DIFFUSION MODEL FOR DEPOSITION OF EPITAXIAL GaAs LAYERS PREPARED BY THE MOCVD METHOD

Jindřich LEITNER[a], Petr VOŇKA[b], Josef STEJSKAL[c], Přemysl KLÍMA[c] and Rudolf HLADINA[c]

[a] *Department of Materials for Electronics, Prague Institute of Chemical Technology, 166 28 Prague 6*
[b] *Department of Physical Chemistry, Prague Institute of Chemical Technology, 166 28 Prague 6*
[c] *Research Institute of Radiocommunication A. S. Popov, 142 21 Prague 4*

Received November 29, 1990
Accepted February 13, 1991

The authors proposed and treated quantitatively a kinetic model for deposition of epitaxial GaAs layers prepared by reaction of trimethylgallium with arsine in hydrogen atmsosphere. The transport of gallium to the surface of the substrate is considered as the controlling process. The influence of the rate of chemical reactions in the gas phase and on the substrate surface on the kinetics of the deposition process is neglected. The calculated dependence of the growth rate of the layers on the conditions of the deposition is in a good agreement with experimental data in the temperature range from 600 to 800°C.

The metalorganic chemical vapour deposition (MOCVD) method represents nowadays the most frequently used method for the preparation of epitaxial layers and structures of semiconductors of the type $A^{III}B^{V}$ used in electronics and optoelectronics, especially on the production scale[1,2]. A great attention is therefore paid to the study of both fundamental processes and technical aspects of the deposition of epitaxial layers.

Our preceding work[3] was devoted to results of a detailed thermodynamic analysis of the Ga–As–C–H system. On the assumption that a local thermodynamic equilibrium is established between the formed solid phase and the adjacent gaseous phase, it is possible to find from equilibrium calculations such initial conditions, at which the desired solid phase (GaAs) is the only thermodynamically stable phase in the studied system. However, equilibrium calculations do not enable us to predict, in most cases, the rate of deposition, but only to estimate its upper limit. Therefore, our present work is devoted to a kinetic model of deposition of epitaxial GaAs layers prepared by the MOCVD method.

The results of a kinetic analysis of the deposition process for various types of epitaxial reactors were published by a number of authors[4–17], especially during the

last years. Various models are distinguished mainly by the extent of treatment of hydrodynamic conditions in the reactor and of the chemical conversion of the system under study. The most elaborated model appears to be that of Tirtowidjojo and Pollard[13-15], involving, besides multicomponent heat and mass transfer, as many as 232 chemical reactions which can proceed homogeneously in the gas phase and 115 reactions proceeding at the substrate surface. Also Jensen et al.[7,8] and Coltrin and Kee[17] consider a simultaneous influence of transport and kinetic processes on the deposition rate of GaAs layers. Based on these sophisticated models, it is possible to predict or to correlate the growth rates of the GaAs layers in a wide range of the deposition conditions (e.g. temperature from 450 to 1 050°C). In the temperature range about 600 – 800°C, the growth rate can be predicted by using simpler models based on the assumption that the kinetic characteristic of the deposition process is governed mainly by transport of gallium to the substrate surface[5,6,11,12].

The aim of the present work is the formulation and quantitative treatment of the diffusion model for the deposition of epitaxial GaAs layers on the assumption that the transport of gallium to the substrate surface is the controlling process. The commonly used type of a rectangular reactor with a cold wall will be assumed and the starting substances are trimethylgallium (TMGa), arsine, and hydrogen.

THEORETICAL

The results of experimental studies of the growth rate of GaAs layers at various deposition conditions[18-25] suggest that at the usual conditions (atmospheric pressure, 600 – 800°C, $x^0(\mathrm{TMGa}) = 10^{-4} - 10^{-3}$, initial ratio of $AsH_3/\mathrm{TMGa} = 10 - 100$) the growth rate is little dependent on the temperature, increases almost linearly with the initial concentration of TMGa but is independent of the initial AsH_3 concentration, increases with the rate of flow of the gas phase, and is independent of the crystallographic orientation of the substrate. These results suggest that the transport of gallium to the substrate surface has a dominant influence on the deposition rate.

Trimethylgallium is very unstable at elevated temperatures and may undergo partial or complete decomposition during transport to the surface of the substrate. Its thermal decomposition was studied by many authors[26-34], however only some partial steps of the complicated mechanism were satisfactorily elucidated. Since sufficiently accurate data for evaluation of the kinetics of thermal decomposition of TMGa are not available and the coefficients of diffusion and thermal diffusion of decomposition products can only roughly be estimated, the transport of gallium is involved in our model only in the form of TMGa.

A two-dimensional model (2D model, Fig. 1) is used to determine the distribution of the deposition velocity along the reactor axis. The 2D model does not take into account the deformation of the velocity and temperature fields by the side walls of the reactor and the formation of turbulence as a result of thermal convection. We assume that a Poiseuille velocity profile is established in the flowing gas, so that the solution of Navier–Stokes equations, which would be tedious, is unnecessary. The simplified system of partial differential equations describing stationary heat and mass transfer in the reactor reads[35] (cf. Symbols)

$$\frac{\partial}{\partial x}(\varrho u) = 0 \tag{1}$$

$$u = 6u_0(y/h)(1 - y/h) \tag{2}$$

$$\varrho u \frac{\partial}{\partial x}(c_p T) = \frac{\partial}{\partial x}\left(k \frac{\partial T}{\partial x}\right) + \frac{\partial}{\partial y}\left(k \frac{\partial T}{\partial y}\right) \tag{3}$$

$$cu \frac{\partial x_{\mathrm{TMGa}}}{\partial x} = \frac{\partial}{\partial x}\left[cD_{\mathrm{TMGa}}\left(\frac{\partial x_{\mathrm{TMGa}}}{\partial x} + k_{\mathrm{T}} \frac{\partial \ln T}{\partial x}\right)\right] + \frac{\partial}{\partial y}\left[cD_{\mathrm{TMGa}}\left(\frac{\partial x_{\mathrm{TMGa}}}{\partial y} + k_{\mathrm{T}} \frac{\partial \ln T}{\partial y}\right)\right], \tag{4}$$

where

$$\varrho = \overline{M}p/RT \tag{5a}$$

and

$$c = p/RT\,. \tag{5b}$$

The boundary conditions for this system can be written as (Fig. 1)

$$y = 0,\ x < 0:\ u = 0,\ v = 0,\ T = T_c,\ N_{\mathrm{TMGa}} = 0 \tag{6a}$$

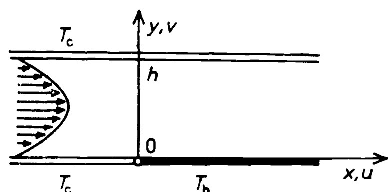

FIG. 1
Schematic representation of a 2D epitaxial reactor

$$y = 0, x \geqq 0: u = 0, v = 0, T = T_h, x_{TMGa} = 0 \quad (6b)$$

$$y = h: u = 0, v = 0, T = T_c, N_{TMGa} = 0 \quad (6c)$$

$$0 < y \leqq h, x = -5h: u = 6u_0(y/h)(1 - y/h), v = 0 .$$
$$T = T_c, x_{TMGa} = x^0_{TMGa} \quad (6d)$$

and the flux of TMGa along the y axis with respect to the fixed coordinates is given by the equation

$$N_{TMGa} = -cD_{TMGa}\left(\frac{\partial x_{TMGa}}{\partial y} + k_T \frac{\partial \ln T}{\partial y}\right). \quad (7)$$

The boundary condition (6d) is formulated for $x = -5h$ (ref.[11]), enabling a realistic modification of the velocity profile at a discontinuous change of the temperature of the reactor bottom (for $x = 0$) to be effected.

The growth rate of the GaAs layer can be calculated as

$$G(\mu m/s) = 10^4 (M_{GaAs}/\varrho_{GaAs}) (N_{TMGa})_{y=0} , \quad (8)$$

where $M_{GaAs} = 144{\cdot}64$ g mol^{-1} and $\varrho_{GaAs} = 5{\cdot}31$ g cm^{-3} (ref.[36]).

COMPUTATION AND DATA

The system of equations (1)–(5) was solved by the finite-difference method[37] with the usual choice of $\Delta x = 0{\cdot}05-0{\cdot}1$ mm and $\Delta y/h = 0{\cdot}02-0{\cdot}04$ taking into account the stability of the solution. It turned out that when the derivatives with respect to x on the right-hand sides of Eqs (3) and (4) are neglected, the results are not appreciably changed and the implicit scheme used is stable for any choice of Δx and Δy. A considerable simplification follows from the fact that the concentrations of the starting substances (TMGa and AsH_3) are very low. Accordingly, the physical properties of the gaseous mixture can be approximated by the properties of pure hydrogen, and further the calculation of the velocity and temperature fields in the reactor can be separated from the calculation of the concentration gradient of TMGa in the region above the subsrate.

The temperature dependence of the heat conductivity of hydrogen was expressed as

$$k(W/cm\ K) = 1{\cdot}05 \,.\, 10^{-3} + 3{\cdot}21 \,.\, 10^{-6}T + 2{\cdot}5 \,.\, 10^{-11}T^2 \quad (9)$$

and the specific heat capacity of hydrogen $c_p(H_2) = 14{\cdot}824$ J/g K (mean value in the temperature range 300–1 100 K) (ref.[38]). Since the gaseous mixture is very dilute, the effective values of the diffusion coefficient and thermal diffusion coefficient of

TMGa can be replaced by the values for the binary system $TMGa/H_2$. The value of D_{TMGa/H_2} at 300 K was calculated from the equation following from the Chapman–Enskog kinetic theory[39]. Based on the parameters of the Lennard–Jones potential for hydrogen[39] and TMGa (Ref.[40]), we found $D_{TMGa/H_2} = 0.349 \text{ cm}^2/\text{s}$. This value is in good agreement with $D_{TMGa/H_2} = 0.373 \text{ cm}^2/\text{s}$ calculated from the Fuller equation[39] with the use of the diffusion volume of hydrogen[39] and of that of TMGa calculated by adding the contributions for hydrogen, carbon[39] and gallium[41]. The Fuller equation can also be used to estimate the upper limit of the diffusion coefficient of TMGa in such a way that the contribution for gallium is neglected in calculating the diffusion volume of TMGa. The resulting value is $0.438 \text{ cm}^2/\text{s}$. In this connection, the experimentally found value[42] $D_{TMGa/H_2} = 0.598 \text{ cm}^2/\text{s}$ at 300 K seems to be too high. We employed the temperature dependence of the diffusion coefficient

$$D_{TMGa/H_2}(\text{cm}^2/\text{s}) = 1.614 \cdot 10^{-5} T^{1.75} \quad (10)$$

derived from the above value of $D_{TMGa/H_2} = 0.349 \text{ cm}^2/\text{s}$ (300 K) and the temperature dependence of the diffusion coefficient given by the Fuller equation. The thermal diffusion coefficient k_T in Eq. (*4*) depends on the composition; in binary gas mixtures we have

$$k_T = \alpha x_i (1 - x_i) . \quad (11)$$

The temperature dependence of the thermal diffusion coefficient was expressed by the equation

$$\alpha = 0.93 + 6.44 \cdot 10^{-4} T - 2.57 \cdot 10^{-7} T^2 - 3.82 \cdot 10^4 T^{-2} \quad (12)$$

which was obtained by correlating the values of α calculated by Holstein[40] with the simplified relation obtained from the Chapman–Enskog kinetic theory.

RESULTS AND DISCUSSION

Our model was tested by using a set of deposition conditions taken from the literature[5]. The calculated dependences of the layer growth rate on the distance from the leading edge of the substrate were compared with experimental data (Figs 2 and 3). It can be seen from the diagrams that the calculated values are at higher gas flow rates always lower than the experimental ones, especially at the inlet portion of the substrate in a reactor in which the ratio of the width to the depth, b/h, is smaller. Ouazzani et al.[11] introduced a correction in calculating the deposition rate, involving the influence of b/h on the mean linear velocity of the gas phase (for the 2D model, the ratio of b/h is considered infinite). For $b/h = 6.3$ (Fig. 2), the effective linear velocity of the gas phase is given by $u_0^* = 1.11\ u_0$ and for $b/h = 2.8$ (Fig. 3) by

$u_0^* = 1{\cdot}26\, u_0$. The deposition rates thus calculated are for various x values summarized in Table I together with the experimental values and the values calculated without the mentioned correction. It can be seen that the correction leads to a moderate increase of the deposition rate along the substrate.

To obtain an idea about the role of the diffusion coefficient of TMGa in the calculations and about the possible error in the deposition rate due to neglection of any other gaseous gallium compounds, some calculations were carried out with the diffusion coefficient value increased by 50% (Table I). It can be seen that this higher value leads to an increase of the deposition rate at the inlet portion of the substrate, i.e. to a stronger exhaustion of Ga from the gas phase, which in turn leads to a decrease of the deposition rate in the region of higher x values. We estimated the diffusion coefficients of dimethylgallium, $D_{DMGa/H_2} = 0{\cdot}422\ cm^2/s$, and of methylgallium, $D_{MGa/H_2} = 0{\cdot}503\ cm^2/s$, in hydrogen at 300 K based on the Fuller equation. These are, respectively, by 14 and 35% higher than the value for TMGa.

As mentioned above, neglection of the derivatives with respect to x on the right-hand sides of equations (*3*) and (*4*) (representing the heat transport by conduction and diffusion of TMGa in the direction of the gas flow) does not lead to significant deviations in the calculated deposition rate. This circumstance was studied by Ouazzani et al.[11], who concluded that the axial diffusion of TMGa can be neglected if the Peclet number is at least equal to 5. This condition was satisfied in our case.

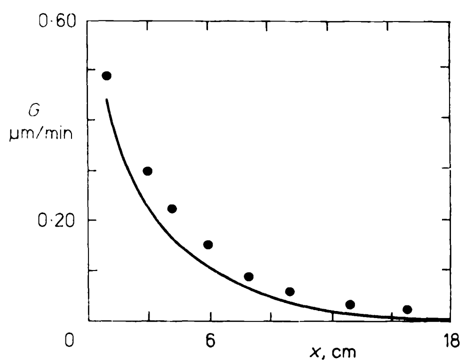

FIG. 2

Comparison of calculated (———) and experimental (●) dependence of the growth rate of GaAs layers, G, on the distance from the substrate border. Parameters: $b = 5$ cm, $h = 0{\cdot}8$ cm, $p = 101{\cdot}3$ kPa, $T_c = 300$ K, $T_h = 973$ K, $u_0 = 6{\cdot}9$ cm/s, $x^0_{TMGa} = 5{\cdot}92 \cdot 10^{-4}$

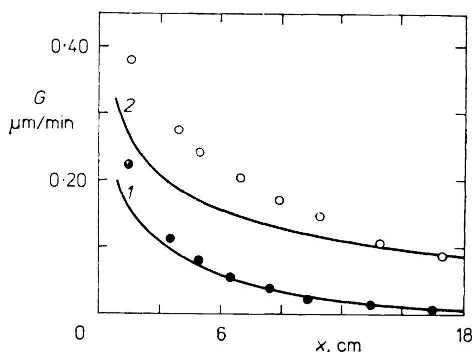

FIG. 3

Comparison of calculated (———) and experimental (●, ○) dependences of the growth rate of GaAs layers on the distance from the substrate border. Parameters: $b = 5$ cm, $h = 1{\cdot}8$ cm, $p = 101{\cdot}3$ kPa, $T_c = 300$ K, $T_h = 973$ K, $u_0 = 2{\cdot}0$ cm/s (●) and 9·2 cm/s (○), $x^0_{TMGa} = 4{\cdot}93 \cdot 10^{-4}$

To find out whether our kinetic model can predict the growth rate of the GaAs layers at widely varying deposition conditions, the growth rates were calculated at atmsopheric pressure in the temperature range 500–900°C for linear velocity of the gas phase $u_0 = 1-20$ cm/s and initial concentration $x^0_{TMGa} = 10^{-4}-10^{-3}$. The height of the reactor was 2 cm. The results of calculations are illustrated in Figs 4–6. The following conclusions can be drawn:

1. In the temperature range about 600–800°C, our model describes well the experimental dependence of the deposition rate on the temperature. The apparent activation energy, about 0·8 kJ, corresponds to the value measured in the interval 0–10 kJ (refs[5,19,20,22]). At other temperatures, however, the model does not give the experimentally found decrease of the deposition rate.

2. At 700°C, the model describes well the dependence of the deposition rate on the linear velocity of the gas phase in the reactor. The calculated data are in accord

TABLE I

Comparison of calculated and experimental values of the growth rate of GaAs layers at various deposition conditions ($p = 101{\cdot}325$ kPa and $T_h = 973$ K in all cases)

x cm	G(exp) μm/min	G(calc), μm/min[a]		
		(1)	(2)	(3)
$h = 0{\cdot}8$ cm; $u_0 = 6{\cdot}9$ cm/s; $x^0_{TMGa} = 5{\cdot}92 \,.\, 10^{-4}$				
1·0	0·49	0·436	0·456	0·554
6·0	0·16	0·102	0·119	0·074
16·0	0·02	0·008	0·012	0·002
$h = 1{\cdot}8$ cm; $u_0 = 2{\cdot}0$ cm/s; $x^0_{TMGa} = 4{\cdot}93 \,.\, 10^{-4}$				
1·5	0·23	0·158	0·164	0·182
5·0	0·08	0·073	0·088	0·073
14·0	0·02	0·015	0·025	0·007
$h = 1{\cdot}8$ cm; $u_0 = 9{\cdot}2$ cm/s; $x^0_{TMGa} = 4{\cdot}93 \,.\, 10^{-4}$				
1·5	0·38	0·279	0·297	0·341
5·0	0·24	0·170	0·187	0·211
14·0	0·11	0·102	0·116	0·115

[a] (1) Calculated values; (2) calculated values with correction for finite value of b/h; (3) calculated values for $D_{TMGa/H_2} = 2{\cdot}421 \,.\, 10^{-5}\, T^{1.75}$ (diffusion coefficient by 50% higher) without correction for finite value of b/h.

with experiments[19,22] indicating that beginning from a certain value the dependence of the deposition rate on the square root of the linear velocity is practically linear.

3. At 700°C, the model gives the correct dependence of the deposition rate on the initial concentration of TMGa, which is practically linear[5,19,20,25].

4. As follows already from the formulation of the model, the calculated deposition rate does not depend on the initial concentration of AsH_3 and crystallographic orientation of the substrate.

It is apparent from our results that in the temperature range about 600–800°C, commonly used for the deposition of epitaxial GaAs layers, a relatively simple dif-

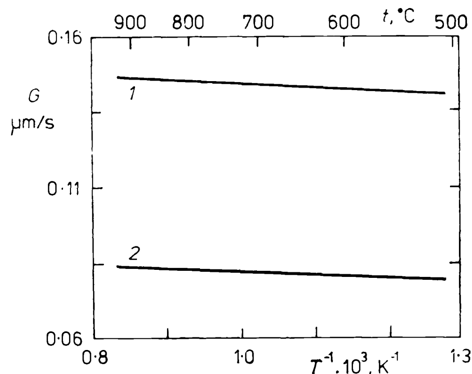

Fig. 4

Dependence of the growth rate of GaAs layers on temperature. Parameters: $x^0_{TMGa} = 5 \cdot 10^{-4}$, $u_0 = 5$ cm/s; *1* $x = 4$ cm, *2* $x = 12$ cm

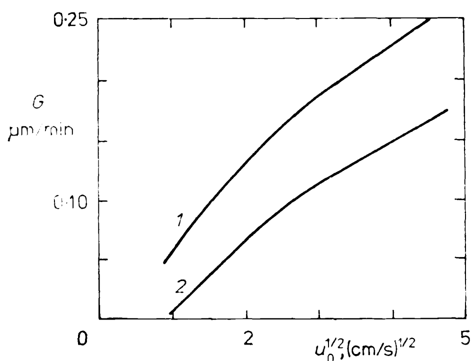

Fig. 5

Dependence of the growth rate of GaAs layers on the linear velocity of gas phase in the reactor. Parameters: $t = 700$°C, $x^0_{TMGa} = 5 \cdot 10^{-4}$; *1* $x = 4$ cm, *2* $x = 12$ cm

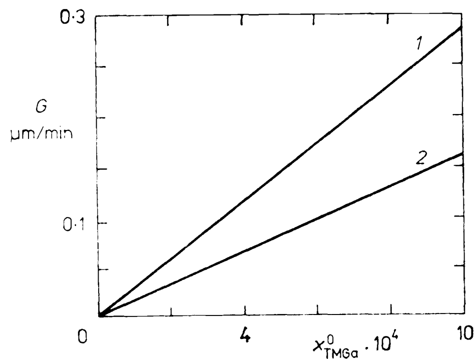

Fig. 6

Dependence of the growth rate of GaAs layers on the initial concentration of TMGa. Parameters: $t = 700$°C, $u_0 = 5$ cm/s; *1* $x = 4$ cm, *2* $x = 12$ cm

fusion model can be used to predict the deposition rate, assuming that the transport of gallium to the substrate surface is the controlling step. In a wider temperature range, the chemical reaction rates proceeding in the gas phase and on the substrate surface become important for the description of the deposition kinetics and therefore they should be taken into account.

SYMBOLS

b	width of reactor, cm
c	molar concentration, mol/cm^3
c_p	specific heat capactiy at constant pressure, J/g K
D_i	effective diffusion coefficient of species i, cm^2/s
$D_{i,j}$	binary diffusion coefficient, cm^2/s
G	linear growth velocity of a layer, µm/min
h	reactor height, cm
k	heat conductivity, W/cm K
k_T	thermal diffusion coefficient
M_i	molar mass of species i, g/mol
$\bar{M}$	mean molar mass of a gas mixture, g/mol
N_i	flux of species i referred to fixed coordinates, $mol/s\ cm^2$
p	pressure, kPa
Pe	Péclet number, equal to $u_0 h/D$
R	universal gas constant (8·314 J/mol K)
T	absolute temperature, K
T_c	temperature of the cold wall of the reactor, K
T_h	temperature of the substrate, K
t	temperature, °C
u	x-component of the linear velocity of flow, cm/s
u_0	mean linear velocity of flow of the gas phase, cm/s
u_0^*	effective value of u_0, cm/s
v	y-component of the linear veloctiy of flow, cm/s
x	coordinate parallel to the reactor axis, cm
x_i	molar fraction of species i
y	coordinate normal to the substrate, cm
z	dimensionless coordinate, equal to y/h
α	thermal diffusion coefficient
ϱ	density, g/cm^3

REFERENCES

1. Ludowise M. J.: J. Appl. Phys. *58*, R31 (1985).
2. Schumaker N. E., Stall R. A., Wagner W. R., Polgar L. G.: J. Metals *38*, 41 (1986).
3. Leitner J., Voňka P., Mikulec J., Stejskal J., Klíma P.: J. Cryst. Growth *112*, 437 (1991).
4. Kusumoto Y., Hayashi T., Komiya S.: Jpn. J. Appl. Phys. *24*, 620 (1985).
5. van de Ven J., Rutten G. M. J., Raaijmakers M. J., Giling L. J.: J. Cryst. Growth *76*, 352 (1986).
6. Moffat H., Jensen K. F.: J. Cryst. Growth *77*, 108 (1986).

7. Mountziaris T. J., Jensen K. F.: *Mater. Res. Soc. Symp. Proc.* (C. W. Tu, V. D. Mattera and A. C. Gossard, Eds), Vol. 131, p. 116. MRS, Pittsburgh 1989.
8. Jensen K. F., Mountziaris T. J., Fotiadis D. I.: *Mater. Res. Soc. Symp. Proc.* (C. W. Tu, V. D. Mattera and A. C. Gossard, Eds), Vol. 145, p. 107. MRS, Pittsburgh 1989.
9. Sato M., Suzuki M.: J. Electrochem. Soc. *134*, 1540 (1987).
10. Sato M., Suzuki M.: Jpn. J. Appl. Phys. *26*, 428 (1987).
11. Ouazzani J., Chiu K.-Ch., Rosenberger F.: J. Cryst. Growth *91*, 497 (1988).
12. Ouazzani J., Rosenberger F.: J. Cryst. Growth *100*, 545 (1990).
13. Tirtowidjojo M., Pollard R.: J. Cryst. Growth *93*, 108 (1988).
14. Tirtowidjojo M., Pollard R.: J. Cryst. Growth *98*, 420 (1989).
15. Tirtowidjojo M., Pollard R.: *Mater. Res. Soc. Symp. Proc.* (C. W. Tu, V. D. Mattera and A. C. Gossard, Eds), Vol. 131, p. 109, MRS, Pittsburgh 1989.
16. Tanaka H., Komeno K.: J. Cryst. Growth *93*, 115 (1988).
17. Coltrin M. E., Kee R. J.: *Mater. Res. Soc. Symp. Proc.* (C. W. Tu, V. D. Mattera and A. C. Gossard, Eds), Vol. 145, p. 119. MRS, Pittsburgh 1989.
18. Duchemin J. P., Bonnet M., Koelsch F., Huyghe D.: J. Electrochem. Soc. *126*, 1134 (1979).
19. Leys M. R., Veenvliet H.: J. Cryst. Growth *55*, 145 (1981).
20. Reep D. H., Ghandhi S. K.: J. Electrochem. Soc. *130*, 675 (1983).
21. Kräutle H., Roehle H., Escobosa A., Beneking H.: J. Electron. Mater. *12*, 215 (1983).
22. Heinecke H., Veuhoff E., Pütz N., Heyen M., Balk P.: J. Electron. Mater. *13*, 815 (1984).
23. Takahashi Y., Soga T., Sakai S., Umeno M., Hattori S.: Jpn. J. Appl. Phys. *23*, 709 (1984).
24. Kamon K., Shimazu M., Kimura K., Mihara M., Ishii M.: J. Cryst. Growth *84*, 126 (1987).
25. Schmitz D., Strauch G., Knauf J., Jürgensen H., Heyen M., Wolter K.: J. Cryst. Growth *93*, 312 (1988).
26. Jacko M. G., Price S. J. W.: Can. J. Chem. *41*, 1560 (1963).
27. Nischizawa J., Kurabayashi T.: J. Electrochem. Soc. *130*, 413 (1983).
28. Denbaars S. P., Maa B. Y., Dapkus P. D., Danner A., Lee H. C.: J. Cryst. Growth 77, 188 (1986).
29. Yoshida M., Watanabe H., Uesugi F.: J. Electrochem. Soc. *132*, 677 (1985).
30. Larsen C. A., Buchan N. I., Stringfellow G. B.: Appl. Phys. Letl. *52*, 480 (1980).
31. Larsen C. A., Buchan N. I., Li S. H., Stringfellow G. B.: J. Cryst. Growth *102*, 103 (1990).
32. Haigh J., O'Brien S.: J. Cryst. Growth *67*, 75 (1984).
33. Butler J. E., Bottka N., Sillmon R. S., Gaskill D. K.: J. Cryst. Growth *77*, 163 (1986).
34. Lückerath R., Koss H. J., Tommack P., Richter W., Balk P.: *Mater. Res. Soc. Symp. Proc.* (C. W. Tu, V. D. Mattera and A. C. Gossard, Eds), Vol. 131, p. 91. MRS, Pittsburgh 1989.
35. Bird R. B., Stewart W. E., Lightfoot E. N.: *Přenosové jevy*, p. 579. Academia, Praha 1968.
36. Jacob H., Müller G. in: *Numerical Data and Functional Relationships in Science and Technology; Landolt-Börnstein New Series*, Group III, Vol. 17d (M. Schulz and H. Weiss, Eds), p. 13. Springer, Berlin 1984.
37. Kubíček M.: *Numerické algoritmy řešení chemicko-inženýrských úloh*, Chap. 9. SNTL, Praha 1983.
38. Kleijn C. R., van der Meer T. H., Hoogendoorn C. J.: J. Electrochem. Soc. *136*, 3423(1989).
39. Reid R. C.: Prausnitz J. M., Poling B. E.: *The Properties of Gases and Liquids*, Chap. 11. McGraw-Hill, New York 1987.
40. Holstein W. L.: J. Electrochem. Soc. *135*, 1788 (1988).
41. Erdös E., Voňka P., Stejskal J., Klíma P.: Collect. Czech. Chem. Commun. *49*, 2425 (1984).
42. Suzuki M., Sato M.: J. Electrochem. Soc. *132*, 1684 (1985).

Translated by K. Micka.