

A NEW CVD REACTOR FOR SEMICONDUCTOR FILM DEPOSITION

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Abstract—The concept and design of a new chemical vapor deposition (CVD) reactor is presented for both epitaxial and nonepitaxial film deposition in semiconductor processing. The reactor is designed in such a way that a stagnant semiconductor source fluid of uniform concentration is provided for the film deposition without causing free or forced convection. The supply of the source gas for the deposition is by diffusion through a porous material such as quartz or graphite. Compared to the low pressure CVD (LPCVD) reactor with mounted wafer configuration, the new reactor should give a better film thickness uniformity and about an order of magnitude reduction in the amount of the source gas required. Further, at least for polycrystalline silicon deposition, the deposition rate can be much higher than is currently practiced with the LPCVD reactor. Design equations for the reactor are given. Details on the design for the polycrystalline silicon deposition are also given.

INTRODUCTION

Polycrystalline and amorphous film deposition on a substrate is an important part of any semiconductor fabrication. As a result, considerable attention has been paid to the deposition apparatus or reactor. The chemical vapor deposition (CVD) reactors are usually classified [1] as horizontal reactor, vertical reactor, barrel-type reactor, which is an outgrowth of a horizontal reactor with tilted susceptor, and "pancake"-type reactor, which is similar to Bertly reactor. Although the classification is based on the placement of substrates, the major factors affecting the mode of operation are temperature and pressure.

The CVD reactors are increasingly operated at low pressures, typically around one torr, because of better thickness uniformity of grown film afforded by the low pressure. Although the better uniformity over the atmospheric CVD is often attributed to higher diffusivity at lower pressure, the major factor for the better uniformity has to do with the stabilization of flow or absence of buoyancy effects at low pressures [2]. The question of which regime (kinetic or mass-transfer) controls at low pressure is not dependent solely on the diffusivity but rather on the relative magnitude of diffusion and intrinsic growth rate. Note in this re-

gard that the growth rate decreases with decreasing pressure.

For polycrystalline silicon film growth, the usual choice is a hot wall, low pressure reactor with wafer mounted on a rack. The feed gas flows in the direction perpendicular to the mounted wafers [3]. Typical flow rates are around 50 sccm for pure silane feed or 500 sccm with nitrogen carrier gas with approximately one fourth silane by volume for the polysilicon film growth. The corresponding growth rate is approximately 20 nm/min. The flow in the annulus between the tube wall and the mounted wafers serves as the source of the gas; the film growth on the mounted wafers is mainly by the supply of the gas through the diffusion of the gas between two adjacent wafers. The wafer-to-wafer uniformity is usually controlled by choosing a sufficiently high flow rate so that the reactants are not depleted toward the reactor outlet, often in conjunction with temperature control in the direction of flow.

Although the low pressure CVD (LPCVD) reactor is a significant improvement over the atmospheric pressure reactor, it is not without problems. The streamlines intruding into the space between two adjacent wafers [e.g.9] can cause local nonuniformity. The wafer-to-wafer nonuniformity is still a problem. More importantly, the wafer-to-wafer nonuniformity problem limits the number of wafers that can be processed. Another problem is the waste of quite costly, high

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purity gases: the amount of the gases used is too far in excess of what is needed for the actual film growth. Unlike the usual chemical reactors and processes, the gases in semiconductor processing are not recycled because of the impurity problem the recycle can cause. Note in this regard that the purity of interest is in the ppm range.

Proposed in this paper is the concept and the design of a novel CVD reactor that can assure the thickness uniformity for any number of wafers processed and yet the amount of source gas used can be reduced at least by an order of magnitude.

CVD REACTOR

Consider polycrystalline film deposition with the mounted wafer configuration. Suppose that the stagnant fluid medium of constant concentration and temperature can be provided for the film deposition in the absence of buoyancy-driven free convection. This latter condition can be assured by operating the reactor at a low pressure. Suppose further that the supply of gases for the deposition is by diffusion rather than forced convection or flowing fluid. Then the amount of gas used could be made just enough (by diffusion) to assure uniform deposition across a wafer rather than across a number of wafers, thereby reducing substantially the amount of gas used.

The fact that diffusive flux is involved means that the supply of gas should be by diffusion through a porous material. The fact that a low pressure is to be used suggests that a part of reactor should be under vacuum and that there should be a buffer zone between the low pressure region and the vacuum to preserve the stagnancy of fluid in the low pressure region. A reactor satisfying these requirements is shown in Fig. 1.

The reactor consists of five parts in a rectangular cell. The empty upper chamber is maintained at a desired total pressure by regulating the feed rate of the source gases. If a total pressure less than atmospheric pressure is desired, the upper chamber is first evacuated to a pressure lower than the desired pressure by a mechanical pump and then the pressure maintained at the desired level by the feed rate. The second part is a column (block) of porous material, preferably quartz or graphite with a desired average pore diameter. A wide range of average pore size material can be chosen for specific design. The third part is the deposition chamber wherein wafers mounted on a rack are placed in the middle. Because of the vacuum in the empty lower chamber, which is the fifth part,

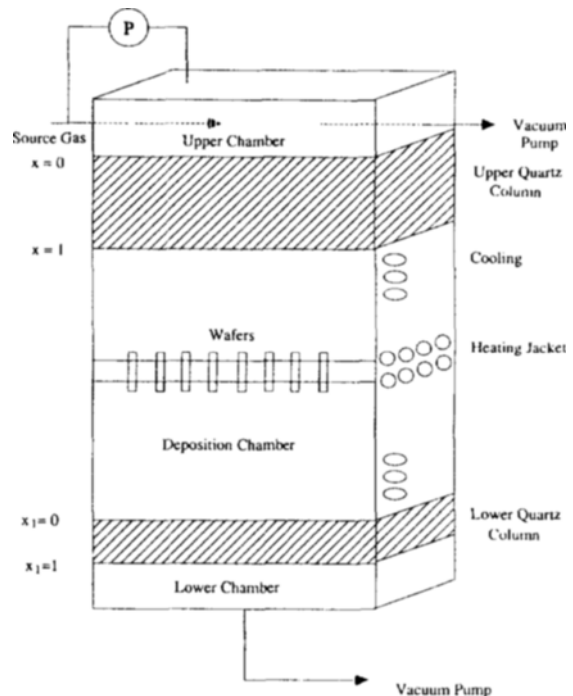


Fig. 1. New CVD reactor.

the deposition chamber is at a lower pressure. As discussed later, the total pressure in the deposition chamber can be maintained at a desired level at steady state.

The gas around the mounted wafers is heated by a furnace jacket and the part of the chamber close to the quartz columns are cooled to prevent homogeneous nucleation outside the heated region. It is noted that the deposition chamber is symmetric around the mounted wafer, in particular with respect to cooling. The fourth part, which is another column of porous quartz, ensures gas stagnancy in the deposition chamber. The relative length of the quartz column with respect to that of the upper quartz column is an important design parameter for the pressure in the deposition chamber.

In the basic unit, the wafers are placed on a hung rack in the direction of "diffusion" so that if there were any thickness nonuniformity it would be nonuniformity within a wafer. This means that there is essentially no limitation on the number of wafers that can be processed; the only limitation being the size (side width) of the reactor. Since the supply of source gas to the wafer is through diffusion rather than the usual flow of gas (approximately 500 sccm for a typical LP-CVD reactor with approximately 23% source gas such

as silane by volume [3]), a significant reduction of the costly source gas can be realized.

DESIGN CONSIDERATIONS

There are essentially two requirements for uniform deposition in the reactor : uniform concentration and absence of free or forced convection in the deposition chamber when the temperature is uniform. Consider the steady-state mass balance in the deposition chamber for the first requirement:

$$A_e [N_m - N_{out}] = G_i + D_i = E \quad (1)$$

where A_e is the external area of the quartz column facing the deposition chamber, N_m is the flux of the source gas from the upper quartz column, N_{out} is the flux into the lower quartz column, G_i is the total growth rate in moles/time ($G_i = \rho_s G n A_w / M$; G : growth rate in length/time, n : number of wafers, ρ : density of film, A_w : wafer area, M : molecular weight of the film), and D_i is the deposition rate onto the hot walls. If the flux from the upper quartz column is much higher than the deposition rate E , the concentration in the deposition chamber should be sufficiently uniform. Dividing both sides of Eq. (1) by $A_e N_m$ yields

$$1 - \frac{N_{out}}{N_m} = \frac{E}{A_e N_m} \quad (2)$$

Therefore, the concentration uniformity is assured within a desired limit if

$$\frac{E}{A_e N_m} < \delta \quad (3)$$

where δ is a small number, say 0.001 (less than 0.1% change in the concentration).

The second requirement of no free convection can be met [4] if

$$Ra = \frac{b^3(-\beta_h)(T_h - T_c)g}{\nu\alpha} < 1708 \quad (4)$$

In view of the fact that the number is for the case of two parallel plates, a more conservative (smaller) number than 1708 may have to be used. This criterion is for a stagnant fluid confined by two parallel plates, one of which is at a cold temperature T_c and the other at a higher temperature T_h (lower plate) separated by a distance b . Here Ra is the Rayleigh number, β_h [$-\rho\{\partial(1/\rho)/\partial T\}_p$] is the coefficient of volume expansion at T_h , ρ is the fluid density, ν is the kinematic viscosity, α is the thermal diffusivity, and g is the gravity.

In light of the two requirements, consider the flux in the upper quartz column. In general, the flux of species i in one direction [5] consists of viscous flow, gaseous diffusion, and surface diffusion. If the surface diffusion is neglected and an effective diffusivity is used, the flux of species i , N_i , in the absence of reaction can be written in terms of pressure as follows:

$$N_i = -p_i \frac{B_o}{R_g T \mu} \frac{dP}{dz} - \frac{D_i}{R_g T} \frac{dp_i}{dz} \quad (5)$$

where p_i is the partial pressure of species i , B_o is the Darcy permeability, R_g is the gas constant, T is the temperature, μ is the fluid viscosity, P is the total pressure, D_i is the effective diffusivity, and z is the axial coordinate in the direction of diffusion. The ideal gas law was used in writing Eq. (5). The mass conservation equations for the upper quartz column can be written in the following dimensionless form:

$$\frac{d^2 y_i}{dx^2} + \frac{\beta}{\gamma_i} \left(y_i \frac{d^2 Y}{dx^2} + \frac{dy_i}{dx} \frac{dY}{dx} \right) = 0 \quad (6)$$

$$\sum_i y_i = Y \quad (7)$$

where

$$x = z/L_u, \quad y_i = p_i/P_o, \quad Y = P/P_o, \quad \beta = \frac{B_o P_o}{\mu D_i}, \quad \gamma_i = \frac{D_i}{D_c} = \left(\frac{M_i}{M_c} \right)^{1/2} \quad (8)$$

Here P_o is the total pressure in the empty upper chamber, L_u is the length of the upper quartz column, D_i is the effective diffusivity of carrier gas, M_c is the molecular weight of carrier gas, and M_i is the molecular weight of species i . Eq. (7) follows from the definitions of y_i and Y . The boundary conditions are:

$$Y = 1, \quad y_i = (y_i)_f \quad \text{at } x = 0 \quad (9)$$

where $(y_i)_f$ is the mole fraction of species i in the feed. One can also write the same equations as Eqs. (6) and (7) for the lower quartz column and the boundary conditions are:

$$Y = y_i \cong 0 \quad \text{at } x_i = 1 \quad (10)$$

where x_i is the axial coordinate normalized with respect to the lower quartz column length L_c with the origin at the top of the column. These conditions follow from the fact that the empty lower chamber is under vacuum. A specific value of Y , which could be very small, may be used. In such a case, the condition on y_i should be replaced by the flux ratios at the end of the column. However the value of Y and y_i are

so small that they can be set zero without causing any deviation. The other boundary conditions for solving the two conservation equations are the continuity conditions at the deposition chamber:

$$\begin{aligned} N_i|_{x=1} &= N_i|_{x_l=0} \\ Y|_{x=1} &= Y_i|_{x_l=0} \end{aligned} \quad (11)$$

The flux in terms of dimensionless y_i is given by

$$N_i = -\alpha_i \left(\frac{dy_i}{dx} + \frac{\beta}{\gamma_i} y_i \frac{dY}{dx} \right) \quad (12)$$

where

$$\alpha_i = \frac{D_i P_o}{R_g T L} ; L = L_u \text{ or } L_l \quad (13)$$

The solution of these equations yields the necessary information required for specifying the design operating conditions.

Consider the design of the reactor for polycrystalline silicon growth. The polycrystalline growth in the LPCVD reactor is usually carried out with either pure SiH_4 or 20 to 30% SiH_4 in nitrogen carrier gas at around 0.2-1 torr. The growth rates are 10 to 20 nm/min [6]. Consider the case of pure SiH_4 . In these case, only one conservation equation [Eq. (6)] is involved, which is

$$\frac{d^2 Y}{dx^2} + \beta \left[Y \frac{d^2 Y}{dx^2} + \left(\frac{dY}{dx} \right)^2 \right] = 0; \quad \text{upper quartz column} \quad (14)$$

$$\frac{d^2 Y}{dx_l^2} + \beta_l \left[Y \frac{d^2 Y}{dx_l^2} + \left(\frac{dY}{dx} \right)^2 \right] = 0; \quad \text{lower quartz column} \quad (15)$$

where

$$\begin{aligned} x_i &= z_i/L_i, \quad \beta_i = \frac{(B_o)_i P_o}{\mu(D)_i}; \quad x_i = 0 \\ &\text{at the top of the lower quartz column} \end{aligned} \quad (16)$$

$$\begin{aligned} Y &= 1 \quad \text{at } x=0 \\ Y &= \omega \quad \text{at } x_l=1 \\ \text{with } Y|_{x=1} &= Y|_{x_l=0} = Y_d \\ N|_{x=1} &= N|_{x_l=0} \end{aligned} \quad (17)$$

The last two conditions in Eq. (17) are the (approximate) continuity conditions in the deposition chamber when the requirement of uniform concentration given by Eq. (3) is satisfied. Here ω is Y in lower chamber, which should be close to zero under vacuum condition, and L_l is the length of the lower quartz column. The subscript l is for the lower quartz column. Note that β and β_l do not have to be the same since different

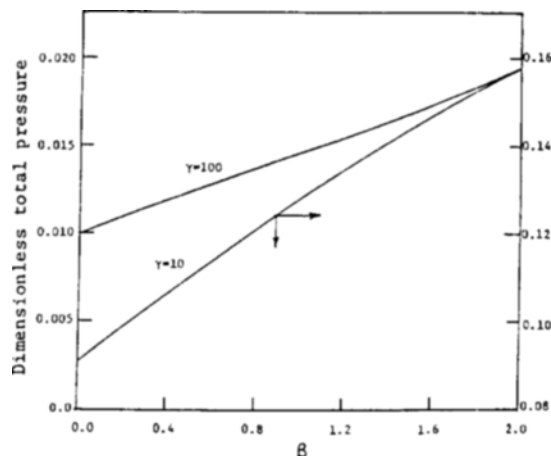


Fig. 2. Dimensionless total pressure in the deposition chamber.

quartz pore structure can be used for the two columns.

The solutions yield:

$$\begin{aligned} \frac{1}{2} Y^2 + \frac{1}{\beta} Y &= \frac{1}{\beta} + \frac{1}{2} - x \left\{ \frac{1}{\beta} (1 - Y_d) + \frac{1}{2} (1 - Y_d^2) \right\}; \\ &\text{upper quartz column} \end{aligned} \quad (18)$$

$$\begin{aligned} \frac{1}{2} Y^2 + \frac{1}{\beta_l} Y &= \frac{1}{\beta_l} Y_d + \frac{1}{2} Y_d^2 - x_l \left\{ \frac{1}{\beta_l} (Y_d - \omega) + \frac{1}{2} (Y_d^2 - \omega^2) \right\}; \\ &\text{lower quartz column} \end{aligned} \quad (19)$$

where

$$\begin{aligned} Y_d &= \left\{ \frac{1 + \bar{\alpha}}{\bar{\alpha} \beta_l + \beta} \right\} \\ &\left(\left[1 + \frac{(\bar{\alpha} \beta_l + \beta)(2 + \beta + \bar{\alpha}(2\omega + \beta_l \omega^2))}{(1 + \bar{\alpha})^2} \right]^{1/2} - 1 \right) \end{aligned} \quad (20)$$

$$\bar{\alpha} \equiv \frac{\alpha_l}{\alpha_u} = \frac{D_l}{L_l} \frac{L}{D}; \quad D_l: D \text{ in the lower chamber}$$

It is instructive to examine the dimensionless total pressure (normalized with respect to the total pressure in the empty upper chamber, P_o) when quartz columns are of the same pore structure, i.e., $\beta = \beta_l$. Then Eq. (20) reduces to

$$Y_d = \frac{1}{\beta} \left\{ \left[1 + \frac{\beta(2 + \beta)}{1 + \gamma} \right]^{1/2} - 1 \right\} \quad \omega \rightarrow 0; \quad \gamma = \frac{L}{L_l} \quad (21)$$

since then $\bar{\alpha}$ reduces to γ . The dimensionless total pressure in the deposition chamber is shown in Fig. 2 as a function of β with γ as the parameter. It is seen that the pressure is dependent mainly on the value of γ . For a given γ , the pressure increases with increasing β . The dimensionless pressure given

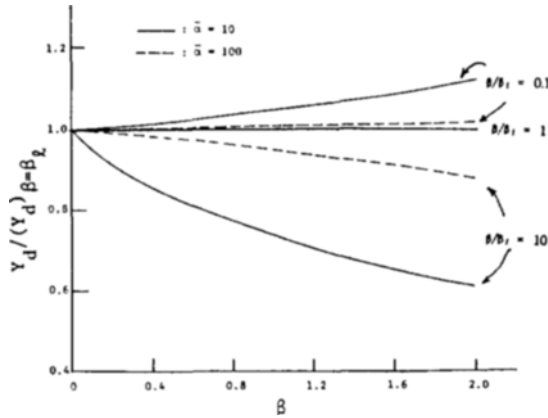


Fig. 3. Effect of using quartz columns of different pore structure on the dimensionless total pressure in the deposition chamber.

by Eq. (20) when the two quartz columns are made of different pore structure, is shown in Fig. 3 for $\omega \rightarrow 0$. The dimensionless pressure is normalized with respect to the dimensionless pressure for the case of $\beta = \beta_0$ in Fig. 3 to examine the effect of using quartz columns of different pore structure. It is readily seen that the effect is not significant when $\tilde{\alpha}$ is large as seen from the dashed curves for $\tilde{\alpha} = 100$. Some leverage for changing the pressure in the deposition chamber can be gained as seen from the solid curves for $\tilde{\alpha} = 10$. It is seen from these curves that the pressure in the deposition chamber can be reduced further by increasing the ratio β/β_0 and that the effect is larger for larger β . According to Eq. (12), the flux is given by

$$N = \alpha_u \left\{ (1 - Y_d) + \frac{\beta_u}{2} (1 - Y_d^2) \right\}; \quad \alpha_u \equiv \frac{D_u P_0}{R_g T L_u} \quad (22)$$

where Eq. (18) has been used and subscript u is for the upper quartz column.

Consider the first requirement on the flux for uniform concentration in the deposition chamber. If it is assumed that the silicon deposition on the deposition chamber walls [$\langle D \rangle$ in the Eq. (1)] is of the same magnitude as that on the wafers and that the area of the quartz column for the flux is the same as the total surface area of the wafers, one can rewrite Eq. (3) as

$$\frac{2G\rho_s/M}{N} < \delta; \quad G \text{ in length/time} \quad (23)$$

The kinetics of polycrystalline silicon growth at low pressure are well established [1] and the growth can be represented by first order kinetics. Since α_u in Eq.

(22) in terms of the concentration in the deposition chamber C_h is given by $(C_h/Y_d)(D_u/L_u)(T_h/T)$, Eq. (23) can be rewritten as

$$\frac{2k\rho_s Y_d/M}{\left(\frac{D_u}{L_u}\right)\left(\frac{T_h}{T}\right)\left[1 - Y_d + \frac{\beta_u}{2}(1 - Y_d^2)\right]} < \delta \quad (24)$$

where k is the apparent rate constant for the first order kinetics. Noting that $\rho_s = 2.33 \text{ g/cm}^3$, $T_h = 873\text{K}$ (typical growth temperature of 600°C for pure silane), $T = 298\text{K}$, $M = 29 \text{ g/gmol}$ and $(1 - Y_d) < [1 - Y_d + \beta_u(1 - Y_d^2)/2]$, one has from Eq. (24)

$$\frac{0.057kY_d}{\left(\frac{D_u}{L_u}\right)(1 - Y_d)} < \delta \quad (25)$$

According to Rosler [3], the polycrystalline growth rate is 125 nm/min at 600°C and 0.6 torr , which means that k is approximately $1.91 \text{ cm}^4/(\text{s} \cdot \text{gmol})$. Thus, the final form of Eq. (23) is:

$$\frac{0.11Y_d}{\left(\frac{D_u}{L_u}\right)(1 - Y_d)} < \delta; \quad (D_u/L_u) \text{ in cm/s} \quad (26)$$

Consider now the second requirement for the absence of free convection in the deposition chamber [Eq. (4)], which can be written as

$$\begin{aligned} Ra = GrPr &= \frac{\rho^2 g L^3}{\mu^2} \left(\frac{T_h - T}{T_h} \right) Pr \\ &= \frac{gb^3}{\mu^2} \left(\frac{T_h - T}{T_h} \right) \left(\frac{P_0 Y_d M}{R_g T_h} \right)^2 Pr \\ &= 1361b^3 (P_0 Y_d)^2 Pr < 1708; \\ L \text{ in cm, } P_0 \text{ in atm} \end{aligned} \quad (27)$$

where b is the distance between the hot region and the cooled section of the deposition chamber. The viscosity of silane at low pressure is $279 \text{ } \mu\text{P}$ at 873K according to the formula based on Lennard-Jones parameters for silane [7]. Thus, one has

$$b^3 (P_0 Y_d)^2 < 1.79 \quad (28)$$

if the Prandtl number is taken as 0.7 . If the distance b is taken as 10 cm , the requirement is given by

$$P_0 Y_d < 0.042; \quad P_0 \text{ in atm} \quad (29)$$

which means that the pressure in the deposition chamber be less than 30 torr .

The reactor design problem is now reduced to specifying P_0 , γ and D_u/L_u for a given desired growth rate with the constraints of Eqs. (26) and (29) if both the upper and lower quartz columns have the same

pore structure. Note that β is still a free parameter although it was not used in arriving at Eq. (26). Otherwise, one has an additional freedom to choose β . Suppose that the desired growth rate is 75 nm/min. Since 125 nm/min growth rate is possible at 0.6 torr and the growth is proportional to silane concentration, the pressure in the deposition chamber should be 3.6 torr at 600°C. This pressure certainly satisfies the constraint of Eq. (29), i.e. less than 30 torr. If, for example, (L_u/D_u) is chosen as 100 cm/s and δ as 0.001 (less than 0.1% change in the concentration in the deposition chamber), Y_d that satisfies the constraint of Eq. (26) is 0.5, which in turn means that $P_u=7.2$ torr. The Darcy permeability can be calculated [8] by

$$B_u = 10^{-9} \frac{\epsilon_p^3}{(1 - \epsilon_p)^2} \text{ (cm}^2\text{)}$$

where ϵ_p is the porosity of porous material. For a typical value of $\epsilon_p=0.4$, β is 0.127. Use of this value in Eq. (21) for a calculation of γ yields 1.06. Thus, the length of the upper quartz column is 5 cm and that of the lower column is 4.7 cm.

The case of polycrystalline silicon growth in nitrogen carrier gas is nearly the same as the growth with pure silane since γ , in this case is close to unity, i.e. $\gamma_{\text{SiH}_4}=0.94$. Therefore, the dimensionless total pressure behavior is similar to the one for the silane case. For rigorous treatment, however, Eq. (6) written for N_2 and SiH_4 needs to be solved with the aid of Eq. (7).

IN SUMMARY

The concept and design of a new CVD reactor has been presented for film deposition. An ideal CVD reactor is one in which a stagnant source gas of constant composition is provided without free or forced convection. The stagnancy is provided in the new reactor by feeding source gas by diffusion through a porous material. The constant composition is achieved by ensuring that the flux of the source gas is much larger than the growth rate. This condition is given by Eq. (3). The absence of free convection is achieved by maintaining the total pressure in the deposition chamber of the reactor low enough to satisfy Eq. (4). Solutions of Eqs. (6) and (7) provide the information necessary for the flux and the total pressure in the deposition chamber. For a given desired growth rate, these equations together with Eqs. (3) and (4) can be used to specify the total pressure in the upper empty chamber, the lengths of the upper and lower quartz columns, the total pressure and the partial pressure of

the source gas in the deposition chamber and the type of quartz or graphite with desired diffusivity and Darcy's permeability. It should be noted in this regard that if the same material of construction with the same pore characteristics is used for the quartz columns, only one set of equations applicable to both columns needs to be solved instead of one set for the upper column and another set for the lower column.

For polycrystalline silicon deposition, which follows first order kinetics, the growth rate can be set at any desired level as long as Eq. (4) is satisfied. This means that the deposition rate in the new CVD can be made much higher than is now practiced in an LPCVD reactor.

Lastly, a substantial reduction in the amount of source gas used for the polycrystalline deposition can be realized with the new reactor. For the mounted wafer configuration in an LPCVD reactor, the concentration has to be uniform over the entire number of wafers for the wafer-to-wafer uniformity. For the new reactor, it has to be uniform over a single wafer in the direction of diffusion. Therefore, a one order of magnitude reduction in the amount of silane required for polycrystalline silicon deposition could be realized, for instance, if the number of wafers in the LPCVD reactor exceeds one hundred.

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NOMENCLATURE

- A_c : cross sectional area of quartz column [L^2]
- A_w : wafer external surface area [L^2]
- b : distance between hot and cold region [L]
- B_u : Darcy permeability [L^2]
- C_m : concentration of the source gas in the upper chamber in Fig. 1 [mol/L^3]
- D : effective diffusivity of carrier gas [L^2/t]
- D_i : effective diffusivity of species i [L^2/t]
- D_l : D_i for key species in the lower quartz column in Fig. 1 [L^2/t]
- D_t : total deposition rate on walls [mol/t]
- D_u : effective diffusivity of key species in the upper chamber in Fig. 1 [L^2/t]
- E : total deposition rate [mol/t]
- g : gravity [L/t^2]

G : linear growth (deposition) rate [L/t]
 G_r : Grashof number
 G_t : molar growth rate [mol/t]
 k : rate constant for polycrystalline silicon deposition [L³/mol t]
 L : length of upper quartz column in Fig. 1 [L]
 L_r : length of lower quartz column in Fig. 1 [L]
 L_u : length of upper quartz column in Fig. 1 [L]
 M : molecular weight [M]
 M_c : M for carrier gas [M]
 M_i : M for species i [M]
 n : number of wafers
 N_i : molar flux of species i [mol/L²/t]
 N_{in} : N for incoming source gas [mol/L² t]
 N_{out} : N for outgoing source gas [mol/L² t]
 P_i : partial pressure of species i [M/L t²]
 P : total pressure [M/L t²]
 P_u : total pressure in the upper chamber in Fig. 1 [M/L t²]
 Pr : Prandtl number
 Ra : Rayleigh number
 R_g : gas constant [ML²/t² Tmol]
 T : temperature; cold region temperature [T]
 T_c : cold region temperature [T]
 T_h : hot region temperature [T]
 x : dimensionless axial coordinate for upper quartz column in Fig. 1
 x_l : dimensionless axial coordinate for lower quartz column in Fig. 1
 y_i : mole fraction of species i
 $(y_i)_f$: y_i of feed
 Y : dimensionless total pressure [M/Lt²]
 Y_d : Y in the deposition chamber in Fig. 1 [M/Lt²]
 z : axial coordinate for the upper quartz column in Fig. 1 [L]
 z_l : axial coordinate for the lower quartz column in Fig. 1 [L]

Greek Letters

α : quantity defined by Eq. (13) [mol/L³t]
 α_l : α in the lower quartz column in Fig. 1 [mol/L³ t]
 α_u : α in the upper quartz column in Fig. 1 [mol/L³ t]
 $\bar{\alpha}$: α_l/α_u

β : dimensionless quantity defined by Eq. (8)
 β_h : volume expansion coefficient evaluated at T_h [T⁻¹]
 β_l : β for the lower quartz column in Fig. 1
 γ : L/L_u
 γ_l : dimensionless quantity defined by Eq. (8)
 δ : dimensionless fractional allowance in concentration variation
 ϵ_p : porosity of the column material in Fig. 1
 μ : fluid viscosity [M/L_t]
 ν : kinematic viscosity of fluid [L²/t]
 ρ : fluid density [M/L³]
 ρ_s : solid density [M/L³]

Subscripts

c : cold
 h : hot
 l : lower quartz column in Fig. 1
 u : upper quartz column in Fig. 1

Dimension

L : length
 M : mass
 t : time
 T : temperature

REFERENCES

1. Bloem, J.: *J. Crystal Growth*, **50**, 581 (1980).
2. Westphal, G. H.: *J. Crystal Growth*, **65**, 105 (1983).
3. Rosler, R. S.: *Solid State Tech.*, p. 63, April (1977).
4. Fahien, R. W.: *Fundamentals of Transport Phenomena*, p. 541, McGraw-Hill, New York (1983).
5. Peng, C. F. and Stewart, W. E.: *Ind. Eng. Chem. Fund.*, **12**, 143 (1973).
6. Sze, S. M., ed.: *VLSI Technology*, McGraw-Hill, New York (1983).
7. Reid, R. C., Prausnitz, J. M. and Sherwood, T. K.: *The Properties of Gases and Liquids*, Chap. 9, 3rd ed., McGraw-Hill, New York (1983).
8. Blik, A., van Poelje, W. M., van Swaaij, W. P. M. and van Beckum, F. P. H.: *AIChE J.*, **31**, 1666 (1985).
9. Middleman, S. and Yeckel, A.: *J. Electrochem. Soc.*, **133**, 1951 (1986).