A Stagnant Layer Model for the Epitaxial Growth of Silicon from Silane in a Horizontal Reactor

F. C. Eversteyn, P. J. W. Severin, C. H. J. v.d. Brekel, and H. L. Peek

Philips Research Laboratories, N. V. Philips' Gloeilampenfabrieken, Eindhoven, Netherlands

ABSTRACT

Flow patterns have been made visible in a horizontal water-cooled epitaxial reactor by injection of TiO_2 particles into the gas flow. From these experiments, a stagnant layer model has been developed with which the epitaxial growth of silicon from silane can be described. In the case of a nontilted susceptor, the model predicts an appreciable nonuniformity in thickness along the susceptor, whereas a small angle of tilting of the susceptor should yield a much better uniformity in thickness (2% over a length of 22 cm). Experiments agree very well with the theoretical predictions of the model.

Silane as source for the epitaxial growth of silicon has several advantages over $SiCl_4$ and $SiHCl_3$: lower deposition temperature, less autodoping from substrate, less outdiffusion, and no back etching. On the other hand, silane is rather unstable and will decompose also on the hot reactor wall. This means that, as far as the construction is concerned, reactors for SiH_4 and $SiCl_4$ or $SiHCl_3$ will be different. For the time being, the most popular type is the horizontal reactor. The construction is rather simple. A large number of slices can be grown in one charge and the reactor is easy to load and unload. For SiH_4, a water-cooled reactor is preferable, because otherwise silicon would be deposited on the reactor wall, which would make measurement of the temperature of the substrates with an optical pyrometer impossible.

In the past, several authors have described the process of epitaxial growth, though mostly in vertical reactors. Bradshaw (1) developed a boundary layer model with which he was able to explain the growth experiments in a vertical reactor. Here the deposition efficiency is low, so that the decrease in concentration of the silicon compound along the susceptor can be neglected. Shepherd (2) described the epitaxial growth of silicon from $SiCl_4$ in a small horizontal reactor, assuming a parabolic gas flow in the reactor and omitting the decrease in $SiCl_4$ concentration in the length direction of the reactor. For large horizontal reactors, the deposition efficiency is found to be about 35%. This means that the decrease in concentration of the silicon compound along the reactor cannot be neglected. Taking this into account, Rundle (3) investigated the epitaxial growth of silicon from $SiCl_4$ and $SiHCl_3$, assuming that the gas flow in the reactor has a velocity component only along the axis of the reactor and that the whole system is at a constant temperature, ignoring temperature gradients.

In this paper, it is shown that these assumptions cannot be justified. From gas flow pattern experiments, it is seen that there is a thermal convection in the reactor vertically *overturning* the main gas flow and resulting in complete mixing of the gas. Directly above the heated susceptor, the temperature then drops over a thin stagnant layer which is excluded from the main gas flow. These experiments are discussed and a model derived for the epitaxial growth in a horizontal reactor is analyzed. The rate of epitaxial growth is calculated in the case of a nontilted and tilted susceptor. It is shown that there is close agreement between the predictions derived from the model and the experiments.

Investigation of the Flow Pattern

Experimental arrangement.—To make the flow pattern in the reactor visible, TiO_2 particles are injected into the gas stream by means of the equipment shown schematically in Fig. 1 (4). The horizontal reactor used for these experiments is the same as is considered later for the epitaxial deposition of silicon from silane. It consists of a water-cooled quartz tube with a rectangular cross section (5 x 4 cm) and a graphite susceptor, 30 cm long, 4 cm wide, 8 mm thick, positioned in a quartz boat which fits in the reactor. In our case, the free space between a side wall of the reactor and the quartz boat is about 0.5 mm. The quartz boat rests on the floor of the reactor. The free height above the susceptor is 2 cm. Dry hydrogen and hydrogen containing water vapor flows into the reactor where the temperature of the susceptor is 1050°C. By switching two electromagnetic valves, the dry hydrogen can be passed over $TiCl_4$. Mixing of the water vapor-loaded and the $TiCl_4$-loaded hydrogen causes the formation of TiO_2 particles, resulting in a white haze. The flow pattern in the reactor can then be observed. Photographs of the gas flow patterns have been made through the window on the right-hand end of the reactor (see Fig. 1) and are discussed below.

Experimental results.—In Fig. 2, the streamline pattern is given for the water-cooled reactor with the susceptor at room temperature. It is seen from this

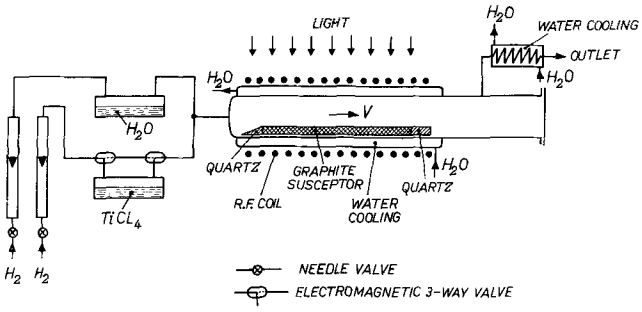

Fig. 1. Schematic view of the equipment to make gas flow patterns in the reactor visible.

Fig. 2. Streamline pattern above the nonheated susceptor ($T_S \approx 20$°C) with water-cooling around the tube (wall temperature approximately 10°C). Convective flow is visible under the influence of a small vertical temperature gradient. Rotation speed: about 1 rps.

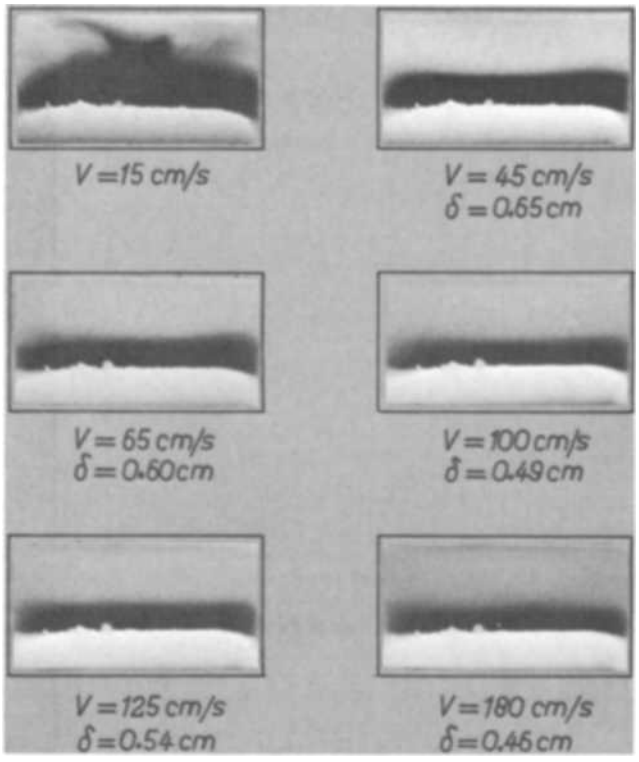

Fig. 3. Streamline patterns with TiO_2 at different gas velocities of hydrogen (corrected for heating up). Susceptor temperature 1050°C. Stagnant layer thickness decreases with increasing gas velocity.

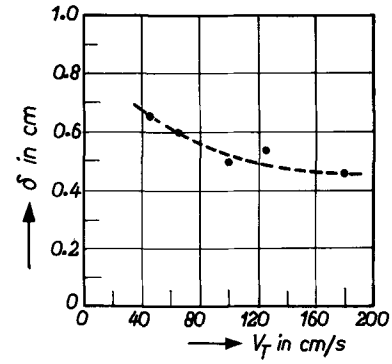

Fig. 4. Dependence of the stagnant layer thickness on the mean velocity of hydrogen in the reactor. Susceptor temperature, 1050°C.

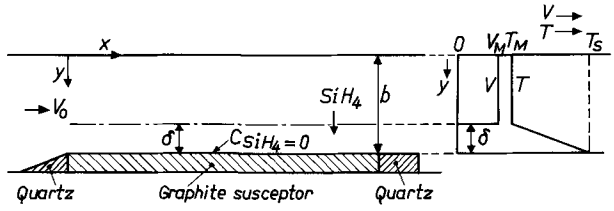

Fig. 5. Stagnant layer model for a horizontal reactor with a nontilted susceptor.

photograph that, although the vertical temperature gradient is very small ($\Delta T \approx 10$°C over about 2 cm), a convection flow already exists which, combined with the net gas flow, results in a double screw flow pattern. When the susceptor is heated up, the vertical temperature gradient increases. Due to a buoyancy force, increased in this way, a higher rotation velocity of the spirals results and the separate streamlines mix. This is shown in Fig. 3 where hydrogen with different gas velocities flows through the reactor in which the susceptor is at 1050°C. In the case of low gas velocity the convection flow lines can be seen separately, but at higher velocity the streamline pattern is restricted to a region separated from the substrate by a dark layer. The latter is indicative of a region where the gas velocity is zero and the incoming TiO_2 particles will flow only in a region separated from the susceptor by a stagnant layer. The thickness of the stagnant layer depends on the gas velocity, as can be seen from Fig. 4. When the linear velocity in the reactor is increased, the stagnant layer thickness tends to decrease.

Epitaxial Growth with a Nontilted Susceptor

Calculations on the model.—From the experiments described above, a model has been developed with which the epitaxial growth in a horizontal reactor can be physically described and mathematically analyzed.

In Fig. 5, a schematic diagram of a horizontal reactor with a nontilted susceptor is drawn with the assumptions implied in the model for epitaxial growth. They are mainly based on the gas flow pattern experiments with TiO_2 and explicitly given below:

I. Due to vertical *overturning* caused by buoyancy forces in the gas (thermal convection), the velocity, V_m, and the temperature, T_m, are constant over the height ($b - \delta$) of the reactor tube.

II. The temperature of the gas (T_m) is taken to be constant in the length direction of the reactor. This assumption is valid for a water-cooled reactor.

III. In the stagnant layer, the gas velocity is zero and the temperature increases linearly with y from T_m to the susceptor temperature T_s: $T(y) = T_s - (T_s - T_m)(b - y)/\delta$.

IV. SiH_4 diffuses through the stagnant layer to the susceptor. At the surface of the susceptor, the SiH_4 concentration is assumed to be zero since the unstable SiH_4 reaching the susceptor decomposes immediately at the temperature considered (1050°C). Thus, the deposition rate of silicon is diffusion controlled.

With these assumptions, the rate of epitaxial growth in a horizontal reactor has been calculated and turns out to be described by the following expression (see Appendix A):

$$G(x) = 7.23 \times 10^6 \frac{D_0 T_s p_0}{R T_0^2 \delta} \exp\left(-\frac{D_0 T_s x}{T_0 V_0 b \delta}\right) \quad [1]$$

where G is the growth rate of silicon (μm/min); D_0, the diffusion coefficient at 300°K of silane in hydrogen ($D_0 = 0.2$ cm^2/s); T_s, the susceptor temperature (°K); $T_0 = 300$°K; p_0, the partial pressure of silane (dynes/cm^2 $\approx 10^{-6}$ atm) at the inlet of the reactor; V_0, the mean velocity (cm/s) of the gas as calculated from the incoming gas flow and the free cross section of the tube; R is the gas constant (8.31 x 10^7 erg/°K). The free height above the susceptor is indicated by b (cm). The only unkown variable is δ, the thickness of the stagnant layer.

Experimental results.—Experiments were carried out on the epitaxial growth of silicon from silane in an atmosphere of hydrogen. Figure 6 shows an outline of the apparatus used for these experiments. Substrates were 0.01 ohm-cm (N- and P-type), polished slices of silicon single crystals. After etching the slices at 1200°C with HCl, the temperature of the susceptor is lowered to 1050°C, at which temperature the epitaxial growth is initiated by admitting a flow of hydrogen containing SiH_4. The temperature has been measured with an optical pyrometer and corrected for the emissivity of the silicon.

The growth rate, G, is obtained by measuring both the time of growth and the thickness of the deposited silicon layer. It has been verified that the growth rate, G, is a constant over the time of growth. The accuracy in the growth rate is affected by the accuracy with which the time of switching can be measured and by the accuracy of the thickness measurement method. The accuracy in time measurement amounts to ±0.2%.

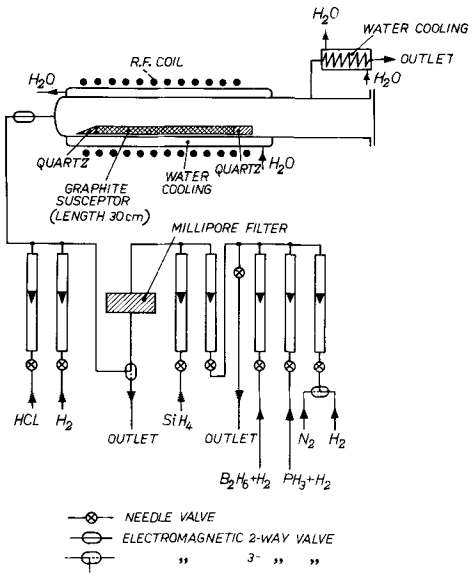

Fig. 6. Outline of the epitaxial growth equipment

The thickness has been measured by infrared multiple interference with a Hitachi EPI-G2 spectrophotometer. The data obtained allow for a final accuracy in thickness of ±0.5%. The evaluation technique used will be described by Severin (5).

According to Eq. [1], the growth rate is linearly dependent on the concentration of SiH_4 in the carrier gas. This was verified up to 10^{-1} vol % SiH_4 in a series of growth experiments. In each experiment, the slice was positioned at the same place in the reactor. The results of these experiments are given in Fig. 7. It is clear from this figure that growth rate varies linearly with the SiH_4 concentration.

Another series of experiments has been carried out to study the dependence of growth rate, G, on position x along the axis of the reactor. A typical result of these experiments is given in Fig. 8. It can be seen that, as required by Eq. [1] derived from the model, log G indeed decreases linearly with x.

Calculated and experimental values for the thickness of the stagnant layer.—From the slope of the line in Fig. 8 as well as from the value of G at $x = 0$ the thickness of the stagnant layer, δ, can be calculated. In Fig. 9, these values are given for gas flow rates (V_T) between 50 and 500 cm/s corresponding to V_o between 20 and 200 cm/s. This figure also shows the thickness of the stagnant layer as found by direct observation in gas flow pattern experiments with TiO_2 as shown in Fig. 3. Good agreement exists between these measured values of the stagnant layer thickness and the values of δ derived from $G(0)$. The values obtained from the slope turn out to be at variance. In order to proceed with the theory, a simple formal relation between δ and the velocity, V, is needed. A number of relationships have been tried but the relation:

$$\delta = \frac{A}{\sqrt{V_T}} - B \qquad [2]$$

with $A = 7$ cm$^{3/2}$ s$^{-1/2}$ and $B = 0.2$ cm appears to be the best compromise.

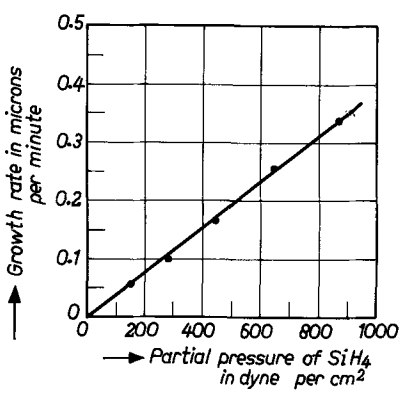

Fig. 7. Dependence of the growth rate on the partial pressure of silane (10^3 dynes/cm^2 relates to 0.1 vol % SiH_4).

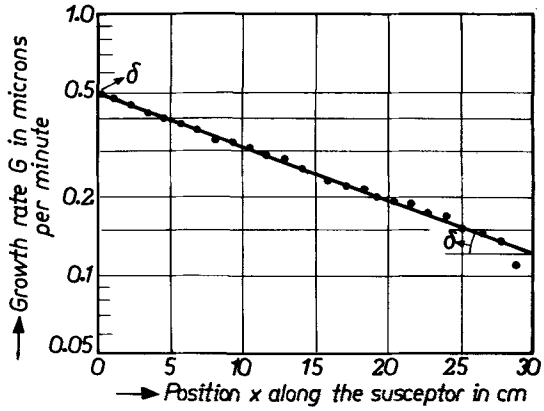

Fig. 8. Growth rate vs. position along the susceptor for V_o = 17.5 cm/s. Substrate temperature, 1050°C.

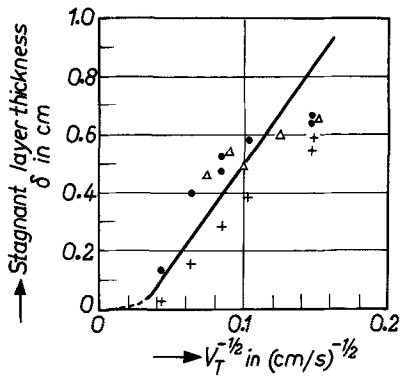

Fig. 9. Dependence of the stagnant layer thickness on the mean gas velocity in the reactor (corrected for heating up): △, from flow pattern experiments using TiO_2, + from the slope of the log $G(x)$-x curve; ●, from the growth rate at $x = 0$.

Theoretically and experimentally, the growth rate, G, decreases with increasing values of the position parameter, x. The magnitude of this effect depends on the gas flow rate. When Eq. [2] is inserted into Eq. [1], it follows that the slope of the log G *vs.* x curve as a function of V_o shows a rather flat minimum. This can be seen in Fig. 10. Some experimental data regarding this effect are also presented in this figure. It appears that the same trend is found in the experimental and in the calculated course of the slope of the log G *vs.* x curve as a function of the mean velocity, V_o. The differences in actual value of d log G/dx are due to the inaccuracies introduced by Eq. [2].

Increasing the gas flow rate from 9 to 34 cm/s results in a decrease of the slope of the log G *vs.* x curve. However, a further increase of the gas flow rate has no influence on the slope of the growth curve.

From these experiments and calculations, it is clear that it is impossible to get the same deposition rate in all positions on the susceptor. A smaller gradient in the thickness of deposited silicon should be obtainable with a stagnant layer thickness decreasing in the length direction of the reactor to compensate the effect of decreasing partial pressure of SiH_4. A gradually decreasing stagnant layer can be achieved by tilting the susceptor, as is dealt with below.

Reactor efficiency.—At this stage, it is interesting to consider the reactor efficiency, η, defined as the fraction

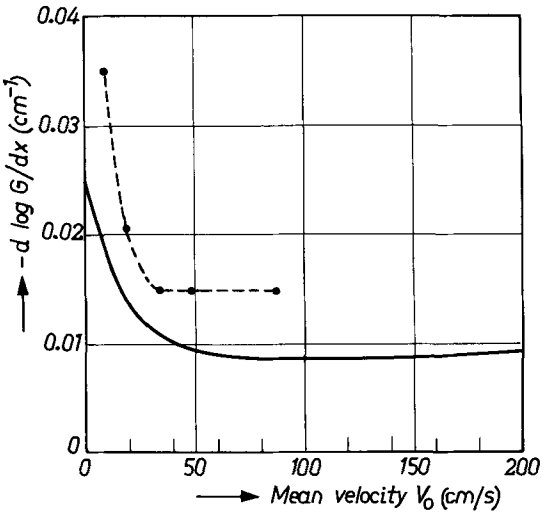

Fig. 10. Calculated values of the slope of the log $G(x)$-x curve from Eq. [1] and Eq. [2] at different values of the gas velocity (V_0). Some experimental values are also inserted into this figure (dotted line).

of the incoming SiH_4 decomposed on the hot susceptor. The reactor efficiency, η, can be expressed by

$$\eta = \frac{c\int_0^L J_y(x)\,dx}{n_0 V_0 b d} \cdot 100\% \qquad [3]$$

where L is the length of the susceptor, c the width of the susceptor, d the width of the reactor, $J_y(x)$ the molecular flux in the y-direction as a function of x, and $n_0 = p_0/kT_0$.

Integrating Eq. [3] yields

$$\eta = \frac{c}{d}\left\{1 - \exp\left(-\frac{D_0 T_s L}{b V_0 \delta T_0}\right)\right\} \cdot 100\% \qquad [4]$$

From Eq. [2] and Eq. [4], the dependence of the reactor efficiency, η, on the gas flow rate can be calculated. This is shown in Fig. 11. At low V_0, the efficiency, η, decreases with increasing V_0 and flattens up to the upper limit of the range of validity of Eq. [2]. Figure 11 also gives the experimental values of η, measured by gas chromatographic analysis of the incoming and outgoing gas. The experimental values are close to the calculated ones. As predicted, η decreases with increasing V_0 and tends to be constant at the right value of about 35%.

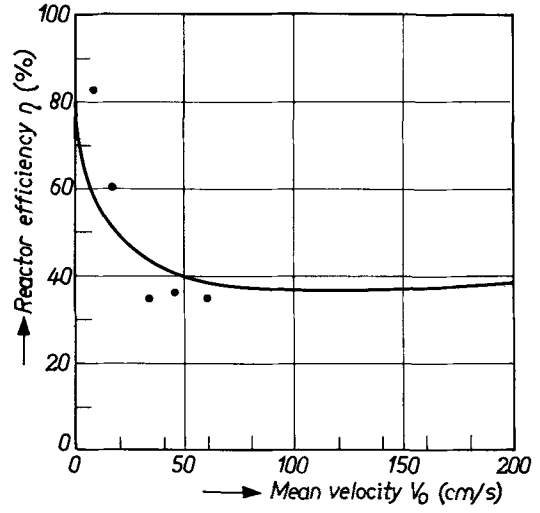

Fig. 11. Dependence of the reactor efficiency, η, on the mean gas velocity (V_0).

Epitaxial Growth of Silicon in a Horizontal Reactor with a Tilted Susceptor

Calculations on the model.—In Fig. 12, a schematic view is given of a horizontal reactor with a tilted susceptor. The assumptions for this model are the same as used for the nontilted susceptor. However, the decrease in stagnant layer thickness along the axis due to increasing gas velocity is taken into account. For this we use Eq. [2], which was found as a result of the experiments in a horizontal reactor with a nontilted susceptor. In the case of a tilted susceptor, the gas velocity is a function of x because of decreasing cross section of the free space above the susceptor

$$V_T(x) = \frac{V_0\, b\, T_m}{(b - x\tan\psi)\, T_0} \qquad [5a]$$

Of course, it has been avoided that the gas streams around and under the susceptor by filling up the space under and adjacent to the susceptor. The stagnant layer thickness, δ, and the temperature decay in the stagnant layer are supposed to be determined by the local velocity only and, hence, Eq. [2] and assumption III are modified to be a function of x

$$\delta(x) = \frac{7}{\sqrt{V_T(x)}} - 0.2 \qquad [5b]$$

and

$$T(x,y) = T_s + \frac{(T_s - T_m)\,(y - b + x\tan\psi)}{\delta(x)} \qquad [5c]$$

With these relations, the growth rate (μm/min) in a horizontal reactor with a tilted susceptor is found to be

$$G(x) = 7.23 \times 10^6 \frac{D_0 T_s p_0}{R T_0^2 \delta(x)} \exp \left\{ -\frac{2 D_0 T_s T_m}{4 g T_0^2 \tan\psi}\left(\delta(0) - \delta(x) + 0.2 \ln\frac{\delta(0)}{\delta(x)}\right)\right\} \qquad [6]$$

This formula is derived in Appendix B.

Experimental results.—With Eq. [6], calculations have been carried out for a fixed angle of tilting of 2.9° and different gas velocities of 10-70 cm/s. This is shown in Fig. 13. It is seen that at low gas velocity (V_0 = 10 cm/s) the growth rate decreases in the length direction, while at a high gas velocity ($V_0 > 40$ cm/s) even an increase in growth rate is found. Between these two values, a region should exist where the growth rate is nearly independent of position x. Experiments carried out under the same conditions as those for which these curves have been calculated show a very good agreement with the calculations, as can be seen in Fig. 14. With a gas velocity V_0 = 18 cm/s a decrease in growth rate along the axis is found, while at 48.7 cm/s an increase in growth rate is obtained. With a gas velocity of 34 cm/s, the growth rate is constant within 2% over a region of 22 cm (≈80% of the susceptor length). Not only the predicted trends in the deposition rates are found experimentally, but also the experimental value of the growth rate in the case of nearly even deposition is close to the calculated value. As can be seen from Fig. 13, we obtain for p_0 = 1000 dynes/cm² and V_0 = 34 cm/s a growth rate G = 0.43 μm/min. This implies for p_0 =

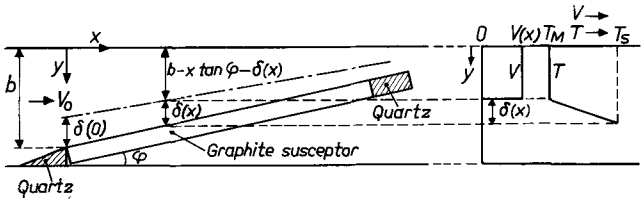

Fig. 12. Stagnant layer model for a horizontal reactor with a tilted susceptor.

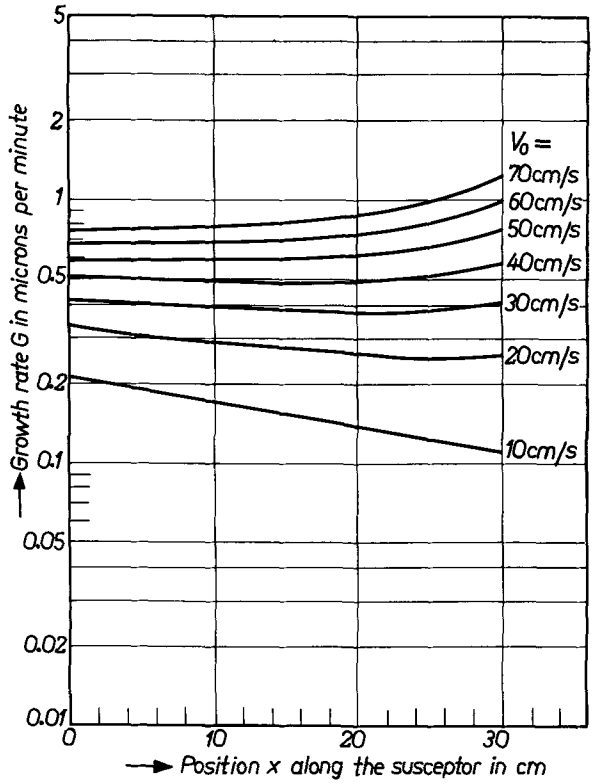

Fig. 13. Calculated values of the growth rate vs. position along the susceptor at different gas velocities for an angle of tilting of 2.9°. T_m = 700°K, T_s = 1350°K, b = 2.05 cm, p_o = 10^3 dynes/cm², D_o = 0.2 cm²/s.

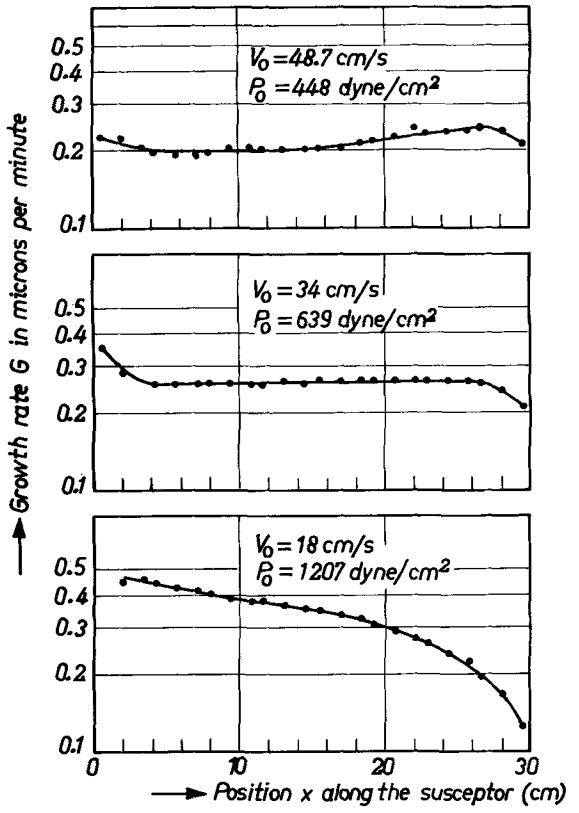

Fig. 14. Growth rate vs. position along the susceptor for an angle of tilting of 2.9° and V_o = 18, 34 and 48.7 cm/s, b = 2.05 cm, T_s = 1350°K.

639 dynes/cm² that G = 0.27 μm/min which agrees well with the experimental value G = 0.26 μm/min given in Fig. 14.

The decrease in growth rate at the end of the susceptor, as observed experimentally, is due to geometrical properties of the system where the flow and the temperature should match boundary conditions.

The uniformity in growth rate across the susceptor depends on how well the susceptor fits into the reactor. Variations in growth rate across the susceptor smaller than 5% have been found.

Discussion

From the experiments described in this paper, it is clear that the epitaxial growth of silicon from silane in a horizontal reactor can be adequately explained with a stagnant layer model. According to this model, above the heated susceptor a layer of stagnant gas is present in which the temperature gradient is very high (>100°C/mm). Above the stagnant layer the gas is vertically mixed by the high convection. When the vertical temperature gradient is small, the streamlines have been seen individually with a haze of TiO_2 and the rotation speed of the convection flow increases with the vertical temperature gradient. The experiment to which Fig. 2 refers yields for 5°C/cm a rotation speed of about 1 rps, as has also been calculated using an elementary theory in Appendix C. However, in the case of a heated susceptor, this temperature gradient is rather high resulting in so high a rotation speed that separate streamlines mix and that only a very small temperature gradient is left in the convective layer. In fact, we assume the temperature is uniform within this upper layer. This justifies the model as a whole.

The interpretation of the results presented in the sections on the experiments are based only on the equations of conservation of mass, which is expressed in the upper, convective, layer by the equation of continuity and in the lower, stagnant, layer by equating the silane diffusion flow to the growth rate of the silicon epitaxial layer, as shown in Appendixes A and B. The reactive species involved is supposed to be SiH_4.

We realize that a number of simplifying assumptions have been made in deriving the above results, particularly as regards the diffusion process. The diffusion of silane to the susceptor is rather complicated because several effects combine, which results in an effective diffusion coefficient of silane in hydrogen of 0.2 cm²/s at room temperature. In general, the temperature dependence of the diffusion coefficient is given by $D = D_o(T/T_o)^m$ with $1.75 \leq m \leq 2$. For mathematical convenience (see Appendix A), a value of $m = 2$ is used. However, a value of $m = 1.75$ would have been more reasonable because most gases diffuse in hydrogen with this m value (6). To obtain the same diffusion coefficient at high temperature (~1000°C), it is clear that the D_o value must be higher when $m = 1.75$ is used instead of $m = 2$.

Normally, thermal diffusion is neglected. In the case of a stagnant layer with a temperature gradient above 100°C/mm, this diffusion should be taken into account, as has been found by Bloem (7). Thermal diffusion results in a mass transport from the susceptor to the main gas flow and this diffusion flow is to be subtracted from the diffusion flow to the susceptor caused by a concentration gradient. The diffusion coefficient of silane in hydrogen due to a concentration gradient is not known, but on the basis of molecular weight a value of D_o = 0.6 cm²/s seems reasonable (2). The value of D_o = 0.2 cm²/s, with which the results described in this paper can be explained, should be considered as incorporating a correction for neglected thermal diffusion and the mathematically simplified temperature dependence of D.

APPENDIX A

As can be seen from Fig. 5, the space above the susceptor is divided into a convective and a stagnant layer. The derivation involves the equation of continuity in the convective layer and the diffusion equa-

tion in the stagnant layer. The solutions are coupled at the common boundary.

In an arbitrarily chosen point in the convective layer the equation of continuity should be satisfied, which reads, since only the stationary state is being considered and no decomposition of silane in the gas phase is assumed

$$\frac{\partial J_x(x)}{\partial x} + \frac{\partial J_y(x,y)}{\partial y} = 0 \qquad [A1]$$

Here, J_x is not a function of y, because due to convection all parameters concerned—temperature, T; density, n; partial pressure of silane, p; velocity, V—are uniform over the convective part of the cross section of the tube.

The second pertinent equation describes the diffusion process in the stagnant layer from the top surface to the substrate surface

$$J_y(x) = -D\frac{\partial n(x,y)}{\partial y} \qquad [A2]$$

According to elementary transport theory, $D \sim T^{1/2}/n(T)$ and a relation $D \sim T^{3/2}$ would be a reasonable assumption. Experimental evidence, however, is in favor of a $D \sim T^m$ relationship with $1.75 \leq m \leq 2$. We shall use here $m = 2$ for mathematical convenience so that $D = D_oT^2/T_o^2$.

Finally, we need the equation of state which, for the highly diluted silane, can be given by

$$p = nkT \qquad [A3]$$

where the temperature lapse rate in the stagnant layer is given by

$$T = T_s - (T_s - T_m)(b - y)/\delta \qquad [A4]$$

and $T = T_m$ in the convective part of the tube.

By straightforward calculation, it can be shown that Eq. [A2], [A3], and [A4] can be combined to give

$$J_y(x)T_o^2k\delta/D_o(T_s - T_m) = -T^2\frac{d\frac{p}{T}}{dT} \qquad [A5]$$

which can be integrated between $p = p(x,b - \delta)$, $T = T_m$ and $p(x,b) = 0$, $T = T_s$ to give

$$p(x) = J_y(x)kT_o^2\delta/D_oT_s \qquad [A6]$$

An alternative relation between $p(x)$ and $J_y(x)$ can be obtained by integrating Eq. [A1] over the cross section $0 < y < (b - \delta)$ and unit width, which yields, because J_y has a finite value only at the convective layer bottom surface and V and T are constant

$$J_y(x) = \frac{-(b - \delta)V}{kT_m}\frac{\partial}{\partial x}p(x) \qquad [A7]$$

When V, which is related to the inside parameters $(b - \delta)$ and T_m, is expressed in the corresponding outside parameters b and T_o as

$$V = V_obT_m/(b - \delta)T_o \qquad [A8]$$

Eq. [A7] can be written as

$$J_y(x) = \frac{-V_ob}{kT_o}\frac{d}{dx}p(x) \qquad [A9]$$

Eliminating $p(x)$ from the two solutions [A6] and [A9], we find

$$\frac{\partial}{\partial x}J_y(x) + \frac{D_oT_sJ_y(x)}{bV_oT_o\delta} = 0$$

which, with the boundary condition derived from [A6]

$$p(o) = \frac{J_y(o)kT_o^2\delta}{D_oT_s} \qquad [A10]$$

has the solution

$$J_y(x) = \frac{p_oD_oT_s}{kT_o^2\delta}\exp\left(-\frac{D_oT_sx}{bV_o\delta T_o}\right) \qquad [A11]$$

Using the density of silicon (2.33 g/cm^3) and the gas constant R instead of k, the growth rate of silicon in microns per minute can now easily be found to be

$$G(x) = 7.23 \cdot 10^6\frac{D_oT_sp_o}{RT_o^2\delta}\exp\left(-\frac{D_oT_sx}{bV_o\delta T_o}\right) \qquad [A12]$$

APPENDIX B

As has been made clear in the text, tilting the susceptor over a small angle, ψ, can be helpful to produce uniform thickness along the length of the reactor. As shown schematically in Fig. 12, the cross section varies both because of the tilting and because of the x-dependent stagnant layer thickness, $\delta(x)$. Equation [A8] should be written now as

$$V(x) = \frac{V_ob}{b - \delta(x) - x\tan\psi}\cdot\frac{T_m}{T_o} \qquad [B1]$$

where $V(x)$ is the velocity at high temperature in the reactor. From the experiments with a nontilted susceptor, the thickness of the stagnant layer, δ, has been found and plotted in Fig. 9 as a function of V_T where now

$$V_T(x) = \frac{V_obT_m}{(b - x\tan\psi)T_o} \qquad [B2]$$

and assuming a quasi-stationary state

$$\delta(x) = \frac{A}{\sqrt{V_T}(x)} - B \qquad [B3]$$

The temperature in the stagnant layer can be written as

$$T(x,y) = T_s + \frac{(T_s - T_m)(y - b + x\tan\psi)}{\delta(x)} \qquad [B4]$$

It can easily be verified that, apart from T being a function of x and y instead of y only, Eq. [A2], when combined with Eq. [A3] and [B4], yields the same Eq. [A5] and the same solution [A6]

$$p(x) = \frac{J_y(x)\delta(x)kT_o^2}{D_oT_s} \qquad [B5]$$

Under the actual conditions of the experiments, there is no decomposition on the wall. This means that the equation of continuity over the convective part of the cross section of the tube can be written as

$$J_y(x,b - x\tan\psi - \delta(x)) = -\frac{d}{dx}(b - \delta(x) - x\tan\psi)J_x \qquad [B6]$$

which yields for Eq. [B6] applying Eq. [A3]

$$-\frac{d}{dx}\frac{(p(x)V(x)(b - \delta(x) - x\tan\psi)}{kT_m} = J_y(x,b - x\tan\psi - \delta(x)) \qquad [B7]$$

or applying Eq. [B1]

$$J_y(x) = -\frac{V_ob}{kT_o}\frac{dp(x)}{dx} \qquad [B7a]$$

Eliminating $J_y(x)$ from Eq. [B5] and [B7a], it is found that

$$\frac{dp(x)}{dx} = -\frac{D_oT_sp(x)}{T_oV_ob\delta(x)} \qquad [B8]$$

which has the solution

$$p(x) = p(o)\exp\left[-\frac{D_oT_s}{T_oV_ob}\int_o^x\frac{dx}{\delta(x)}\right] \qquad [B9]$$

Hence, in terms of $J_y(x)$ from Eq. [B5]

$$J_y(x) = \frac{D_oT_sp(o)}{kT_o^2\delta(x)}\exp\left[-\frac{D_oT_s}{T_oV_ob}\int_o^x\frac{dx}{\delta(x)}\right] \qquad [B10]$$

It can easily be verified that for constant δ Eq. [B10] is equal to Eq. [A11], as it should be.

Integrating the exponent in Eq. [B10] with Eq. [B2] and [B3] yields the following rather cumbersome expression

$$J_y(x) = \frac{D_o T_s p_o}{kT_o^2 \delta(x)} \exp\left[-\frac{2D_o T_s T_m}{A^2 T_o^2 \tan\psi} \left(\delta(o) - \delta(x) + B \ln \frac{\delta(o)}{\delta(x)} \right) \right] \quad [B11]$$

Using the density of silicon (2.33 g/cm^3) and the gas constant R instead of k, the growth rate of silicon in microns per minute can now easily be found to be

$$G(x) = 7.23 \cdot 10^6 \frac{D_o T_s p_o}{RT_o^2 \delta(x)} \exp \left[-\frac{2D_o T_s T_m}{A^2 T_o^2 \tan\psi} \left(\delta(o) - \delta(x) + B \ln \frac{\delta(o)}{\delta(x)} \right) \right] \quad [B12]$$

APPENDIX C

In the experiments with small vertical temperature gradient (see Fig. 2), the TiO_2 particles were visibly moving upward along streamlines. This is a basic phenomenon in meteorology (8). The velocity y of the rising lump of gas can be assessed in the following way.

Denoting the vertical coordinate by y and the acceleration of gravity by g, the force experienced by a given parcel of gas, primed if not in equilibrium with the environment, increases with the deviation from hydrostatic equilibrium. This yields in a first order of approximation a force proportional to this deviation, or, since the unprimed quantities refer to equilibrium

$$\ddot{y} = -g \frac{\rho - \rho'}{\rho'} = -g \frac{\Delta T}{T} \quad [C1]$$

Integrating this equation, it is found that

$$y_o - y = \frac{g \Delta T t^2}{2T} \quad [C2]$$

which yields, when the height of the tube is b and the characteristic time to traverse this distance is τ

$$\tau = \left(\frac{2bT}{\Delta T g} \right)^{1/2} \quad [C3]$$

Numerically, this fits well: $b = 2$ cm, $\Delta T \approx 10°$K, $T = 300°$K, $g \approx 10^3$ cm/sec^2 yields $\tau \approx 1/3$ sec, which means, taking into account the lateral and downward motion, 1 rps. This has been observed experimentally.

NOMENCLATURE

x,	position on the susceptor
y,	position above the susceptor
$G(x)$,	growth rate at position x on the susceptor, μm min^{-1}
$J_x(x,y)$,	molecular flux of SiH_4 in the x-direction as a function of x and y, cm^{-2} s^{-1}
$J_y(x,y)$,	molecular flux of SiH_4 in the y-direction as a function of x and y, cm^{-2} s^{-1}
δ,	stagnant layer thickness, cm
$\delta(x)$,	stagnant layer thickness at position x on the susceptor, cm
$\delta(o)$,	stagnant layer thickness at $x = 0$, cm
c,	width of the susceptor, cm
d,	width of the reactor, cm
b,	free height of the reactor at $x = 0$, cm
L,	length of the susceptor, cm
T_m,	temperature of the gas in the convective part of the reactor, °K
T_s,	susceptor temperature, °K
$T(x,y)$,	temperature of the gas in the stagnant layer as a function of x and y
T_o,	300°K
$p(x)$,	partial pressure of silane at the position x, dynes cm^{-2}
p_o,	partial pressure of silane at the inlet of the reactor, dynes cm^{-2}
$V(x,y)$,	linear gas velocity in the reactor as a function of x and y, cm s^{-1}
V_o,	mean gas velocity in the case of a nonheated susceptor cm s^{-1}
V_m,	linear gas velocity in convective part of the reactor, cm s^{-1}
$V_T(x)$,	mean gas velocity in reactor as a function of x (corrected for heating up), cm s^{-1}
R,	gas constant (= 8.31 x 10^7 erg °K^{-1})
k,	Boltzmann constant (= 1.3 x 10^{-16} erg °K^{-1})
ρ,	gas density, g cm^{-3}
n,	number of silane molecules per unit volume, cm^{-3}
D_o,	diffusion coefficient for silane at 300°K (corrected value for thermodiffusion: 0.2 cm^2 s^{-1})
η,	deposition efficiency, %
t,	time, s
ψ,	angle of tilting of the susceptor
g,	acceleration of gravity, cm s^{-2}
τ,	characteristic time for convective flow, s

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