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An experimental study on buoyancy-driven convective mass transfer from spheres embedded in saturated porous media

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Abstract Buoyancy-driven convective mass transfer coefficients from copper spheres embedded saturated porous media have been experimentally obtained. Limiting diffusion current technique based on cathodic reduction of cupric ions is used. The data correspond to high Rayleigh number and, expectedly, exhibit non-Darcian effects. It is found that mass transfer from the spheres embedded in saturated porous media can be predicted by $Sh_D^* = 5.609 Ra_D^{*0.241} Da^{-0.214}$, where Ra_D^* and Da are modified Rayleigh number and Darcy's number respectively.

List of symbols

A	mass transfer area
C_1, C_2	constants in Eq. (8)
C_b	bulk concentration
D	sphere diameter
Da	Darcy's number, Eq. (10)
D_e	effective diffusivity, Eq. (2)
D_M	molecular diffusivity of cupric ions in acidic solution
d_p	particle diameter
F	Faraday's constant
g	acceleration due to gravity
Gr_D	average Grashof number, $gd_p^3(\varrho/\varrho_0 - 1)/\nu^2$
I_L	limiting current
K	permeability, Eq. (3)
k_D	average mass transfer coefficient
Ra_D	average Rayleigh number $ScGr_D$
Ra_D^*	average modified Rayleigh number, Eq. (7)
Sc	Schmidt number, ν/D
Sh_D	average Sherwood number estimated with molecular diffusivity, $k_D D/D_M$
Sh_D^*	average Sherwood number estimated with effective diffusivity, $k_D D/D_e$
z	number of electrons in reduction reaction

Greek symbols

ϱ	bulk density of the solution
ϱ_0	density of the solution at the surface
ε	porosity
ν	kinematic viscosity
τ	tortuosity

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1 Introduction

Buoyancy driven convection from spheres suspended in free fluid has constantly attracted the attention of numerous researchers [1–6]. Nevertheless, the identical transport problem in porous media has received relatively less attention despite of its importance in many areas such as petroleum geology, geophysics, hydrology, catalytic reactions, nuclear waste disposal. The steady state flow and heat transfer from a sphere embedded in saturated porous media was studied by Yamamoto [7]. This theoretical work presented asymptotic solutions for velocity and temperature fields for small Rayleigh numbers. Pop and Ingham [8], utilizing boundary layer approximation, predicted steady state heat transfer for higher Rayleigh number. In an effort to generalize the results, Merkin [9] investigated heat transfer from axi-symmetrical surface of arbitrary profile. Free convective boundary layer equations were solved leading to an analytical expression for heat transfer. Steady state similarity solution of boundary layer equations for sphere, which is a special case of axi-symmetrical body, was given by Cheng [10]. Nilson [11] used Mangler's transformation to analyze boundary layers on axi-symmetrical surface of bodies of revolution. Sano and Ohihara [12] proposed a model but with the present case for small Rayleigh number.

Natural convection from spheres situated in porous media was studied under various conditions, namely; non-Newtonian fluid by Chen and Chen [13], mixed convection by Cheng [14], coupled heat and mass transfer by Kumari and Nath [15] and Lai and Kulack [16], transient heat transfer by Pop et al. [17], Ganapathy and Purushothaman [18], Ganapathy [19] and Ganapathy [20]. Recently, Paik et al. [21] studied transient conjugate mixed convection for pure/saline water.

All of these works are theoretical and assume applicability of Darcian flow model. It has been demonstrated that for vertical surface embedded in porous media the Darcy's flow-model does not hold at higher Rayleigh number and larger packing particles [22]. In the absence of validation with experimental data, the applicability of the Darcian flow and similarity solution is questionable for the present hydrodynamics.

The objective of this work is to obtain average mass transfer coefficients from spheres embedded in saturated porous media. The limiting diffusion current technique based on reduction of cupric ions has been used. At limiting current the surface concentration is constant at approximately zero value. This situation is identical to steady state heat transfer from isothermal spheres. The results will be compared with the prediction of the available steady state models for isothermal spheres. Finally the data are correlated to give a relation valid for non-Darcian conditions.

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Experimental

The experimental set-up used for this study is shown in Fig. 1. The packed bed was formed in a one liter cylindrical vessel with five different packing materials; namely, glass spheres of 16, 6, 4, 3 and 2 mm diameters. Porosities of the packing materials were determined by water replacement method. Copper reduction from acidic cupric sulfate solution was chosen to estimate the limiting diffusion current. This system was selected for its well-defined limiting current plateau [23]. A solution of 0.618 M $CuSO_4$ and 3.09 M H_2SO_4 was prepared. Concentrations of Cu^{++} and H_2SO_4 were determined through idiometric and acid-base titration respectively.

Four copper spheres of 0.98, 1.51, 1.98 and 2.59 cm diameters were fabricated. A thin copper rod was welded on each sphere for electrical connection. The rod was painted with an insulating paint. The surface of the spheres was prepared by application of increasing grades of emery papers (100, 400, 600, 1500 grit size) and finally by washing with acetone to remove any oil/grease. The prepared assembly was embedded in the packing material to act as the working electrode. A copper cylinder, kept inside the bed, near to the wall, worked as the counter electrode. A saturated calomel electrode was embedded in the bed to function as a reference electrode. The temperature was measured by a thermometer of 0.1°C least count. The working, counter and reference electrodes were connected to a potentiostat (Model 273A, EG&G PARC). The potentiostat was driven by a software (Model 352, EG&G PARC) via an IBM486 computer. Potentiostatic linear polarization and chronoamperometric curves were obtained for samples spheres embedded in different packing materials and while suspended in free solution.

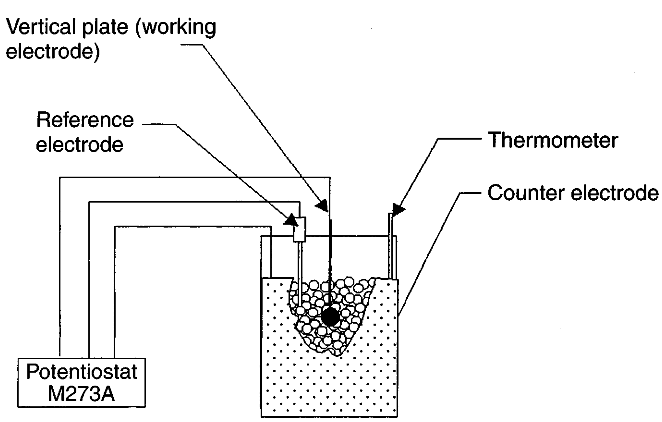

Fig. 1. Experimental set-up for measuring limiting current

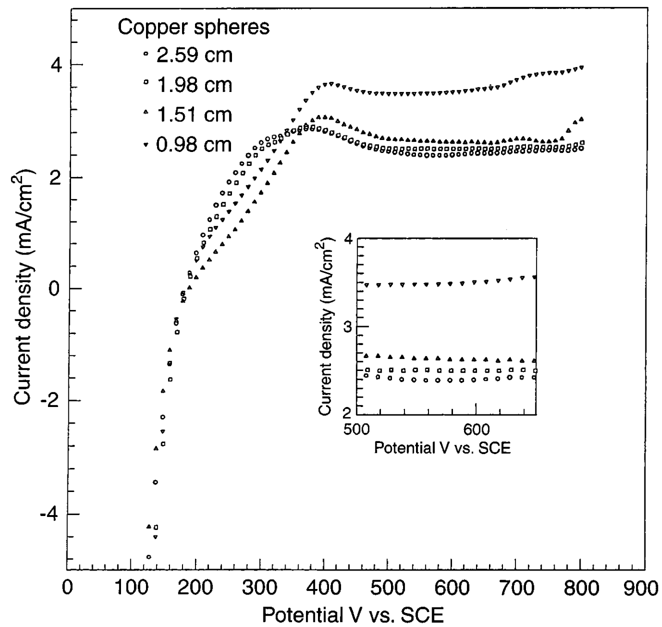

Fig. 2. Potentiostatic polarization curves of copper spheres in free solution

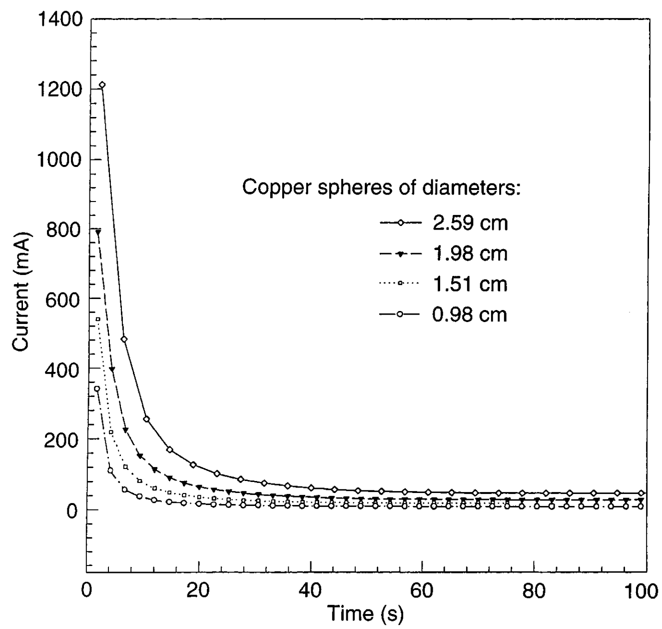

Fig. 3. Chronoamperometric curves of different copper spheres suspended in free solution at 550 mV SCE

3
Data reduction

Typical polarization curves obtained for spheres of different size immersed in free solution are shown in Fig. 2. Similar curves were obtained when spheres were embedded in porous media. These curves exhibit pronounced limiting current plateau between 500 to 650 mV SCE. Chronoamperometric curves from these spheres at potential 550 mV SCE are shown in Fig. 3. These curves indicate that steady state was reached in less than 45 seconds. The limiting current determined from these curves is used to calculate the averaged mass transfer coefficients (k_D):

$$k_D = \frac{I_L}{zFAC_b} \tag{1}$$

The physicochemical properties of the acidic cupric sulfate solution which are required in estimation of the Sherwood and Rayleigh numbers, were estimated from temperature dependent empirical correlation given in reference [24]. Density of the solution at the surface (ϱ_0) is also needed to evaluate Rayleigh number. Its evaluation necessitates estimation of concentration of H_2SO_4 and $CuSO_4$ at the surface. For mass transfer controlled regime, the surface concentration of $CuSO_4$ can be taken as zero while concentration of H_2SO_4 is calculated by utilizing principle of electroneutrality and the correlation given in reference [24].

The effective mass diffusivity for a given porous media is estimated from the molecular diffusivity by following relation:

$$D_e = \frac{D_M \varepsilon}{\tau} \tag{2}$$

where, τ is the tortuousity of the porous media. For regular packing materials its recommended value is 4 [25]. The permeability of the media is estimated using Kozeny's equation:

$$K = \frac{d_p^2 \varepsilon}{180(1-\varepsilon)^2} \tag{3}$$

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Results and discussion

The ranges of various parameters used in this study are given in Table 1. The data correspond to high Rayleigh numbers. The average mass transfer coefficients estimated from Eq. (1) are plotted against sphere diameter (D) in Fig. 4. Average mass transfer coefficients decrease with increasing sphere diameter, conforming to the theory of natural convection from spheres in free fluid. For a given size of sphere, the mass transfer coefficient increase with increasing size of the packing particles. The value approaches asymptotically to the values of free fluid.

In Fig. 5 the average Sherwood number is plotted against Rayleigh number for free fluid. Experimental data match well with various theoretical/empirical equations, for example, Acrivos [1], Steinberger [3] and Raithby [4]. This comparison establishes the applicability of the technique and accuracy of correlation for physicochemical properties. Identical plots for various packing particles are also shown in Fig. 5. It can be observed that for a given Ra_D, Sh_D increases with particles size of the packing spheres, asymptotically approaching the values for free solution.

Yamamoto [7] assumed Darcy's flow model and obtained uniformly valid asymptotic solution for temperature field. In this present notations, corresponding to mass transfer, the model equation can be written as:

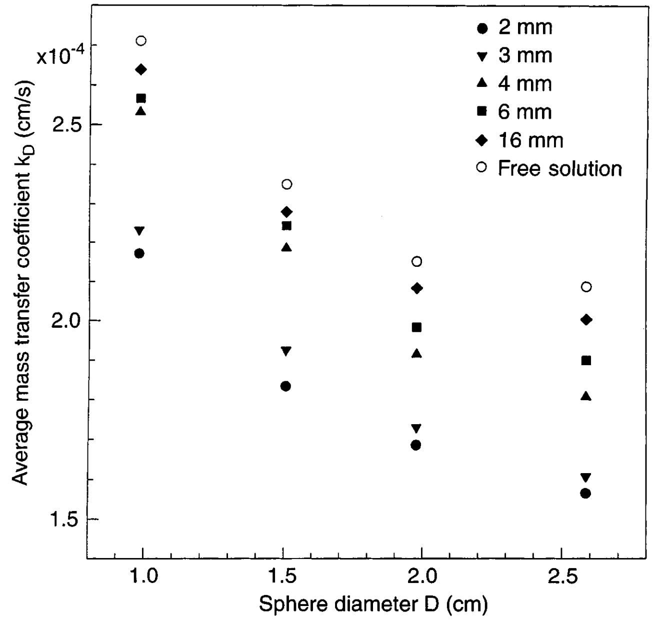

Fig. 4. Average mass transfer coefficient versus diameter of sphere for different packing particles

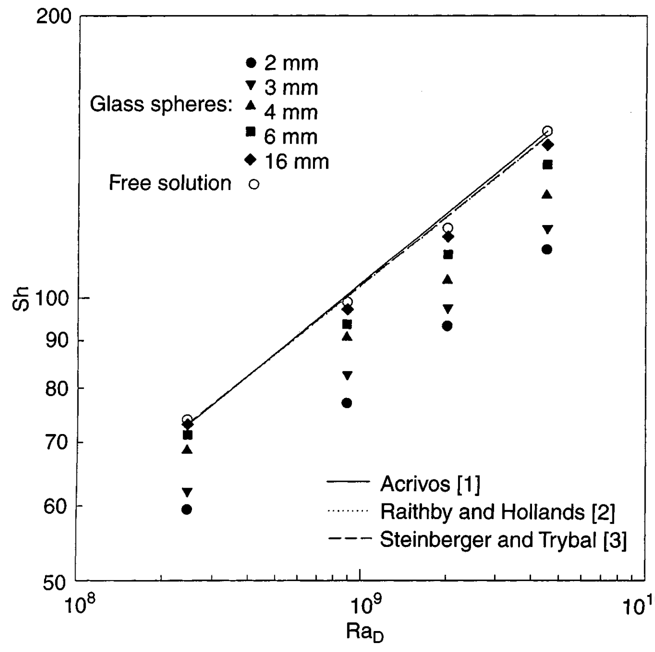

Fig. 5. Average Sherwood number versus Rayleigh number plots for different packing particles

Table 1. Range of various parameters in this study

Parameter	Range
Diameter of the spheres	0.98–2.59 cm
Packing particle diameter	2.0–16.0 mm
Temperature	22.0–23.6°C
Porosity	0.371–0.417
Permeability	1.03×10^{-6}–3.38×10^{-3} cm^2
Sc	3953.4–4242.6
Ra_D	2.45×10^8–4.51×10^9
Ra_D^*	9.39×10^4–2.22×10^7

$$Sh_D^* = \frac{2\tau}{\varepsilon} + \frac{602\varepsilon\left(Ra_D^*\right)^2}{315\tau} \quad (4)$$

This model is valid only for smaller Ra_D^* and therefore do not match with the present data. For Darcian flow model, Merkin [9] analyzed free convection boundary layer on axi-symmetric bodies in saturated porous media. Similarity solution of energy and momentum equations was presented leading to a relation for local heat transfer. Cheng [10] obtained similarity solution for sphere, which is specific case of Merkin's analysis. The result can be written in the non-dimensional form as follows for mass transfer:

$$Sh_D^* = 0.362 Ra_D^{*\frac{1}{2}} \quad (5)$$

where, Sh_D^* is the Sherwood number calculated using effective diffusivity:

$$Sh_D^* = \frac{k_D D}{D_e} \quad (6)$$

and, Ra_D^* is the modified Rayleigh number given as:

$$Ra_D^* = \frac{gKD(\varrho/\varrho_\infty - 1)}{\nu D_e} \quad (7)$$

In Fig. 6 experimental data are plotted as Sh_D^* versus Ra_D^* for each size of packing particles. In this plot, the effect of the size of the packing particles is apparent. The average Sherwood number (Sh_D^*) is higher for smaller packing particles for a given value of Ra_D^*. Equation (5) represent one single straight line and does not indicate dependance on the size of packing particles. This is due to the assumption of Darcy's flow model in its derivation. For embedded spheres, models that include non-Darcian effects are non-existent. Albeit, for vertical surface embedded in saturated porous media several mathematical works include non-Darcian effects. Rahman et al. [22] have compared these models with the experimental data and concluded that the model due to Kim and Vafai [26] that utilizes Brinkman-extended Darcy's flow model, predicted Sh_D^* close to the observed values. According to this model, Sh_D^* is proportional to one-fourth power of the ratio of modified Rayleigh number and Darcy's number. Since the transport at vertical surface and sphere is identical except the geometry, Kim and Vafai's model can be applied but with different constants. Therefore an equivalent relation for the present case can be written as;

$$Sh_D^* = C_1 \left(\frac{Ra_D^*}{Da}\right)^{C_2} \quad (8)$$

where Da is Darcy's number based on particle diameter:

$$Da = \frac{K}{\varepsilon D^2} \quad (9)$$

Equation (8) is plotted in Fig. 6 for comparison. The model equation exhibit reasonable match with the experimental data with a constant of value of $C_1 = 3.8$ and $C_2 = 0.25$. However, some deviation can be observed that gets pronounced for smaller packing particles. Through some empirical trials, it was found that the data fall on one single straight line when Sh_D^* is plotted against $Ra_D^* Da^{-0.89}$, as shown in Fig. 7. The linear regression analysis results into following equation with $R^2 = 99.3\%$:

$$Sh_D^* = 5.609 Ra_D^{*0.241} Da^{-0.214} \quad (10)$$

The plots of Eq. (10) for all packing particles in Fig. 6 exhibit a better match with the experimental data.

5 Conclusion

Mass transfer coefficients from copper spheres embedded in spherical glass particle forming the porous media sat-

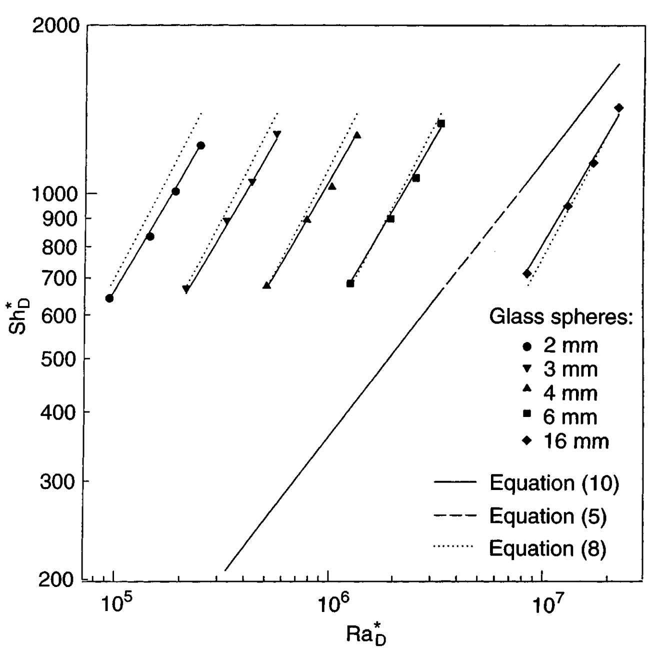

Fig. 6. Variation of Sherwood number with modified Rayleigh number for different packing particles

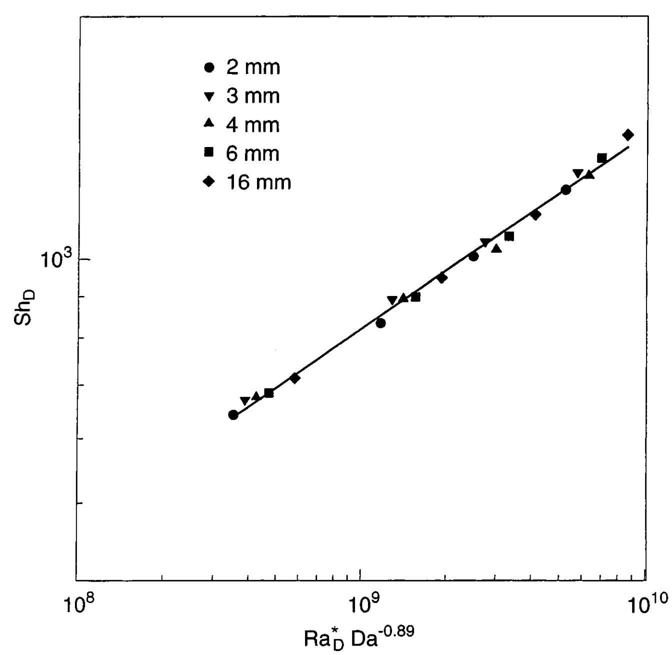

Fig. 7. Average Sherwood number versus $Ra_D^* Da^{-0.89}$ curves for spherical particles

urated with liquid were obtained experimentally using limiting diffusion current technique based on cupric ion reduction in acidic electrolyte. Average Sherwood numbers were calculated and data were compared with existing mathematical models. Model based on simple Darcy's flow fail to predict mass transfer coefficients. In the absence of a non-Darcian model for sphere, results of Kim and Vafai's model is used to obtain Ra_D^* and Da as correlating parameters resulting into Eq. (10).

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