t	= temperature	[°C]
U	= dimensionless velocity in X -direction	[—]
u	= velocity in x -direction	[cm/sec]
V	= dimensionless velocity in Y -direction	[—]
v	= velocity in y -direction	[cm/sec]
W	= dimensionless velocity in Z -direction	[—]
w	= velocity in z -direction	[cm/sec]
X, Y, Z	= dimensionless rectangular co-ordinates	[—]
x_A	= half width of reaction zone	[cm]
x, y, z	= rectangular co-ordinates	[cm]
α	= thermal diffusivity	[cm ² /sec]
β	= volumetric coefficient of expansion	[1/°C]
ζ	= dimensionless vorticity	[—]
λ	= thermal conductivity	[cal/cm·sec·°C]
μ	= viscosity	[g/cm·sec]
ν	= kinematic viscosity	[cm ² /sec]
ρ	= density	[g/cm ³]
ϕ	= dimensionless stream function	[—]
<Subscripts>		
H	= reaction zone	
W	= wall	

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CHARACTERISTICS OF THE HORIZONTAL EPITAXIAL REACTOR — IN CASE OF MASS TRANSFER RATE CONTROL —

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Concentration distributions of reactant and mass transfer coefficient in a horizontal epitaxial reactor were numerically calculated from fundamental equations under some assumptions in the case of mass transfer rate control. The concentration distribution in the cross-section of the channel became uniform when Gr and the distance from the leading edge were increased or the feed rate of gas was decreased. To check the calculated results, carbon plates were burned in this reactor under the mass transfer rate control and local concentration of carbon dioxide was measured. The calculated concentration and mass transfer coefficient were a sufficiently good approximation to the experimental ones.

Introduction

In an epitaxial reactor (chemical vapor deposition reactor), deposition reaction occurs on the surface of a susceptor heated up in 700–1200°C. Thickness and purity of reaction products must be controlled for the application purpose. Especially, in the case of production of semiconductor material, the thickness must be

as strictly uniform as possible. Thickness and purity of reaction products are influenced by the concentration distribution of reactants in the reactor.

In a horizontal epitaxial reactor, which has been generally used, vigorous three-dimensional free convective flow is generated by the buoyancy force induced by large temperature differences. Therefore, velocity distributions are not easily calculated from fundamental equations, and concentration distributions of reactant gas and temperature distributions have been determined experimentally^{1,3–6,8,13,14}. There are few

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theoretical studies for transport phenomena in this reactor^{12,15}).

In order to analyze the transport phenomena in this reactor theoretically, the cross-sectional and axial velocity profiles and the temperature distributions in a horizontal epitaxial reactor were obtained by calculating numerically from fundamental equations, and these results agreed well with the experimental ones¹⁰).

In this paper, the concentration distributions of reactant gas and the mass transfer coefficient in the case of mass transfer rate control were calculated numerically from fundamental equations under some suitable assumptions.

Furthermore, to check these calculated results, carbon plates in the horizontal epitaxial reactor were burned under the mass transfer rate control, and the local concentration of carbon dioxide in the reactor was measured.

1. Fundamental Equations and Numerical Calculation Procedure

Let us consider mass transfer in three-dimensional flow with laminar natural convection in a horizontal rectangular channel, of which a part of the bottom is heated and the other walls are kept at low constant temperature, as in the previous paper¹⁰). From the observed flow pattern in this reactor, the following assumptions are made to calculate the concentration of reactant.

1) Since the reactant gas is well preheated before entering the reaction zone, the temperature distribution and the flow pattern in the reactor are established and not affected by its axial length.

2) Overall reaction rate is controlled by the mass transfer rate from the bulk of gas to the solid surface.

3) Since Pe is quite large for this reactor, the axial diffusion term in a mass balance equation may be neglected.

4) The physical properties of the fluid, such as viscosity, thermal conductivity, etc., are independent of temperature, but the buoyancy force is dependent upon the temperature difference.

Fundamental equations are the equations of continuity, momentum, energy¹⁰) and the mass balance. The dimensionless variables, the dimensionless stream function ψ and the vorticity ζ are defined as follows.

$$\left. \begin{aligned} (U, V, W) &= (u, v, w)H/\nu \\ (X, Y, Z) &= (x, y, z)/H \\ T &= (t - t_w)/(t_H - t_w), \quad P = pH^2/\rho\nu^2, \quad C = c/c_0 \\ Gr &= g\beta H^3(t_H - t_w)/\nu^2, \quad Pr = C_p\mu/\lambda, \quad Sc = \nu/D \end{aligned} \right\} \quad (1)$$

$$U = -\partial\psi/\partial Y, \quad V = \partial\psi/\partial X \quad (2)$$

$$\zeta = \partial V/\partial X - \partial U/\partial Y = (\partial^2\psi/\partial X^2 + \partial^2\psi/\partial Y^2) \quad (3)$$

After some manipulations the nondimensional fundamental equations are written as Eqs. (4)–(6)

and (8), and boundary conditions become Eqs. (7) and (9).

$$U \frac{\partial \zeta}{\partial X} + V \frac{\partial \zeta}{\partial Y} = \left(\frac{\partial^2 \zeta}{\partial X^2} + \frac{\partial^2 \zeta}{\partial Y^2} \right) + Gr \frac{\partial T}{\partial X} \quad (4)$$

$$U \frac{\partial T}{\partial X} + V \frac{\partial T}{\partial Y} = \frac{1}{Pr} \left(\frac{\partial^2 T}{\partial X^2} + \frac{\partial^2 T}{\partial Y^2} \right) \quad (5)$$

$$U \frac{\partial W}{\partial X} + V \frac{\partial W}{\partial Y} + \frac{\partial P}{\partial Z} = \left(\frac{\partial^2 W}{\partial X^2} + \frac{\partial^2 W}{\partial Y^2} \right) \quad (6)$$

$$\left. \begin{aligned} X=0; \quad \psi &= \partial^2\psi/\partial X^2 = \partial W/\partial X = \partial T/\partial X = 0 \\ X=L/H; \quad \psi &= \partial\psi/\partial X = W = T = 0 \\ Y=0, \quad \left\{ \begin{array}{l} 0 \leq X \leq x_A/H; \quad \psi = \partial\psi/\partial Y = W = 0, \quad T = 1 \\ x_A/H < X \leq L/H; \quad \psi = \partial\psi/\partial Y = W = T = 0 \end{array} \right. \\ Y=1; \quad \psi &= \partial\psi/\partial Y = W = T = 0 \end{aligned} \right\} \quad (7)$$

$$U \frac{\partial C}{\partial X} + V \frac{\partial C}{\partial Y} + W \frac{\partial C}{\partial Z} = \frac{1}{Sc} \left(\frac{\partial^2 C}{\partial X^2} + \frac{\partial^2 C}{\partial Y^2} \right) \quad (8)$$

$$\left. \begin{aligned} X=0; \quad \partial C/\partial X &= 0 \\ X=L/H; \quad \partial C/\partial X &= 0 \\ Y=0, \quad \left\{ \begin{array}{l} 0 \leq X \leq x_A/H; \quad C = 0 \\ x_A/H < X \leq L/H; \quad \partial C/\partial Y = 0 \end{array} \right. \\ Y=1; \quad \partial C/\partial Y &= 0 \\ Z=0; \quad C &= 1 \end{aligned} \right\} \quad (9)$$

Since these equations could not be solved analytically, in this study Eqs. (2)–(9) were solved numerically.

The calculation procedure is as follows: At first each equation was written in a finite difference form with “upwind method”^{2,9,11}). Then, Eqs. (2)–(5) and (7) were solved numerically with relaxation method and U and V were obtained from Eq. (2). After establishing the velocity components in X and Y directions, the axial velocity component, W , was calculated from Eqs. (6) and (7) with relaxation method as a function of X and Y . Finally, concentration distributions of reactant gas were calculated from Eqs. (8) and (9).

In a parabolic partial differential equation such as Eq. (8), axial step size ΔZ must be small enough to satisfy the stability conditions. In this calculation ΔX and ΔY were 1.03×10^{-1} and 9.09×10^{-2} , respectively. If ΔZ was smaller than 3.8×10^{-4} , the calculated results had good reproducibility.

The calculation has been carried out for $D_e = 3.24, 3.86, 5.0$ cm, $L/H = 0.827, 1.204$, $x_A/H = 0.25-0.87$, $Z = 1.25-8.70$, $Pr = 0.69$, $Gr = 10^3-10^5$, $Sc = 0.594$ and $Q = 1.2-4.9$ l/min ($Re = 12.8-52.7$).

2. Apparatus and Experimental Procedure

To check the calculated results, carbon plates were burned on a susceptor heated by a high-frequency induction coil and local concentrations of carbon dioxide were measured.

Fig. 1 Schematic diagram of experimental apparatus

w_z [cm]	length [cm]	thickness [cm]
1.0	10.0–20.0	0.20
2.0	"	"
3.0	"	"
4.0	"	"

Fig. 2 Concentration distributions with $Q=1.2, 2.5$ and 4.9 l/min ($Re=12.8, 26.8$ and 52.7) for $L/H=0.827$, $x_A/H=0.310$, $Gr=1.5 \times 10^4$, $Pr=0.69$, $Sc=0.594$ and $Z=3.85$ (---: $U=V=0$, $\circ$: exp. results)

Figure 1 shows a schematic diagram of the experimental apparatus. The rectangular channel with a cross sectional area of $2.6 \times 4.3 \text{ cm}^2$ ($L/H=0.827$)* and 170 cm long was made of transparent quartz glass. A Ni-plate susceptor ($2.1 \times 1.0 \times 100 \text{ cm}^3$) was put on the bottom of the channel and was heated by a high-frequency induction coil. A carbon plate was put on this susceptor. The temperatures of the carbon plate and the channel wall were measured by a micro-pyrometer and Pt-PtRh thermocouples ($0.3 \text{ mm} \phi$), respectively. The carbon plate used in this experiment was the graphitic carbon SEG-5 made in Nippon Carbon Co., Ltd. Its sizes are shown in Table 1. Air from a compressor was passed through a dehydration tower filled with silica-gel and a CO_2 -absorption tower filled with $\text{KOH}-\text{CaO}$, and then entered the reactor. To establish the temperature and velocity distribution of the reactant gas (air) towards the axial direction, the carbon plate was put on middle part of the Ni-plate.

Gas chromatography was used to analyse carbon dioxide in the gas. Charcoal was packed in a chromatograph column, and hydrogen was the carrier gas. To investigate the local concentration of carbon dioxide, a narrow sampling tube made of porcelain was put into the reactor and the gas sample was taken by this tube.

The carbon plate was burned in the range of 1000

* Rectangular channel, $2.3 \times 5.5 \text{ cm}^2$ ($L/H=1.204$), was also used for this experiment.

to 1100°C . In this temperature range, the overall combustion rate was controlled by the mass transfer rate of oxygen to the carbon plate⁸. The feed flow rates of air were in the range of 1.2 to 4.9 l/min (at 20°C , 1 atm). In these flow rates, steady spiral flow was established in the reactor⁷. No carbon monoxide was detected in these experimental conditions.

3. Comparison of Calculated with Experimental Results

Since the concentration distributions of a reactant gas were symmetrical to Y axis in a horizontal epitaxial reactor, these distributions were calculated in the right-hand half of the cross-section.

Figure 2 shows dimensionless concentration distributions of the reactant gas in the cross-section at the distance of 10 cm from the leading edge of carbon plate where the flow rates of feed air are 1.2, 2.5 and 4.9 l/min, respectively.

The solid lines show the concentration distributions calculated by the method mentioned in section 1 and the dotted lines show the concentration distributions in the case of $U=V=0$ (W was calculated from Eq. (6) under the condition of $U=V=0$). When the feed rate of air, Q , is increased, the concentration distributions of the reactant gas become less uniform towards cross-sectional direction. This means that the effect of vortex flow upon convective mass transfer is comparatively decreased by increasing Q .

Fig. 3 Concentration distributions with $Z=1.92, 5.77$ and 7.69 for $L/H=0.827$, $x_A/H=0.310$, $Gr=1.5 \times 10^4$, $Pr=0.69$, $Q=1.2$ l/min ($Re=12.8$) and $Sc=0.594$ (○: exp. results)

Fig. 4 Comparison of calculated concentrations with experimental ones

In Fig. 2 the numerical values written near the experimental point are the local concentrations of reactant gas (oxygen) obtained by combustion of carbon plates under the same conditions as those of this numerical calculation. As can be seen from these values, the calculated concentrations are in a sufficiently good approximation to the experimental ones. Furthermore, such a good agreement between the calculated results and the experimental ones was also obtained in other cases ($L/H=1.204$ ($H=2.30$ cm, $L=2.77$ cm), $x_A/H=0.43-0.870$). Figure 2 also shows that the concentration distributions of the reactant gas in the case of $U=V=0$ are quite different from those of spiral flow.

Figure 3 shows concentration distributions of the reactant gas at distances of 5, 15 and 20 cm from the leading edge of carbon plate, i.e., $Z=1.92, 5.77$ and 7.69 , respectively.

With increasing the distance Z , the reactant concentration decreases successively and the concentration distribution across the cross-section becomes more uniform. The numerical values at each point also show the experimental ones of local reactant

concentrations. The experimental results are a sufficiently good approximation to the computed ones. Furthermore, such a good agreement is obtained in other cases ($L/H=1.204$ ($H=2.30$ cm, $L=2.77$ cm), $x_A/H=0.43-0.870$).

The comparison of the calculated results with the experimental ones are shown in Fig. 4. In this figure all experimental data in the cases of $L/H=0.827$ and 1.204 are plotted. It appears that the experimental values agree approximately with the calculated ones although the plotted points are somewhat scattered.

Figure 5 shows the concentration distributions of the reactant gas in the cases where Gr are 10^3 , 5×10^4 and 10^5 , respectively. The effect of Gr on the concentration distributions is quite significant. When Gr is increased, the intensity of vortex flow in the channel becomes large, and then the concentration distributions of the reactant gas become more uniform.

Figure 6 shows the concentration distributions of the reactant gas with the distance from the leading edge of carbon plate as a parameter in the cases where values of x_A/H are 0.517 and 0.724, respectively. Comparing Fig. 6 with Fig. 2, if the width ratio of the reaction zone is increased, the reactant concentration decreases. In the case of $x_A/H=0.724$, the reaction zone almost occupies the bottom of the channel and the concentration distributions of the reactant gas are quite different from those under the other conditions.

4. Discussion

4.1 Mass transfer coefficient

The mass transfer coefficient, k , may be obtained from concentration gradients near the reaction surface, as follows:

$$k = - \frac{\mathcal{D}}{C_{AV} x_A} \int_0^{x_A/H} \left(\frac{\partial C}{\partial Y} \right)_0 dX$$

or from the mass balance of reactant gas between subsequent two planes perpendicular to the horizontal axis with a distance of ΔZ . Since the concentration

Fig. 5 Concentration distributions with $Gr=10^3$, 5×10^4 and 10^5 for $L/H=0.827$, $x_A/H=0.310$, $Pr=0.69$, $Q=2.5$ l/min ($Re=26.8$) and $Sc=0.594$

gradient is very steep near the surface, very small mesh size is required to evaluate its gradient. Therefore, in this study, the mass transfer coefficient was obtained by the mass balance of reactant gas between two planes. But in some operating conditions, the coefficient was obtained by both two methods mentioned above and these values agreed well with each other.

From the calculated results, the calculated mass transfer coefficient, k , increased as Gr was increased, and k was directly proportional to $Gr^{0.15}$. If Gr was changed from 10^3 to 10^6 under $Re=36$, k changed from 3.1 to 6.1 cm/sec. The value of k is quite high in comparison with that in the case of no vortex flow. The large value of k is due to the large concentration gradient induced by the vortex flow near the reaction zone.

The empirical equation for the overall mass transfer coefficient k_{ov} in a horizontal epitaxial reactor was already obtained⁸⁾. Figure 7 shows comparison of the experimental mass transfer coefficient with the calculated one. From Fig. 7, the calculated result for k_{ov} is a sufficiently good approximation to the measured one. Furthermore, both the calculated mass transfer coefficient for k_{ov} and the experimental one have the same tendency for such factors as Gr , Q , W and Z .

Conclusion

The concentration distribution of reactant gas in a horizontal epitaxial reactor was calculated numerically from the fundamental equations with certain assumptions, and following results were obtained.

(1) In this calculation, the concentration distributions in the cross-section of the channel become uniform when Gr and the distance from the leading edge are increased or the feed rate of gas is decreased.

(2) The calculated results agreed well with the experimental ones.

(3) The theoretical value of the mass transfer co-

Fig. 6 Concentration distributions with $x_A/H=0.517$ and 0.724 for $L/H=0.827$, $Gr=1.5 \times 10^4$, $Pr=0.69$, $Q=2.5$ l/min ($Re=26.8$) and $Sc=0.594$

Fig. 7 Comparison of calculated k_{ov} with experimental one

efficient agrees approximately with the experimental one.

Nomenclature

C	= dimensionless concentration of reactant gas	[—]
C_{AV}	= average concentration of reaction gas in cross-section	[—]
C_p	= heat capacity at constant pressure	[cal/g·°C]
c	= concentration of reactant gas	[mol/cm ³]
D_e	= equivalent diameter of tube ($=4LH/(2L+H)$)	[cm]
$\mathcal{D}$	= molecular diffusivity	[cm ² /sec]
G	= mass velocity of gas	[g/cm ² ·sec]
Gr	= Grashof number ($=g\beta H^3(t_H - t_W)/\nu^2$)	[—]
g	= gravitational acceleration	[cm/sec ²]
H	= channel height	[cm]
k	= mass transfer coefficient	[cm/sec]
L	= half width of channel	[cm]
l	= axial length of carbon plate	[cm]
P	= dimensionless pressure	[—]
Pe	= Peclet number ($=Gl/\mathcal{D}\rho$)	[—]
Pr	= Prandtl number ($=C_p\mu/\lambda$)	[—]
p	= fluid pressure	[g/cm·sec ²]
Q	= volume flow rate of gas	[l/min]
Re	= Reynolds number ($=Q\rho/2L\mu$)	[—]
Sc	= Schmidt number ($=\mu/\rho\mathcal{D}$)	[—]
T	= dimensionless temperature	[—]
t	= temperature	[°C]
U	= dimensionless velocity in X -direction	[—]
u	= velocity in x -direction	[cm/sec]
V	= dimensionless velocity in Y -direction	[—]
v	= velocity in y -direction	[cm/sec]
W	= dimensionless velocity in Z -direction	[—]
w	= velocity in z -direction	[cm/sec]
w_i	= width of carbon plate	[cm]
X, Y, Z	= dimensionless rectangular co-ordinates	[—]
x_A	= half width of reaction zone	[cm]
x, y, z	= rectangular co-ordinates	[cm]
α	= thermal diffusivity	[cm ² /sec]
β	= volumetric coefficient of expansion	[1/°C]

ζ	= dimensionless vorticity	[—]
λ	= thermal conductivity	[cal/cm·sec·°C]
μ	= viscosity	[g/cm·sec]
ν	= kinematic viscosity	[cm ² /sec]
ρ	= density	[g/cm ³]
ϕ	= dimensionless stream function	[—]

<Subscripts>

H	= reaction zone
ov	= overall
W	= wall
0	= inlet to the channel

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