media
Dispersion effect
Inertia effect
Linear stability theory
Electrochemical experiment

1. Introduction
The convective motion driven by the buoyancy forces appears in the fluid-saturated porous layer subjected to heating from below. This is well known as the Horton–Rogers–Lapwood problem [1,2]. The convection in porous media plays an important role in science and engineering applications, such as mantle recurrence, oil recovery, storage of grain and nuclear reactor safety. An interesting extension of the Horton–Rogers–Lapwood problem is the mixed convection under the weak through flow [3]. Many studies on the transient behaviors the mixed convection in the porous layer have been carried out after Prats [4] analyzed the time-dependent oscillating motion. Combarnous and his colleagues [5,6] showed that the phase transition of the time-dependent transverse rolls to the steady longitudinal vortices appears while the rate of through flow increases. Dufour and Neel [7] suggested the criteria on the absolute and convective instabilities using the weakly-nonlinear analysis. Recently, Chung et al. [8] suggested the reasonable stability criteria for the fluid-saturated porous media under the forced convection using both linear and weakly nonlinear analyses, and also the direct numerical simulation. According to the survey of previous analyses, it seems that the longitudinal vortices dominate the transverse rolls for the specific region where an inertial effect is dominant.

These works are mainly focused on the systems with the fully-developed base fields. For the various systems experiencing the developing nonlinear base fields, Choi and his colleagues [9–12] have analyzed the onset of the vortex instabilities successfully by employing their propagation theory. Lee et al. [11] examined the laminar vortex instability on the natural convection flow over the inclined surface embedded the porous layer, where the limiting current method in an electrochemical system has been done for promising the trend line of their stability conditions consistently. For a forced convection flow through the porous media, Chung et al. [12] analyzed the onset of the convective instability including the inertia and the dispersion effects. They showed that these effects make the system stable.

The experimental detection on the natural convection for a high Rayleigh number is very difficult due to the side effects and the difficulties in the control of boundary conditions. Furthermore, the observation of the convective motion in the porous media is hardly detected due to the structural complexity. To overcome the above mentioned problems, electrochemical systems [13–15] under the limiting current condition [16] have been used in the natural convection fields especially for the very large Rayleigh number situations. Lee et al. [11] showed that this method can be extended to the system of the fluid-saturated porous media. In the porous media systems, the dispersion effect caused by the variation of...
porosity, the thermal deviation between solid and fluid, and the fluid velocity \cite{17–20} plays an important role in the determination of stability criteria. But, the stabilization from the buoyancy forces due to the dispersion effects is not examined yet for the convective flows in a horizontal porous channel.

In the present study, we will analyze the onset of a buoyancy-driven instability for the laminar convection flow through the porous layers. The stability condition from the linear stability analysis will be compared with the electrochemical mass transfer experiment. Based on the comparison between the theoretical and experimental results, the inertia and the dispersion effects on the onset of buoyancy-driven secondary motion will be quantified.

2. Theoretical analysis

2.1. Governing equations

The mass transfer system considered here is a fluid-saturated porous layer with uniform superficial velocity \( U_0 \) and uniform concentration in \( C_i \), the fully-developed laminar flow (see Fig. 1). The porous layer is confined between two horizontal plates of depth \( H \). The lower and upper plates are kept at constant concentration \( C_i \) and \( C_s \), respectively. For a small distance in the streamwise \( X \)-direction, the nonlinear concentration profile develops gradually. For \( X > 0 \), the laminar concentration boundary-layer thickness \( \Delta c \) increases with increasing \( X \), and the buoyancy-driven secondary flow will set in at a certain distance. For the isotropic porous media, the governing momentum equation is expressed using the Forchheimer’s equation, Boussinesq approximation and dispersion model:

\[
\frac{\mu}{K} \nabla U \cdot \frac{c_F \rho}{\sqrt{K}} U = -\nabla P - \kappa \rho g C,
\]

where \( U \) is the superficial velocity vector and \( \kappa \) is the unit vector of positive \( Z \)-direction. And the general governing equations for the continuity and mass balances are as follows:

\[
\nabla \cdot U = 0,
\]

\[
\frac{\partial C}{\partial t} + U \cdot \nabla C = \nabla \cdot (D \nabla C),
\]

where \( t \) and \( D \) denote the time and the dispersion tensor, respectively. Hsu and Cheng \cite{17} suggested that \( D = D_e + APe^D_e \), where \( D_e \) is the effective diffusivity, and \( A \) is the constant 2nd-order tensor which is diagonal. They assumed \( n = 1 \) for high \( Re_d \) and \( n = 2 \) for low \( Re_d \). Here, \( Pe_d = U_0 d_i / D_e \) and \( Re_d = \rho U_0 d_i / \mu \) is the Péclet number and the Reynolds number based on the particle diameter \( d_i \), respectively.

\[\mu = \frac{C_F \rho}{\sqrt{K}} \]

(1)

\[\nabla \cdot U = 0,\]

(2)

\[\frac{\partial C}{\partial t} + U \cdot \nabla C = \nabla \cdot (D \nabla C),\]

(3)

\[\frac{\mu}{K} \nabla U \cdot \frac{c_F \rho}{\sqrt{K}} U = -\nabla P - \kappa \rho g C,\]

\[\nabla U = 0,\]

\[\frac{\partial C}{\partial t} + U \cdot \nabla C = \nabla \cdot (D \nabla C),\]

\[\mu = \frac{C_F \rho}{\sqrt{K}}\]

\[\nabla \cdot U = 0,\]

\[\frac{\partial C}{\partial t} + U \cdot \nabla C = \nabla \cdot (D \nabla C),\]

\[\mu = \frac{C_F \rho}{\sqrt{K}}\]

(1)

where \( U \) is the superficial velocity vector and \( \kappa \) is the unit vector of positive \( Z \)-direction. And the general governing equations for the continuity and mass balances are as follows:

\[
\nabla \cdot U = 0,
\]

\[
\frac{\partial C}{\partial t} + U \cdot \nabla C = \nabla \cdot (D \nabla C),
\]

Fig. 1. Schematic diagram of the system considered here.
For the laminar region of \( \text{Re}_K \leq 1 \) and \( Pe(=U_0H/D_2) > 100 \), the volume-averaged, steady state, basic concentration is represented in the dimensionless form:

\[
\frac{\partial c_0}{\partial x} = \frac{\partial}{\partial z} \left( 1 + \gamma_1 \frac{\partial c_0}{\partial z} \right).
\] (4)

For the electrochemical system under the limiting current condition, by considering Kamotani et al.’s work [21] the proper inlet and boundary conditions are expressed as

\[ c_0(0, z) = 0 \text{ and } c_0(x, 0) + 1 = c_0(x, 1) - 1 = 0, \] (5)

where \((x, z) = (X/Pe, Z[H], c_0 = (C_0 - C_1) / \Delta C, \Delta C = (C_u - C_1) = (C_1 - C_0), (1 + \gamma_1) \) is the ratio of the vertical dispersion tensor to the effective diffusivity \( D_e \). The solution of Eq. (4) is obtained analytically for the present system:

\[
c_0 = \sum_{n=0}^{\infty} \left[ \text{erfc} \left( \frac{n + 1/2}{\sqrt{(1 + \gamma_1)x}} \right) - \text{erfc} \left( \frac{n + 1/2}{\sqrt{(1 + \gamma_1)x}} \right) \right] \frac{1}{\sqrt{(1 + \gamma_1)x}}.
\] (6)

where \( \xi = z/\sqrt{(1 + \gamma_1)x} \). For the limiting case of small \( x \), the above concentration field can be approximated as

\[
c_0 = \begin{cases} \text{erfc} \left( \frac{1/2}{\sqrt{(1 + \gamma_1)x}} \right), & z \geq 0.5, \\ -\text{erfc} \left( \frac{1/2}{\sqrt{(1 + \gamma_1)x}} \right), & z \leq 0.5. \end{cases} \] (7)

As shown in Fig. 2, the approximate solution of Eq. (2) represents the exact one given by Eq. (6). Our primary concern is to find the onset of buoyancy-driven convection for \( x \to 0 \), and therefore we use Eq. (7) as a base concentration field.

### 2.2. Linear stability theory

Under the linear stability theory, infinitesimal disturbances are initiated from their unperturbed basic quantities upon the onset position of mixed convection. Then the linearized disturbance equations from Eqs. (1)–(3) are described as

\[
\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} = 0,
\] (8)

\[
\frac{1}{Da} \left( 2 + \text{Re}_K^{-1} \right) u = -\frac{1}{Pe^2} \frac{\partial p}{\partial x},
\] (9)

\[
\frac{1}{Da} \left( 2 + \text{Re}_K^{-1} \right) w = -\frac{1}{Pe} \frac{\partial c}{\partial x},
\] (10)

\[
\frac{1}{Da} \left( 2 + \text{Re}_K^{-1} \right) \frac{\partial c}{\partial x} + \frac{Ra}{Re_K} \frac{\partial c_0}{\partial x} - \frac{Ra}{Re_K} w \frac{\partial c_0}{\partial z} = \frac{1}{2} \left( 1 + \gamma_1 \right) \left( \frac{\partial^2 c}{\partial y^2} + 1 + \gamma_1 \frac{\partial^2 c}{\partial z^2} \right),
\] (11)

where the physical variables are non-dimensionalized by \((u, v, w) = (U_1/Pe, V_1, W_1)R_1H/D_e, y = Y/H, p = P_1H^2/\rho H^2, \gamma = RaC_1/\Delta C \) and \( \tau = D_e \tau H^2 \). Here, \( Ra = \rho g H vC_1/\nu \) and \( Da = k/H^2 \) are defined as the Rayleigh number and the Darcy number, respectively. The subscript 0 denotes the unperturbed quantity, \( U_1, V_1 \) and \( W_1 \) are the perturbed velocity components in the Cartesian coordinates, and \( P_1 \) is the perturbed pressure.

For the steady laminar region of \( \text{Re}_K < 1 \) and \( Pe > 100 \), the terms containing \( 1/Pe^2 \) can be ignored, and therefore \( u = 0 \) and axial diffusion was neglected. By removing the pressure terms with the aid of the continuity equation of Eq. (8), the stability equations are reduced as

\[
\frac{\partial^2 w}{\partial y^2} + \frac{\partial^2 c}{\partial y^2} \frac{\partial^2 c}{\partial z^2} = -\frac{\partial^2 c}{\partial y^2},
\] (13)

\[
\frac{\partial c}{\partial x} + \left( \frac{1}{1 + 2\text{Re}_K} \right) w \frac{\partial c_0}{\partial y} = \left( 1 + \gamma_1 \right) \left( \frac{\partial^2 c}{\partial y^2} + \frac{\partial^2 c}{\partial z^2} \right),
\] (14)

where \( w = w(2 + \text{Re}_K^{-1} Da) \) and \( Ra_0 = Ra Pe ) \) is the Darcy–Rayleigh number. The proper boundary conditions are

\[
w = c = 0 \text{ at } z = 0 \text{ and } 1.
\] (15)

The boundary conditions represent the slip and the constant concentration on each boundary.

Since the coefficients of the above stability equations of (13) and (14) are independent of \( y \), the disturbance quantities in the form of longitudinal vortex rolls are expressed under the normal mode as follows:

\[
\left\{ \begin{array}{l}
\{ w(x, y, z) \\
\{ c(x, y, z) \\
\end{array} \right\} = \left\{ \begin{array}{l}
w(x, z) \\
c(x, z) \\
\end{array} \right\} \exp(\text{i}ay).
\] (16)

In the entrance region of small \( x \), we transformed the disturbance equations such that the eigenfunctions associated with the vertical diffusion operator are localized around the base-concentration front. Following a coordinate transformation to the similarity variable of the basic state \( \zeta = z/\sqrt{(1 + \gamma_1)x} \) and the perturbation equations can be expressed:

\[
(D - a^2) w = a^2 c,
\] (17)

\[
\frac{\partial c}{\partial x} = \left( D^2 + \frac{\zeta}{2} D - a^2 \right) c - Ra_0 w Dc_0,
\] (18)

where \( D = \partial / \partial \zeta, w(\zeta, x) = w(x, z), c(x, \zeta) = c(x, z), a = a/\sqrt{(1 + \gamma_1)x}, Ra_0 = R0 \zeta^{1/2} / (2 + \text{Re}_K)^{1/2} \sqrt{1 + \gamma_1} \) and \( Dc_0 = \exp(-\zeta^2/4) / \sqrt{\pi} \). The boundary conditions are

\[
w = c = 0 \text{ at } \zeta = 0 \text{ and } \zeta \to \infty.
\] (19)

### 2.3. Solution procedure

In order to solve the stability equations (17) and (18) with the boundary conditions (19), let us define the function as

\[
\Phi = (D^2 - a^2) w
\] (20)

and then Eq. (18) is written as

\[
\frac{\partial \Phi}{\partial \zeta} = \left( D^2 + \frac{\zeta}{2} D - a^2 \right) \Phi + a^2 Ra_0 w \frac{1}{\sqrt{\pi}} \exp\left(-\frac{\zeta^2}{4}\right).
\] (21)
The boundary conditions on the bounding surface are equivalent to
\[ w' = \Phi = 0 \quad \text{at} \quad \zeta = 0 \quad \text{and} \quad \zeta \to \infty. \]  
(22)

Using the property of generalized Fourier series, \( \Phi \) can be expressed as
\[ \Phi = \sum_{n=1}^\infty A_n(x)\phi_n(\zeta), \]
(23)
where \( \phi_n \)'s are the orthogonal functions satisfying the following Sturm–Liouville boundary value problem,
\[ \left( D^2 + \frac{\zeta}{2} D \right) \phi_n = -\lambda_n \phi_n \]
(24)
with the following boundary conditions
\[ \phi_n(0) = \phi_n(\infty) = 0. \]
(25)
The present Sturm–Liouville boundary value problem for Eqs. (24) and (25) is solved as
\[ \phi_n = H_{2n-1} \left( \frac{\zeta}{2} \right) \exp \left( -\frac{\zeta^2}{4} \right) \quad \text{and} \quad \lambda_n = n. \]
(26)
where \( H_k \) is the \( k \)th Hermite polynomial.

By combining Eqs. (20) and (23), \( w^* \) is expressed as
\[ w^* = \sum_{n=1}^\infty A_n \psi_n(\zeta), \]
(27)
where \( \psi_n \)'s is obtained by solving
\[ (D^2 - a^2) \psi_n(\zeta) = \phi_n(\zeta) \]
(28)
with the boundary conditions of
\[ \psi_n(0) = \psi_n(\infty) = 0. \]
(29)
The solution of (28) is expressed as
\[ \psi_n = \frac{1}{2a} \left[ \exp (a^2)f(\zeta) - f(\zeta) - \exp (-a^2)f(\zeta) - f(\zeta) \right], \]
(30a)
where
\[ f_{n1} = \int_0^\zeta \exp \left( -a^2 \zeta - \frac{\zeta^2}{4} \right) H_{2n-1} \left( \frac{\zeta}{2} \right) d\zeta, \]
(30b)
\[ f_{n2} = \int_0^\zeta \exp \left( a^2 \zeta - \frac{\zeta^2}{4} \right) H_{2n-1} \left( \frac{\zeta}{2} \right) d\zeta. \]
(30c)
Now, substituting for \( \Phi \) and \( w^* \) from Eqs. (23), (27) and (30) in Eq. (21), we obtain
\[ \sum_{n=1}^\infty \frac{dA_n}{dx} = -\sum_{n=1}^\infty A_n (\lambda_n + a^2) \phi_n(\zeta) 
\]
\[ + Ra_0 a^2 \frac{1}{\sqrt{\pi}} \exp \left( -\frac{\zeta^2}{4} \right) \sum_{n=1}^\infty A_n \phi_n(\zeta). \]
(31)
By using the orthogonal property of \( \phi_n(\zeta) \), multiplying Eq. (31) by \( \exp \left( \frac{\zeta}{2} \right) \phi_n(\zeta) \) and integrating over the range of \( \zeta \), the following system of homogeneous linear equation for the constants \( A_n \) is obtained:
\[ \frac{dA_n}{dx} = -\left( \lambda_n + a^2 \right) A_n \int_0^\zeta \phi_n^2(\zeta) \exp \left( \frac{\zeta^2}{4} \right) d\zeta 
\]+ \[ Ra_0 a^2 \frac{1}{\sqrt{\pi}} \sum_{n=1}^\infty A_n \int_0^\zeta \phi_n(\zeta) \phi_n(\zeta) d\zeta. \]
(32)
Now, Eq. (32) is written in the following matrix form:
\[ x \frac{dA}{dx} = B_a, \]
(33)
where
\[ B_a = -\left( \lambda_i + a^2 \right) A_i + Ra_0 a^2 \frac{1}{\sqrt{\pi}} C_{ij}, \]
\[ C_{ij} = \int_0^\infty \phi_i(\zeta) \phi_j(\zeta) \exp \left( -\frac{\zeta^2}{4} \right) d\zeta, \]
\[ G_{ij} = \int_0^\infty \phi_i(\zeta) \phi_j(\zeta) d\zeta, \]
\[ a = [A_1, A_2, A_3, \ldots, A_n]^T. \]
Due to the orthogonal property of \( \phi_i \)'s, \( G_{ij} = 0 \) for \( i \neq j \) wherein exp \( (\zeta^2/4) \) is the weight function. After integrating them by parts, the following relation is obtained:
\[ G_{ij} = \int_0^\infty \phi_i(\zeta) \phi_j(\zeta) d\zeta = \int_0^\infty \left( D^2 - a^2 \right) \phi_i(\zeta) \phi_j(\zeta) d\zeta 
\]
\[ = \int_0^\infty \phi_i(\zeta) \left( D^2 - a^2 \right) \phi_j(\zeta) d\zeta 
\]
\[ = \int_0^\infty \phi_i(\zeta) \left( D^2 - a^2 \right) \phi_j(\zeta) d\zeta = \int_0^\infty \phi_j(\zeta) \phi_i(\zeta) d\zeta = G_{ji}. \]
(34)
Since the coefficient matrix \( B \) is \( x \)-dependent, normal mode analysis cannot be applied into the present system anymore. To resolve this problem, we introduce the local stability analysis (LSA) where the matrix is fixed at some point, i.e. \( a^2 \) and \( Ra_0 \) are considered to be constant [9–12]. Under the LSA, the neutral stability is determined by the largest eigenvalue of the matrix \( B \), i.e.
\[ A_{max}(B) = 0, \]
(35)
where \( A_{max}(\cdot) \) denotes the largest eigenvalue.

The first approximation to the solution of Eq. (35) is obtained by setting \( B_1 \equiv 0 \), which corresponds to \( \Phi = A_1 \phi_1(\zeta) \) and \( w^* = A_1 \psi_1(\zeta) \). Then, the solution of this first approximation is obtained from
\[ Ra_0 = \frac{1 + a^2}{a^2} \sqrt{\pi} \int_0^\infty \phi_i(\zeta) \phi_j(\zeta) d\zeta = \frac{(1 + a^2)}{a^2} \sqrt{\pi} C_{11}/C_{11}. \]
(36)
If we consider one more term, i.e. \( \Phi = \sum_{n=1}^\infty A_n \phi_n(\zeta) \) and \( w^* = \sum_{n=1}^\infty A_n \psi_n(\zeta) \), Eq. (35) reduces to the following quadratic equation
\[ (Ra_0)^2 \left\{ C_{11} C_{22} - G_{12} \right\} 
\]
\[ - \left\{ \sqrt{\pi} \left( \frac{1}{a^2} + 1 \right) C_{11} C_{22} + \sqrt{\pi} \left( \frac{2}{a^2} + 1 \right) G_{11} C_{22} \right\} R a_0 \]
\[ + \pi \left( \frac{1}{a^2} + 1 \right) C_{11} C_{22} = 0. \]
(37)
Similarly to the above, the higher approximation can be possible. The neutral stability curves from various approximations are summarized in Fig. 3, and the critical conditions which correspond to the minimum point of each curve are given in Table 1.

For the laminar region of \( Ra_{le} \leq 1 \) and \( Pe > 100 \) where the most unstable mode is transverse roll, the neutral stability curves obtained from the linear stability analysis were shown in Fig. 3. Here, the upper region of curve means unstable, while the lower one stable. All these results are valid to \( \Delta c < H \) under the assumption on growth of disturbances in the concentration boundary–layer thickness. In this figure, the stability criteria of minimum \( Ra_{le} \), for a given the critical wavenumber, \( q_c \) were obtained by comparison of 3rd approximation of analytic solution with numerical calculation of Chung et al. [12]. In the deep-pool system of small \( x_c \) the critical values including the dispersion effect are found to be.
The effect of the migration on the onset of buoyancy-driven convection is confined within the narrow regions near both plates. This means the buoyancy force is originated from the density difference between bulk and lower copper-deposited surface. Therefore, the present theoretical predictions can be applied in the present electrochemical experiments on basis of the similarity between the heat transfer and the present mass transfer in conventional boundary-layer systems. To minimize the electromigration effect which breaks the similarity between heat transfer system and mass transfer one, the sulfuric acid as a supporting electrolyte was added like many previous experiments \[9,10,21–26\].

In the present experiment, the electrolyte consists of 0.1 M CuSO$_4$ solution with 1.5 M H$_2$SO$_4$ as a supporting electrolyte, where sulfuric acid was added as a supporting electrolyte to lessen the electromigration effect. Copper ion was deposited on the cathode electrode under the limiting current condition, and was dissolved from the anode one. The electrical information in the cell were obtained by a PC controlling potentiostat (EG&G Parc.) on line. The calomel reference electrode was used to measure the potential difference between the electrolyte solution and the cathode. Experiments were repeated in unstable (cathode facing upward) or stable (cathode facing downward) conditions under various flow rates, $U_D$ and the length of cathode ($L_t=1\sim 35$ cm). From the measured limiting current density the critical distance $X_c$ to mark the onset of instability was determined at the position deviated from the well-known diffusion relation.

Now, we will discuss some simplifications and assumptions applied in the present study. In electrochemical systems, the concentration difference between upper and lower plates is $\Delta C_{ub} = 2\Delta C$ \[24\], where $\Delta C = C_i - C_l$. However, since in the present system the concentration boundary-layer thickness $\Delta_C$ is much smaller than the depth of the porous layer $H$ as shown in Fig. 2, the effect of buoyancy forces is confined within the narrow regions near both plates. This means the buoyancy force is originated from the density difference between bulk and lower copper-deposited surface. Therefore, the present theoretical predictions can be applied the present electrochemical experiments on basis of the similarity between the heat transfer and the present mass transfer in conventional boundary-layer systems. To minimize the electromigration effect which breaks the similarity between heat transfer system and mass transfer one, the sulfuric acid as a supporting electrolyte was added like many previous experiments \[9,10,21–26\].

The effect of the migration on the onset of buoyancy-driven convection was analyzed systematically by Jiang et al. \[25\] and Volgin et al. \[26\]. Volgin et al.’s \[26\] showed that the supporting electrolyte gives minor effects on the onset of convection. We consider the effect of migration on the buoyancy force by following Fenech and Tobias’s method \[14\], where the rate of migration of the supporting
The typical potential–current curve is feature in Fig. 5. As an increasing in the applied potential difference slowly, the current at first increased rapidly and then reached a saturation level such as a ‘plateau’. Only upon relatively higher increase of the applied potential, the current rises appreciably again. At this stage, the hydrogen ions take part in electrochemical reaction and hydrogen gas bubbles evolve. The point at which an increase in potential difference causes almost no increase in current density is known as the limiting current density, \( I_L \). Under the limiting current condition the average mass transfer coefficient \( h_l \) equivalent to the average heat transfer coefficient over the plate length \( L \) can be obtained from the relation of

\[
h_l = \frac{(1 - \tau_{e})h_1}{nFb},
\]

where \( C_b, \tau_{e}, n \) and \( F \) denote the bulk concentration of the deposited metal ion, the transference number which explains the migration effect, the valence of the transferred ion and Faraday’s constant, respectively. Under the limiting current density, the concentration difference \( \Delta C(=C_b - C_i) \) becomes much larger due to the accelerating mass transfer caused by an increasing potential difference, where \( C_i \) is the concentration at the deposited surface. Consequently, the mass transfer will reach to the maximum, because \( \Delta C \) approaches the maximum (\( C_i \to 0 \) when \( C_b \to \) constant). In order to measure mass transfer rate for a given buoyancy force the Sherwood number and the Darcy–Rayleigh number are defined as

\[
Sh = \frac{h_lH}{D_e}, \quad Ra_0 = \frac{g(\rho_b - \rho_s)KH}{D_e\mu},
\]

where \( \rho_b \) and \( \rho_s \) represent the bulk density and the density at the electrode surface, respectively.

To calculate the above dimensionless numbers, the effective diffusivity is essential. Since the local mass transfer rate cannot be measured directly, we measured the time-dependent diffusion coefficient \( D_e \) using the well-known Cottrell equation, \( I_c(t) = nFC_bDI_e^{1/2}/(\pi t)^{1/2} \) [27], the effective diffusivities are obtained by correlating the experimental time-dependent current density as shown in Fig. 6. For the homogeneous CuSO\(_4\)–H\(_2\)SO\(_4\) system, the present experimental diffusivity \( D = 5.921 \times 10^{-6} \text{ cm}^2/\text{s} \) shows a good agreement with Fenich and Tobais’s [14] \( 5.554 \times 10^{-6} \text{ cm}^2/\text{s} \). For the fluid-saturated porous system, we obtained the effective diffusivities as \( D_e = 8.51 \times 10^{-7} \text{ cm}^2/\text{s}, 1.83 \times 10^{-6} \text{ cm}^2/\text{s} \) and \( 4.48 \times 10^{-6} \text{ cm}^2/\text{s} \) for the 1.5 mm, 3 mm and 8 mm diameter glass beads used as packing materials, respectively.

**Fig. 6.** Effective diffusivity determined by relation of current density with time for various diameters of porous media.

**4. Results and discussion**

Combarnous and Bia [5] observed the flow transition from traveling transverse rolls to stationary longitudinal vortices for \( Pe \approx 7.6 \) and \( Ra_0 \approx 50 \). In the present study, we tried to analyze the onset condition of the buoyancy-driven convection for the system of \( Pe > 100, Ra_0 < 100 \) and \( Sc \geq 2000 \), where \( Sc \) denotes the Schmidt number (=\( \mu/(\rho D_e) \)) in the mass transfer system. Even if the flow transition occurs at higher \( Pe \) with increasing \( Ra_0 \) like the previous studies [5–8] and the present experimental condition, it is expected that in the present system the laminar vortex instabilities dominates the oscillating roll-type instabilities.

For the Darcy’s region of \( Re_d \leq 5 \), the present experimental mixed convection mass transfer results are compared with the pure forced convection mass transfer rate in Darcy’s limit in Fig. 7(a), where the solid line represents the mass transfer rate without the inertia and dispersion effects, which is an identical with \( Sh_{Re_0} = 1/\sqrt{(\pi \xi)} \) with \( \gamma_1 = 0 \) (see Eq. (7)). As shown in this figure, the present mass transfer rates follows \( Sh = Sh_{Re_0} \) for a certain region of \( x < x_{uc} \). This means that the inertia and dispersion effects are not significant for small \( Re_d \)-region and the buoyancy effects are sensible for \( x \gg x_{uc} \). Here, we define the characteristic distance \( x_{uc} \) at which \( Sh \) shows the minimum value. For the Forchheimer region of \( 5 < Re_d < 120 \), the mass transfer rate deviates from that of Darcy’s limit, i.e. \( Sh = Sh_{Re_0} \) and the inertia and dispersion effects make the system more stable, as shown in Fig. 7(b). In this region, the effect of dispersion on the onset of the buoyancy-driven motion is more remarkable than those on the purely forced convection mass transfer rate. In other words, the effect of \( Re_d \) on \( x_{uc} \) is more significant than that of purely forced convective \( Sh \).

In Fig. 8, for given \( Ra_0 \) and \( Re_d \), the theoretical critical position \( x_{uc0} \) is compared for the experimental undershoot position \( x_{uc} \) (see Fig. 7). Here \( x_{uc0} \) is the theoretical critical position without dispersion effect, \( x_{uc0} = 167.54 ((1 + 2Re_0)/Ra_0)^{1/2} \). The experimental \( x_{uc} \) for a given \( Ra_0 \) increases with increasing \( Re_d \). It is noted that the bound of \( Re_d \leq 0.1 \) represents the Darcy-flow region through the relation of \( Ra_d \approx 55Re_d \) for the present system of \( \tau \approx 0.36 \).
The dispersion effect depends on the flow complexity in porous layer and the velocity. The effect of the variation of particle diameters on the dispersion seems to be very weak, and the conductivity ratio between solid and fluid is hardly expected because this mass-transfer system consists of the cupric sulfate–sulfuric acid and the glass bead. It is known that the dispersion coefficient is proportional to $\gamma_1 = Pe^{n/2}K$. We defined the dispersion coefficient, $c_L$ as

$$c_L = a_0 Pe^{n/2} \frac{K}{R_d}; \quad (41)$$

which is slightly modified form of the conventional $\gamma_1 = Pe^{n/2}K$. [17].

By letting $x_u = x_c$ and $Sc = 2000$ and combining Eqs. (38a) and (41), we can obtain the following relation:

$$\gamma_1 = \left( \frac{R_d x_c^{1/2}}{12.944(1 + 2Re)} \right)^2 - 1 = a_0 Pe^{n/2} K.$$

As shown in Fig. 9, by comparing the present experimental data and Eq. (42), we construct the following dispersion relation under the least square concept: 

$$\gamma_1 = 0.02 Pe^{1.10}K \quad \text{for } Sc = 2000. \quad (43)$$

Hsu and Cheng [17] suggested $n = 1$ for high $Re_d$ and $n = 2$ for low $Re_d$. Since the present experiment covers the Darcy flow and the Forchheimer one, our exponent 1.10 seems to be reasonable. By combining Eqs. (38a) and (43) we suggest the following onset position relation:

$$x_c = 167.54 \left( 1 + 0.02 Pe^{1.10} \frac{1 + 2 Re}{R_d} \right), \quad (44)$$

in comparison with the present experimental data. As shown in Fig. 8, Eq. (44) represents the present experimental data quite well.

5. Conclusion

For the laminar forced convection in the porous layer, the effects of the inertia and dispersion on the onset condition of the buoyancy-driven motion have been investigated. We employed the linear stability theory on the Forchheimer flow model and also conducted the electrochemical mass transfer experiments. It was found that the inertia and dispersion effects make the system stable. The dispersion effects are determined for the region of $Re_K \gg 0.1$ effectively. It is expected that the new stability criteria including the inertia and the dispersion effects is useful for the application on various scientific and engineering systems.
References