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LETTERS TO THE EDITOR

Discussion of "Double diffusive natural convection in a vertical rectangular enclosure—I. Experimental study"

THE AUTHORS present the results of a series of carefully conducted double-diffusive experiments in a tank with simultaneous horizontal gradients of temperature and concentration [1]. The occurrence of a row of convection cells in the experimental tank was clearly exhibited by the use of Schlieren photography.

The experiments were carried out using an electrochemical system with a $CuSO_4$–H_2SO_4–H_2O solution. The presence of H_2SO_4 is necessary to prevent ionic migration down the electrical potential gradient. The concentration of $CuSO_4$ ranges from 0.015 to 0.05 mol l^{-1} (1.2–4.0 g l^{-1}), and that of H_2SO_4 is 1.5 mol l^{-1} (75 g l^{-1}). With such a large concentration of H_2SO_4 (compared to that of $CuSO_4$) in the solution, any nonuniformity in its distribution may generate a triple-diffusive situation. All available evidence points to such a conclusion.

Wilke *et al.* [2] state that "cathode reaction and migration effects result in a net movement of sulfuric acid toward the interface". Selman and Newman [3] calculated the mass transfer and shear stress along a vertical electrode in a $CuSO_4$–H_2SO_4 solution by using the similarity method to solve the boundary layer equations. They found that the concentration increase of H_2SO_4 at the cathode is 45% of the bulk concentration of $CuSO_4$. (Using the method proposed by Wilke *et al.*, Selman and Newman obtained a value of 35%.) This increase of H_2SO_4 concentration was shown to reduce the convective velocity by approximately 10%. Eventually, this more concentrated solution will be brought by the flow around the corner to lay on top of the bulk fluid, and vice versa at the opposite corner. This process continues throughout the entire duration of the experiment.

According to Cole and Gordon [4], the diffusivity of $CuSO_4$ in an aqueous solution of H_2SO_4 is approximately 6×10^{-6} cm^2 s^{-1} for a 0.31 equivalent of copper sulfate in a 1.0 equivalent of sulfuric acid. The diffusivity of H_2SO_4 is reported by James *et al.* [5] to be approximately 2×10^{-5} cm^2 s^{-1}, more than three times as large as that for $CuSO_4$. Together with thermal diffusion ($\sim 10^{-2}$ cm^3 s^{-1}), a triple-diffusive system is at hand. We note here that a sugar and salt solution with diffusivities that differ by a factor of 3 is a common medium for double-diffusive experiments. With H_2SO_4 participating in the diffusive convective process, the concentration measurements made by light attenuation techniques may be in error because the background concentration of H_2SO_4 can no longer be assumed to be constant.

There is no doubt that the electrochemical system exhibits a host of interesting flow phenomena. However, in order for us to understand the mechanisms responsible for the occurrence of such phenomena, the role played by $CuSO_4$ and H_2SO_4 must be correctly taken into account.

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Author's Reply

AS WAS pointed out by Professor C. F. Chen, the present experiment should be considered as a triple-diffusive convection, which should include the hydrogen transfer in addition to the thermal and cupric-ion transfers in a strict sense. However, it can be approximated by a double-diffusive convection by combining the hydrogen ion diffusion and the cupric ion diffusion into a single solutal diffusion based on the following facts.

(1) The solutal density change in the present study has included both the density changes by Cu^{2+} gradients and H^+ gradients

$$\rho = \rho(T, C, H(C)) \tag{1}$$

$$\left.\frac{\partial \rho}{\partial C}\right|_T = \left.\frac{\partial \rho}{\partial C}\right|_{T,H} + \left.\frac{\partial \rho}{\partial H}\right|_{T,C} \cdot \left.\frac{\partial H}{\partial C}\right|_T \tag{2}$$

where T is the temperature, C the Cu^{2+} ion concentration, and H the H^+ ion concentration. The H and C are related to the ratio of the mass transfer coefficients of cupric sulfate to that of sulfuric acid, which is flow dependent [1]. As Professor Chen pointed out, buoyancy decreases approximately 10% due to the H^+ contribution (the second term on the right-hand side in equation (2)).

(2) The diffusivity ratio of hydrogen ions to cupric ions is roughly 3.3 [2]. The ratio of boundary layer thicknesses would be approximately 1.3 according to the laminar boundary layer analysis. The ratio is, however, significantly small compared to the ratio with the thermal boundary layer thickness.

$$\frac{\delta_H}{\delta_{Cu}} = \left(\frac{D_H}{D_{Cu}}\right)^{0.25} = 1.3$$

$$\frac{\delta_T}{\delta_{Cu}} > 6 \quad [1].$$

Denpo *et al.* [3] actually measured the cupric ion concentration and hydrogen ion concentration distributions near vertical walls using two-wavelength holographic interferometry. The boundary layer thicknesses of the cupric ion and hydrogen ion did not show any appreciable difference. (Both were approximately 0.5 mm.)

(3) Previous studies [4, 5] indicate that the Rayleigh number is simply additive, if one buoyancy is significantly larger than the other and/or if the ratio of diffusivities is close to unity. According to Nilson and Baer [6], the counter-flow at buoyancy ratio of 0.1 and diffusivity ratio 3 falls well within the inner-dominated flow region. The fluid flow can be assumed to be in the simply-additive region [7] and a single solutal Rayleigh number can be used.

(4) The concentration increase of H_2SO_4 at the cathode is approximately 35–45% of the bulk concentration of $CuSO_4$ [8, 9]. In the present experiment, the concentration of $CuSO_4$ is in the range of 0.015–0.05 mol l^{-1}, and it creates a concentration increase of H_2SO_4 by 0.005–0.02 mol l^{-1}, which is considerably smaller than the nominal concentration of H_2SO_4 in the bulk fluid (1.5 mol l^{-1}). The concentration variation of hydrogen ion in the cavity due to the migration effect would be between 1.48 and 1.52 mol l^{-1} at worst.

(5) The triply-diffusive analysis could be suggested for future studies for further refinements in the experimental technique.

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Comments on "Coupled heat and mass transfer by natural convection from vertical surfaces in porous media"

I AM VERY pleased that this new full-length paper [1] confirms our scale-analysis predictions and extends the range covered by our similarity solutions [2]. However, there is one erroneous claim in this new paper, and I must correct it.

On page 1192, the authors state that "Contrary to what has been reported by Bejan and Khair (ref. [2] below), we have found … that solutions in the range of $N < -1$ are impossible." In other words, the solutions reported by us [2] for the negative buoyancy ratios $-5 \leqslant N \leqslant -1$ are fictitious. They explain this claim in the footnote: "Although Bejan and Khair did not state clearly in their paper, the solutions they presented for the range $-5 \leqslant N \leqslant -1$ actually corresponded to a different problem, for which the convective flow is always downward, such that the parallel double boundary-layer structure is maintained."

Lai and Kulacki are wrong. I draw attention to page 913 in our paper [2] in which we pointed out that: "The only change that occurs when N is negative and large in absolute value is that the double boundary layers sketched in (our) Figs. 1 and 2 develop in the negative y direction, since the flow will then be downward." Obviously, our solutions for $-5 \leqslant N \leqslant -1$ are correct, and we explained their physical meaning quite clearly.

Recognition of the possibility of two flow directions, or the condensation of two problems into a unified treatment (for brevity, among other virtues) was one of the central ideas of our paper. We emphasized this by using $|N|$ instead of N, wherever appropriate. In our drawing of the double boundary layer structure (Fig. 2) we did not indicate 'up' or 'down': note that y, the y arrow and the gravity vector are missing.

As an aside, I note that Lai and Kulacki did not mention ref. [3], which is where this problem was first formulated and solved by scale analysis.

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