m = particle mass flux, kg m^{-2} s^{-1}
r = radial position, m
w = argon mole fraction
X^* = core length, m/m
x = axial position, m
1, 2, 3, 4, 5 = test condition

Subscripts

1 = primary jet
c = center line
d = mixing chamber duct
m = completely mixed state

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Magnetically Enhanced Thermal Convection in Oxygen Gas

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The effects of static magnetic fields on heat transfer in oxygen gas have been measured at temperatures between 78 and 112°K and pressures between 2.01 and 4.77 kN/m². After the data are corrected for ordinary thermal conductivity behavior, comparison with theory suggests that the phenomenon is magnetically enhanced thermal convection.

SCOPE

When a magnetic field is applied to a dilute polyatomic gas transporting energy primarily by thermal conduction, the normal response is a small thermal conductivity decrease in the order of 1%. A much different result was found for oxygen gas at low pressures and temperatures near 80°K. The apparent thermal conductivity change was much larger than normal and opposite in sign.

This paper presents the results of a parametric study of the experimental variables in this new phenomenon. The objectives are to identify which variables are dominant, to determine their correlation with the observed effects, and to compare the results with existing theory.

A method is developed here for analyzing the total magnetically induced changes in heat transfer by correcting for the normal thermal conductivity effects. Relationships between the corrected data and each experimental variable are then determined. This information provides insight into the nature of the heat transfer changes which occur when the magnetic field is applied.

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Besides providing a systematic experimental study of the magnetically induced changes in heat transfer through oxygen, this work provides a quantitative test of present theories of magnetothermal convection processes. The measurements show the extent of the range of application, the accuracy of predictions, and give an indication of the directions for further theoretical development.

CONCLUSIONS AND SIGNIFICANCE

The dominant experimental variables determining the magnitude of the magnetic field effects are the gas pressure and absolute temperature, the temperature gradient, and the magnetic field strength. Corrections for the field induced changes in thermal conductivity can be estimated successfully and applied with a maximum expected error to the results in the order of 1%.

The corrected data do not show a constant power law dependence on each variable. But over a narrow range of the experimental conditions, the observed dependencies for pressure, temperature, and temperature gradient agree with those predicted from theory. In particular, the good agreement with the quadratic temperature gradient dependence strongly supports the theoretical assertion that the observed effects are caused by magnetically induced changes in thermal convection. The agreement with theory of the observed magnetic field dependence of the effect is poor, the data showing an approximately cubic field dependence as compared with the predicted linear dependence. It is unlikely that the discrepancy can be attributed to applying the theory, which was developed from a semi-infinite flat plate model, to the concentric cylinder configuration of the experimental cell.

The theoretical predictions of Clark and Honeywell (1977) agree well in absolute magnitude with the experimental results of this study. This suggests that small magnetic field gradients can be important in affecting natural convection heat transfer. Calculations from the earlier theory of Park and Honeywell (1973), which assumed only the presence of homogeneous magnetic fields, are too small by two orders of magnitude.

The data presented here provide a sensitive test for further theoretical attempts to predict the influence of magnetic fields on thermal convection in gases.

When a dilute polyatomic gas transporting energy primarily by thermal conduction is subjected to a magnetic field, the character and intensity of the energy transport change slightly. A small decrease in thermal conductivity occurs when the field is applied. This phenomenon is called the Senftleben-Beenakker Effect (hereafter abbreviated SBE). Analogous effects for mass and momentum transport also occur (Beenakker and McCourt, 1970). The kinetic theory of polyatomic gases has been quite successful in explaining the SBE phenomenon for simple polyatomic gases such as nitrogen, carbon monoxide, methane, and hydrogen, all of which exhibit similar behavior. Small deviations from the normal behavior which have been observed for oxygen (Beenakker et al., 1971; Hulsman et al., 1970) have been successfully explained (Coope et al., 1970; Maksimov, 1972).

The relative decrease in the thermal conductivity is normally of the order of 1% at saturation for most polyatomic gases. The change is uniquely dependent on the product of magnetic field strength and inverse pressure, decreasing monotonically with increasing H/p. Markedly different results were observed for oxygen near 80°K (Honeywell et al., 1972). At small values of H/p, the SBE data are quite similar to the normal behavior observed at 300°K. At higher values of H/p, however, the apparent thermal conductivity change shows a marked departure from the expected decrease. The data reverse in sign, resulting in an increase of the apparent thermal conductivity. Further, the magnitude of this change increases dramatically with increasing pressure (see curves B, C, and D of Figure 1, from Honeywell et al., 1972).

Some time ago, Park and Honeywell (1973) suggested that the observed enhancement of the heat transfer through oxygen is a convective effect. In a recent extension of that work, Clark and Honeywell (1977) provide a quantitative theory which agrees reasonably well with experiment. The phenomenon is referred to as the magnetothermal convection effect. The theoretical analysis is based on an assumed linear superposition of conductive and convective heat transfer through the gas. The additional heat energy transfer induced by the application of the magnetic field is called the magnetoconvection heat flux Q_{Mg}. The functional dependence of Q_{Mg} on experimentally controlled variables is predicted to be

$$Q_{Mg} \,\alpha\, \frac{p^2(\Delta T)^2}{\eta_0 T_0^4} H_x \frac{dH_x}{dz} \tag{1}$$

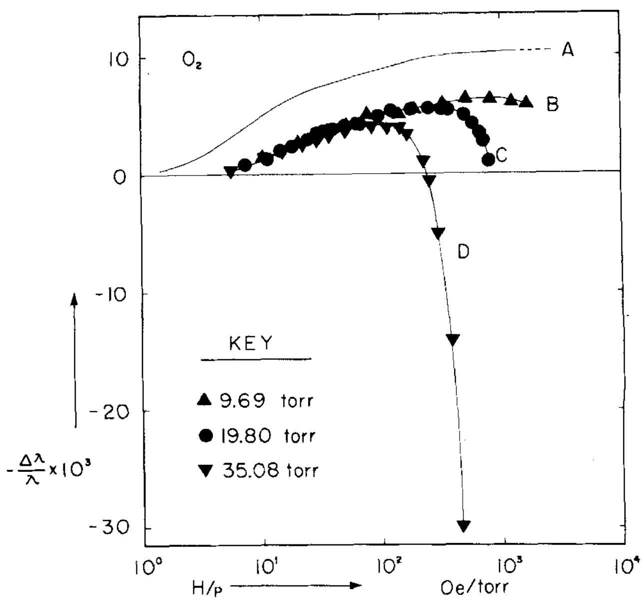

Fig. 1. Relative thermal conductivity change vs. H/p for oxygen at $T = 79°K$ and $\Delta T = 3.8°C$.

Based on the analysis of Park and Honeywell, Vevai (1973) undertook a series of measurements with a predetermined matrix of experimental conditions to determine the exponents a, b, c, and d in the functional relationship

$$Q_{Mg} \propto \frac{p^a(\Delta T)^b}{T_o^c} H^d \qquad (2)$$

An analysis of the experimental measurements and the resulting data are presented here. It is shown that the parametric dependencies suggested by previous theories are reasonably well satisfied for a limited range of experimental conditions.

EXPERIMENTAL

Apparatus

The size of the SBE thermal conductivity effect requires for adequate experimental precision an apparatus with a resolution of at least 1×10^{-5} relative change in thermal conductivity $\Delta\lambda/\lambda_o$. A differential measuring technique was employed to reach this level.

The cell is of the concentric cylinder type, a schematic of which appears in Figure 2. The outer cylinder cell wall OC is relatively massive. During the experimental measurements, the temperature of this cell wall was maintained essentially constant; a drift rate of 1.5×10^{-3} °C/hr is typical. The inner cylinder IC is a 0.305 cm OD thin walled, heated, aluminum tube constructed from aluminum foil. The sample gas is confined to the annular space between the two cylinders by a pair of vacuum tight end seals ES made from 1 mil polyester (Mylar) film. Heat energy is supplied to the inner cylinder by application of a DC current through a length of 0.025 mm diameter nickel-chromium (Evanohm) wire wrapped uniformly and bifilarly around the inner cylinder.

The temperature sensors used for the measurements are miniature strainfree platinum resistance thermometers of the DeGussa type. The primary thermometer PT1 is mounted inside the inner cylinder tube, and a secondary thermometer PT2 is attached to the cell wall. The temperature T_1 of the heated tube is measured relative to the temperature T_2 of the outer cell wall, with the thermometers in opposite arms of a shielded Wheatstone bridge.

The cell is supported in a temperature controlled, high vacuum dewar which is suspended vertically between the 9 in. diameter pole pieces of an electromagnet (Vevai et al., 1978). The magnetic field is applied in a direction perpendicular to the axis of the cell.

Measurements

As mentioned above, the temperature T_2 of the outer cylinder is kept essentially constant, and heat energy is supplied at a steady rate to the inner cylinder by passing a steady current through the heater. When a magnetic field is applied to the cell under these conditions, the change in the heat transfer through the sample gas causes a change δT_1 in the temperature T_1 of the inner cylinder.

In order to establish the functional dependence of Q_{Mg} on the variables T_o, p, ΔT, and H of Equation (2), one would prefer to alter each variable independently. However, the overall temperature difference ΔT could not be changed without simultaneously changing the average temperature T_o of the gas in the cell. Hence, when the dependence of Q_{Mg} upon ΔT was determined, compensation had to be made for the concurrent change in T_o which accompanied the change in ΔT. For the other parametric studies, H, T_o, and p were varied independently.

The experimental conditions for the measurements are given in Table 1. At each experimental condition, data were taken at several values of the magnetic field strength between 0.557 and 1.122 MA/m. The effect is negligibly small for field values smaller than those listed; upper values are limited by apparatus design. The range over which the average gas temperature T_o was varied was 78° to 112°K. Here, the smaller value is limited by the vapor pressure curve for oxygen; the upper limit represents the temperature above which the effect is too small to be accurately measured with the restrictions on the other experimental variables.

Each datum point was obtained using standard procedures for measuring SBE thermal conductivity data for gases (Vevai, 1973; Vevai et al., 1973).

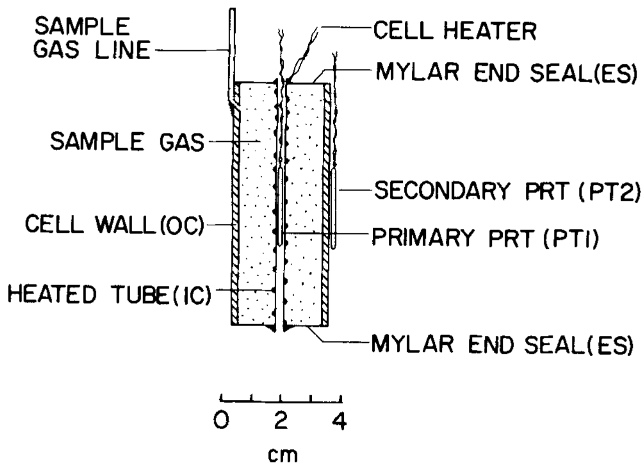

Fig. 2. Cross section of the thermal conductivity cell.

TABLE 1. EXPERIMENTAL CONDITIONS FOR OXYGEN DATA. FOR EACH SET OF CONDITIONS, DATA WERE TAKEN AT MAGNETIC FIELDS OF 0.561, 0.702, 0.844, 0.979, AND 1.22 MA/m

Experimental conditions	T_o, °K	ΔT, °C	P, kN/m²	Q_T, mW
1	77.8	0.91	4.69	1.736
2	78.2	1.80	4.71	3.485
3	79.1	3.53	4.72	6.966
4	79.8	6.74	4.73	13.93
5	81.7	12.5	4.75	27.85
6	86.1	22.5	2.01	50.15
7	85.9	22.7	2.56	51.00
8	86.3	22.6	3.73	55.88
9	86.3	22.5	4.77	56.68
10	79.1	22.6	4.69	55.48
11	98.1	22.6	4.75	59.58
12	107.9	12.4	4.71	33.74
13	112.0	22.6	4.72	65.97

Data Analysis and Results

As shown in the Appendix, the convective contribution to the total field induced change in heat energy transfer is obtained by eliminating the ordinary SBE thermal conductivity contribution at the same average gas temperature. The amount of heat transfer thereby attributed to magnetothermal convection Q_{Mg} is calculated from Equation (A4):

$$Q_{Mg} = S\lambda_o(\Delta T + \delta T_1)\left(\frac{-\Delta\lambda}{\lambda_o}\right)_{\mathrm{SBE}} - Q_T\frac{\delta T_1}{\Delta T} \qquad (3)$$

According to this result, the magnetothermal contribution to the apparent thermal conductivity change is equal to the difference between the total measured field effect and the corresponding value of the SBE effect $(\Delta\lambda/\lambda_o)_{\mathrm{SBE}}$. However, the precise values for the ordinary SBE behavior

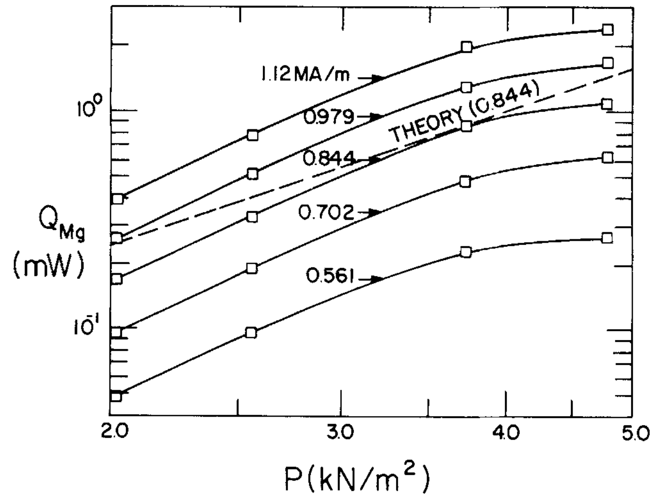

Fig. 3. Variation of the magnetoconvective heat flux in oxygen with pressure at $\Delta T = 22.6 \pm 0.1°C$, $T_0 = 86.1 \pm 0.2°K$.

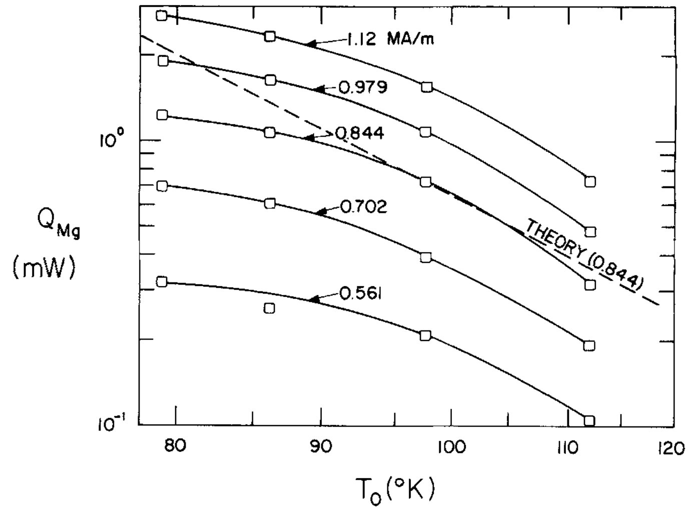

Fig. 4. Variation of the magnetoconvective heat flux in oxygen with average gas temperature at $P = 4.75kN/m^2$, $\Delta T = 22.6 \pm 0.1°C$.

for oxygen at low temperatures have not yet been determined. Therefore, the expected low temperature SBE curve has to be estimated.

To estimate the expected low pressure SBE behavior at low temperature, data were taken with small ΔT for oxygen pressures ranging from 0.27 to 0.93 kN/m^2. The results were then extrapolated to zero pressure at each temperature. Curve B in Figure 1 closely approximates such a low pressure envelope at 80°K. This estimation procedure was repeated for various average gas temperatures covering the range over which the magnetoconvection effect was measured. For most of the experimental conditions, the magnetothermal convection effects are one or two orders of magnitude larger than the SBE values; hence, an error of as much as 10% in estimating the expected SBE curve results in less than 1% error in the calculated values of Q_{Mg}. Curve A represents data for oxygen at 300°K (Hermans et al., 1970). This temperature dependence is in general agreement with that obtained for other gases (Hermans et al., 1970; Vevai et al., 1973).

The experimental results for the SBE corrected data are shown in Figures 3 to 6. The pressure dependence for various values of magnetic field H is shown in Figure 3. For these data, the overall cell temperature difference was fixed at $22.6 \pm 0.1°C$, and the average gas temperature was maintained constant at $86.1 \pm 0.2°K$ to give a reasonably large effect. The data increase with increasing pressure and show a quadratic pressure dependence predicted in Equation (1) only near 3.7 kN/m. For pressures higher than 3.7 kN/m^2, the effect levels off rapidly becoming less strongly dependent on the pressure. Below 2.7 kN/m^2, the data appear to approach a cubic pressure dependence asymptotically.

The effect of changing average gas temperature T_o is shown in Figure 4 for various magnetic field strengths. The values of p and ΔT for these data were 4.75 kN/m^2

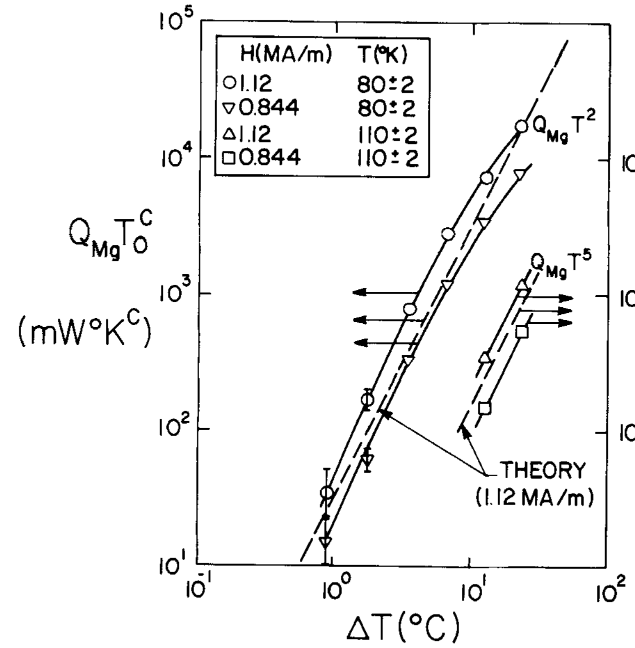

Fig. 5. Variation of the magnetoconvective heat flux in oxygen with the temperature gradient across the cell at $P = 4.75kN/m^2$.

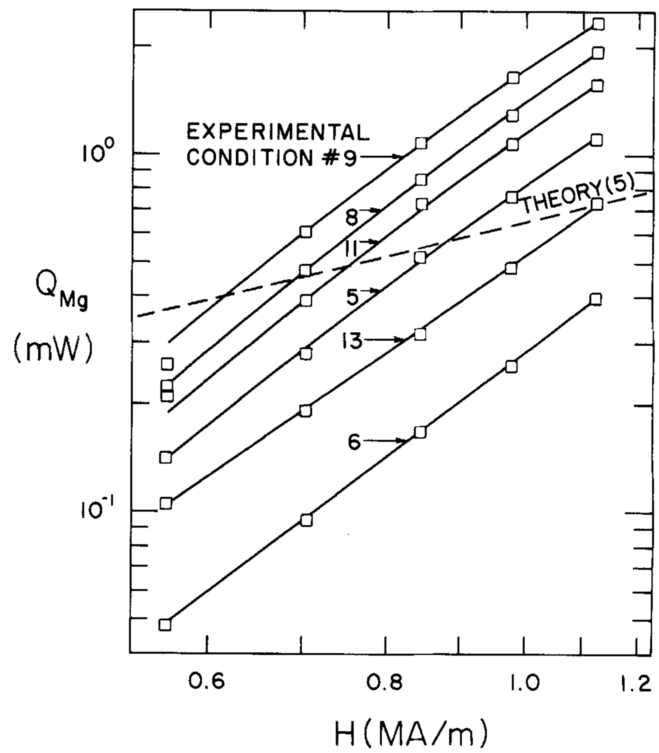

Fig. 6. Variation of the magnetoconvective heat flux in oxygen with magnetic field at various conditions (see Table 1).

and 22.6 ± 0.1°C, respectively. The effect increases uniformly with decreasing temperature. The estimated curves through the data points have a slope of about −5.5 at higher values of T_o and a slope near −2 at temperatures below 90°K. Hence, at least for temperatures higher than 90°K, the temperature dependence is given by $T_o^{-5.0 \pm 0.5}$. The predicted temperature dependence from Equation (1) is $T_o^{-5.07}$ if the viscosity temperature dependence for oxygen gas is included.

As mentioned above, it is not possible to hold the average gas temperature T_o at a constant value in the apparatus while varying ΔT. Hence, data were taken for which T_o was allowed to change as ΔT was changed. The functional dependence of Q_{Mg} on ΔT was then extracted by plotting $Q_{Mg} \times T_o^c$ as a function of ΔT as shown for various magnetic field strengths in Figure 5. The gas pressure was 4.75 kN/m². The value of the exponent c of Equation (2) for each curve is the one appropriate to the temperature at which the data were taken; approximate values for c of 2 and 5 were determined from Figure 4 for average gas temperatures of 80° and 110°K, respectively. Although the data are not exactly linear, a straight line with an average slope of two as predicted from Equation (1) fits these data points reasonably well, especially those obtained at 110°K.

The magnetic field dependence of the data is shown in Figure 6 for a variety of experimental conditions from the list of Table 1. The effect increases with increasing field and is approximately cubic in magnetic field dependence. However, the extrema of the slopes range from 4 to 2.5, decreasing with increasing magnetic field for each set of experimental conditions.

DISCUSSION

Two theoretical treatments of the magnetothermal convection effect, designated (I) (Park and Honeywell, 1973) and (II) (Clark and Honeywell, 1977), respectively, have been developed for the case of paramagnetic or diamagnetic gases confined between two semiinfinite parallel plates. In both analyses, a static magnetic field is applied horizontally along the direction of the temperature gradient in the gas; in (II), vertical components are added. The two treatments attribute the field induced convection to an enhancement of the natural convection currents caused by the temperature dependence of the gas magnetic susceptibility. For the case of a perfectly homogeneous magnetic field (I), the enhancement enters through magnetic pressure effects on the gas density. In (II), the effect is caused by the addition of a magnetic body force on the fluid, a force which can be appreciable even for quite small field gradients.

Both theories predict the same pressure, temperature, and temperature gradient dependencies. In (I), however, the field dependence is quadratic in the field. In (II), the effect is dependent upon the field-field gradient product as given in Equation (1). Detailed calculations have shown that the magnitude of the effect from (I) is two orders of magnitude smaller than those of (II). It is unlikely that the discrepancy between experiment and the theory of (I) can be accounted for by differences in geometry between the theoretical treatment and the experiment conditions. The theoretical predictions from (II) are illustrated in Figures 3 to 6 for the experimental conditions indicated. A constant field gradient of −160 A/m cm has been assumed in each case.

We note finally that magnetoconvection effects are not confined just to oxygen gas, nor theoretically just to paramagnetic gases. Magnetoconvection effects have been observed for nitric oxide at 121°K, 4.7 kN/m² with a temperature difference of 21.9°C (Vevai et al., 1973).

ACKNOWLEDGMENT

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NOTATION

H = externally applied magnetic field, A/m
H_x = externally applied constant magnetic field in the x direction, A/m
K_c = overall natural convection heat transfer coefficient, W/°C
K_L = overall heat transfer coefficient for heat energy losses Q_L, W/°C
P = hydrostatic pressure, N/m²
Q = heat transferred by conduction, W
Q_c = heat transferred by natural thermal convection, W
Q_L = heat energy dissipation other than gas conduction and convection, W
Q_T = total heat transfer rate, W
Q_{Mg} = magnetothermal contribution to the total heat transferred due to the magnetic field gradient effect, W
S = shape factor for heat transfer, m
T = temperature, °K
T_o = average T over ΔT, °K
T_1 = inner cylinder wall temperature, °K
T_2 = outer cylinder wall temperature, °K
δT_1 = increase in inner cylinder wall temperature with application of magnetic field, °C
ΔT = temperature difference across the fluid, $T_1 - T_2$, °C
Z = vertical coordinate direction, m

Greek Letters

η = dynamic viscosity, Ns/m²
η_o = average η over ΔT, Ns/m²
λ_H = thermal conductivity in applied magnetic field, H, W/m°C
λ_o = thermal conductivity in zero magnetic field, W/m°C
$\Delta\lambda$ = thermal conductivity difference, $\lambda_H - \lambda_o$, W/m°C

APPENDIX: THE RELATIONSHIP OF THE MAGNETOCONVECTIVE HEAT FLUX TO EXPERIMENTALLY MEASURED QUANTITIES

The total electrical energy Q_T supplied to the inner cylinder is transformed into heat energy and dissipated through several modes of heat transfer. In a magnetic field free environment at low temperatures, most of the heat energy is transferred from the inner cylinder to the outer cylinder either by conduction through the gas or by convection within the gas. Let the energy transfer by conduction be denoted by Q and that transferred by thermally induced convection currents be denoted by Q_c. For purposes of this discussion, the remainder of the heat energy transport may be lumped together as losses and denoted by the symbol Q_L. It may be thought of as including the energy transported from the inner cylinder by conduction through the end seals, the thermometer and heater leads, etc., to the outer cylinder or to the surroundings. With this simplification we may write

$$Q_T = Q + Q_c + Q_L \tag{A1}$$

The conduction heat transfer may be expressed as $S\lambda_o\Delta T$, where S is the appropriate shape factor for the experimental cell geometry. When radiation heat transfer is negligibly small, the energy losses Q_L may similarly be expressed to good approximation as $K_L \Delta T$. The convection heat transfer Q_c would normally be expressed as $K_c'(\Delta T)^2$, where K_c' is a natural convection heat transfer coefficient for the entire cell. However, for purposes of simplification in the treatment which follows, one power of ΔT will be incorporated into the coefficient. The effect on the resulting analysis is unimportant and results in negligible error for normal measuring conditions. The convection heat transfer will therefore be written as $K_c \Delta T$. With these simplifications, the total thermal energy transfer is given as

$$Q_T = S\lambda_o \Delta T + K_c \Delta T + K_L \Delta T \qquad \text{(A2)}$$

When a magnetic field is applied under conditions of constant outer wall temperature and total heat input to the cell, the field induced change in heat energy transfer causes a compensating change in the inner wall temperature. We may write

$$Q_T = S(\lambda_o + \Delta\lambda)(\Delta T + \delta T_1) + K_c(\Delta T + \delta T_1) + Q_{Mg} + K_L(\Delta T + \delta T_1) \qquad \text{(A3)}$$

Here, all magnetic field influences on convection are collected in the term Q_{Mg}, the quantity representing the amount of heat energy transferred by magnetothermal convection. It is assumed that the coefficient K_L is unaffected by the field.

Solving for Q_{Mg} from Equations (A2) and (A3), we get

$$Q_{Mg} = S\lambda_o(\Delta T + \delta T_1)\left(\frac{-\Delta\lambda}{\lambda_o}\right)_{\rm SBE} - Q_T\frac{\delta T_1}{\Delta T} \qquad \text{(A4)}$$

where $(\Delta\lambda/\lambda)_{\rm SBE}$ is the value of the SBE thermal conductivity effect estimated at the experimental condition corresponding to the datum point δT_1.

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Immobilization of Glucose Isomerase to Cellulose

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Glucose isomerase was covalently bonded to Solka-floc and DEAE cellulose by several techniques. The best results were obtained with techniques that opened the crystalline structure of the cellulose and kept it that way during immobilization. The most active catalyst was prepared by mercerizing Solka-floc, cross-linking it in that expanded configuration with epoxides, and immobilizing the enzyme with cyanogen bromide. The second most active catalyst was made using DEAE cellulose and a mixture of di and triepoxides as immobilization reagent.

The practical consequences of the results were tested by designing reactors to produce high fructose syrup using the activity and stability of several of the preparations. Since the activity decays with time, equations were developed to give the optimum time of running before the catalyst was discarded. The most promising one was the second most active; the most active catalyst yielded a production cost three times as great because of the large quantities of cyanogen bromide needed.

SCOPE

The enzymic isomerization of glucose to fructose has assumed major proportions in the starch industry in the production of high fructose corn syrup (Schnyder, 1973). Although some processors have been content to employ free enzyme in batch processes, the real interest has been in continuous processes using immobilized enzymes. The first process (Lamm and Dworshack, 1972) involved immobilization in heat-fixed microbial cells, followed by a process involving separation of the intracellular enzyme and immobilization on DEAE cellulose, announced by Schnyder

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