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EXPERIMENTAL STUDY OF FREE CONVECTIVE MASS TRANSFER IN POROUS MEDIA

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Abstract—Free convective mass transfer rates in packed beds of spheres and screens were measured by the electrochemical method. It was not possible to correlate the results in the usual manner employed in the analogous heat transfer problem, i.e. Nu or $Sh = f(Re)$. The Sherwood number was correlated as follows:

$$Sh = 0.228\,(ScGr)^{0.32}\,(R_h/d_p)^{0.22}.$$

NOMENCLATURE

c_0, bulk concentration [mol/m^3];
D, diffusivity [m^2/s];
d_b, bed diameter [m];
d_p, particle diameter [m];
g, acceleration of gravity [m/s^2];
Gr, Grashof number $= g\alpha c_0 L^3/\nu^2$;
k, mass transfer coefficient [m/s];
L, depth of packed bed [m];
P, permeability of the porous bed [m^2];
Ra, Rayleigh number $= g\alpha c_0 LP/\nu D$;
Sc, Schmidt number $= \nu/D$;
Sh, Sherwood number $= kL/D$.

Greek symbols

α, densification coefficient [m^3/mole];
ε, void fraction;
ρ, density [kg/m^3];
ν, kinematic viscosity [m^2/s];
φ, specific area of porous bed [m^{-1}].

INTRODUCTION

NATURAL convection in porous media has received considerable attention in the last few years. The convective "heat transfer" behaviour of permeable beds is of increasing interest in the study of geothermal systems, potential alterations in nuclear power reactors and the storage of highly radioactive waste materials. An excellent survey has been published by Hardee and Nilson [1] and Horne and O'Sullivan [2].

However scarce information is available on free convection "mass transfer" in porous media or packed beds. Its knowledge may be of interest in the interpretation of experimental results obtained in various fields:

Free convection may become an important effect in studies of low Péclet number mass transfer in packed beds [3]. This is so, because Sherwood numbers of the same order of magnitude are obtained both in pure natural convection and in flowing fluids.

Several works [4, 5] dealing with the hydrodynamic effects in porous flow-through electrodes working at low flow rates have neglected natural convection, even though it has been shown that free convection is still effective in microcells [6] and narrow spaces [7, 8, 9, 10].

As far as we know, the only data available on natural convection mass transfer coefficients in packed beds are those of Mandelbaum [11] and Karabelas *et al.* [12], but in both cases only single particles of the bed were active. The former worked with Raschig rings, the latter with spheres. Different results should be expected when working with several particles, all of them undergoing a mass transfer process. Accordingly, the present work was undertaken to experimentally determine the mass transfer characteristics for natural convective flow in packed beds. Mass transfer coefficients were determined by the well known electrochemical method. The geometry considered was a porous bed with a liquid layer on top and beneath it, so that the fluid could flow freely in and out of the bed.

EXPERIMENTAL DETAILS

Apparatus

Cells, consisting of cylindrical lucite tubes, were placed in a thermostatic bath, where the temperature was regulated to 25 ± 0.1°C.

It is recognized that wall effects influence low Péclet number mass transfer in packed beds [13]. Consequently, in order to investigate the influence on mass transfer in free convection, two cells of 5 and 10 cm internal diameter and 30 cm height, were constructed. The active porous bed was located at 8 cm from the bottom of the cell.

It is worth mentioning that the aspect ratio (L/d_b) had no influence on the overall mass transfer characteristics, as it was also stated by Hardee and Nilson [1] in their heat transfer studies. These authors worked with wide beds and narrow cylindrical geometries.

Porous media

The porous medium, working as the cathode, was a packed bed either of spheres or of screens.

The spheres were glass beads, initially nickel-plated by autocatalytic deposition and then electrochemically. Only beds of one, two and three layers of spheres were used to ensure limiting current conditions in the whole bed, since the potential drop which builds up in the bed, increases with bed height and hydrodynamic mass transfer through it [14]. In every case a nickel-plated bronze mesh ($d_p = 0.55$ mm ; $\varepsilon = 0.90$) served as support for the beads and simultaneously as current feeder. It was connected to the external circuits through thin cable wires which left at the top of the cell.

The screens were nickel-plated bronze gauzes and in some case up to 5 screens were pressed together to form the packed bed.

In all experiments the counter-electrode consisted of a wire gauze packing, prepared by rolling inox wire cloth, positioned in the upper part of the cell.

The parameters characterizing the geometry of the different beds are disclosed in Table 1. Special care was taken in the determination of particle diameter and bed porosity, since these parameters directly influence the calculation of the specific surface φ, the permeability P and the hydraulic radius R_h.

The mean diameter of the spheres was determined both, from their weight and density and using a micrometer. The mean porosity for each packed bed was obtained by imbibition of water and also from the volumes of the spheres and of the packed bed.

The geometrical characteristics of the screen beds were determined by the method outlined by Blass [15]. Although this method was reported for single screens only, it has been shown [16] that it can be applied with sufficient accuracy to packed beds of screens.

Electrolyte

In almost every case equimolar solutions of potassium ferri- and ferrocyanide, 2 M in sodium hydroxide were used. The electrolytic solution employed by Appel and Newman [3] in their study of low Péclet mass transfer in packed beds was also tested. This solution used potassium nitrate as supporting electrolyte. The properties of the solutions are summarized in Table 2.

Procedure

Special care was taken in all experiments in saturating the electrolyte with nitrogen, blocking off the cell from light exposure and activating the electrodes. Polarization curves were also determined in all runs.

Preliminary measurements were carried out to obtain the limiting current for the feeder-screen. In this case the screen was covered by inert spheres of the same size as those to be investigated in the packed bed. This allowed for the correction of the measured cell current by subtracting the feeder component.

Once the steady state was reached (always within 5–10 min), the limiting current was measured for the

Table 1. Geometric characteristics of porous beds

Symbol	Number of layers or screens	d_p (cm)	ε	L (cm)	φ (cm^{-1})	R_h (cm)	$P \times 10^3$ (cm^2)
Spheres							
●	1	0.975	0.544	0.975	2.81	0.194	4.08
	2	0.975	0.546	1.85	2.79	0.195	4.18
	3	0.975	0.526	2.60	2.92	0.180	3.38
◑	1	0.60	0.501	0.60	4.99	0.100	1.00
	2	0.60	0.476	1.00	5.24	0.091	0.79
	3	0.60	0.455	1.50	5.45	0.083	0.64
○	1	0.50	0.537	0.50	5.56	0.097	1.01
	2	0.50	0.515	0.90	5.82	0.088	0.81
	3	0.50	0.496	1.35	6.05	0.082	0.67
Screens							
△	1	0.198	0.81	0.52	3.84	0.211	4.83
	2	0.198	0.81	1.00	3.84	0.211	4.83
	3	0.198	0.81	1.50	3.84	0.211	4.83
	4	0.198	0.81	2.00	3.84	0.211	4.83
▲	1	0.08	0.85	0.175	7.50	0.113	1.19
	2	0.08	0.85	0.304	7.50	0.113	1.19
	3	0.08	0.85	0.529	7.50	0.113	1.19
	4	0.08	0.85	0.636	7.50	0.113	1.19
	5	0.08	0.85	0.752	7.50	0.113	1.19
▽	1	0.0375	0.84	0.084	17.07	0.050	0.192
	2	0.0375	0.84	0.165	17.07	0.050	0.192
	3	0.0375	0.84	0.260	17.07	0.050	0.192
	4	0.0375	0.84	0.330	17.07	0.050	0.192
	5	0.0375	0.84	0.410	17.07	0.050	0.192

* For both packings the permeability was calculated as described by Happel and Brenner [17].

Table 2. Electrolyte properties at 25°C

Electrochemical system	ρ (g/cm^3)	μ (cp)	$D \times 10^6$ (cm^2/s)	α* (cm^3/mol)
0.01 M $K_3Fe(CN)_6/K_4Fe(CN)_6 + 1$ M KNO_3	1.0580	0.868	5.84	46
0.05 M $K_3Fe(CN)_6/K_4Fe(CN)_6 + 2$ M NaOH	1.0949	1.394	5.34	16.8
0.10 M $K_3Fe(CN)_6/K_4Fe(CN)_6 + 2$ M NaOH	1.1174	1.494	4.98	11.75

* The coefficient was obtained as indicated in reference [18].

different electrolytic solutions to yield the mass transfer coefficient for the packed beds investigated.

CORRELATION OF DATA

Based on the relationships for natural convection heat transfer in porous media mentioned in the literature, the experimental results were correlated in terms of Sherwood number vs. Rayleigh number. Figure 1 shows the experimental data together with empirical and analytical correlations obtained by other authors for bottom heated and volume heat configurations. The wide scattering and net separation of the experimental points suggest that this type of plotting is not suitable for the mass transfer phenomenon investigated. Consequently, the experimental results were correlated using another dimensionless equation, derived by the following reasoning.

Natural convection cannot develop freely in a packed bed because of the neighbouring particles. This resembles the mass transfer in narrow spaces [7, 8, 9, 10] where the Sherwood number is a function of the Schmidt and Grashof numbers and of a geometric ratio. This ratio takes into account the width and the length of the transferring gap.

For packed beds the characteristic dimension of the flow cross-section will be the hydraulic radius, R_h. The particle diameter d_p will, without doubt, influence mass transfer, because the diffusion layer will grow along the particle, break down and grow again on the following particle. So, the geometric ratio entering the correlation should be R_h/d_p and the Sherwood number will be a functional relationship given by the product of powers of the $ScGr$ group and that geometric ratio.

The following equation was obtained from the experiments:

$$Sh = 0.228\,(Sc.Gr)^{0.32}\,(R_h/d_p)^{0.22},$$

$$6.24 \times 10^3 \leqslant Sc.Gr \leqslant 3.03 \times 10^8,$$

$$0.152 \leqslant R_h/d_p \leqslant 1.412,$$

with a mean deviation of 9.88%.

This correlation is drawn in Fig. 2 together with the experimental points and seems to describe adequately the free convection mass transfer in porous media, bearing in mind the widely different packings investigated.

CONCLUDING REMARKS

Free convective mass transfer in porous media was studied experimentally for several bed thicknesses and particle diameters. All the other geometric parameters of the bed were also varied by using different packings.

A new dimensionless equation has been proposed, which improves the correlation of data with respect to the usual relationship employed in the analogous heat transfer studies.

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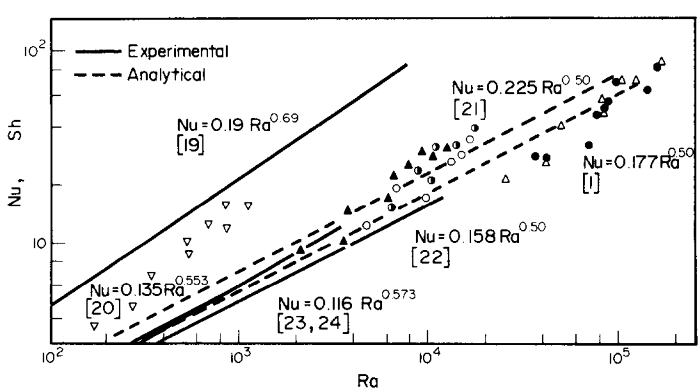

FIG. 1. Dependence of porous bed heat and mass transfer on Rayleigh number. Symbols as listed in Table 1.

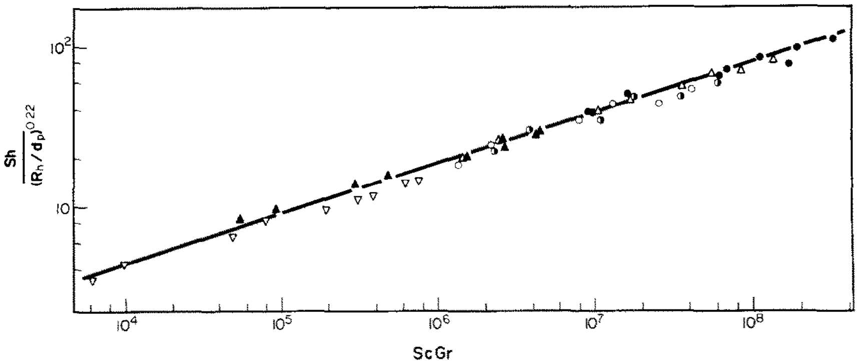

FIG. 2. Dimensionless representation of the natural convective mass transfer in packed beds. Symbols as listed in Table 1.

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ETUDE EXPERIMENTALE DE LA CONVECTION NATURELLE DE MASSE DANS LES MILIEUX POREUX

Résumé—La convection massique naturelle dans des lits fixes de sphères est mesurée par la méthode électrochimique.

Il n'a pas été possible de rassembler les résultats d'une manière analogue à celle relative au transfert de chaleur, c'est-à-dire Nu ou $Sh = f(Ra)$. Le nombre de Sherwood est exprimé par: $Sh = 0{,}228\ (Sc.Gr)^{0.32} \times (Rh/d_p)^{0.22}$.

EINE EXPERIMENTELLE STUDIE ZUM FREI KONVEKTIVEN

Zusammenfassung—Der Stoffübergang durch freie Konvektion in Festbetten aus Kugeln und Gittern wurde nach der elektrochemischen Methode gemessen. Es war nicht möglich, die Ergebnisse in der üblichen Art wie bei dem analogen Wärmeübergangsproblem darzustellen, d.h. Nu oder $Sh = f(Ra)$. Die Sherwood-Zahl wurde wie folgt wiedergegeben:

$$Sh = 0{,}228\,(ScGr)^{0.32}\,(R_h/d_p)^{0.22}.$$

ЭКСПЕРИМЕНТАЛЬНОЕ ИССЛЕДОВАНИЕ СВОБОДНОКОНВЕКТИВНОГО МАССОПЕРЕНОСА В ПОРИСТЫХ СРЕДАХ

Аннотация — Электрохимическим методом измерена интенсивность массопереноса при свободной конвекции в плотных слоях из сферических частиц и сеток. Используемая в аналогичной задаче по теплопереносу зависимость, Nu или $Sh = f(Ra)$, оказалась непригодной для описания полученных результатов. Предложено следующее соотношение для числа Шервуда:

$$Sh = 0{,}228(ScGr)^{0.32}(R_h/d_p)^{0.22}.$$