be due to the influence on the equilibria of additional compounds mentioned before which were not taken into account in the calculations. However the correspondence is such as to have confidence in the given interpretation. The agreement between the experimental data and the calculated curves also indicates that Henry's law defined as $k = p_P/[P]_{Si}$ is obeyed over the whole range of phosphorus concentrations, which were far below the maximum solubility of P in Si ($\sim 1.5 \times 10^{21}/cm^3$) (11).

Conclusion

The mechanism for the incorporation of phosphorus in epitaxial silicon is governed by the position of the Fermi level in the solid and the chemical equilibria in the gas phase. With increasing phosphine partial pressures a slope bending from 1.0 to 0.5 is found due to transitions in the gas phase, at low temperatures a second break to 0.25 occurs when the donor concentration becomes equal to the intrinsic electron concentration of the silicon matrix.

These results also indicate that the gas phase reactions at or near the silicon surface proceed sufficiently rapidly that equilibrium concentrations of the components may be assumed to be present.

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Vapor Phase Epitaxy and Thermodynamic Calculations of $GaAs_{1-x}P_x$ ($0.3 < x < 0.5$)

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ABSTRACT

Various relationships between gas concentrations and solid composition have been established for the vapor phase epitaxy of $GaAs_{1-x}P_x$ ($0.3 < x < 0.5$). A $GaCl$-AsH_3-PH_3-H_2 process was used to grow such materials at a satisfactory temperature of 790°C. The crystal composition was measured as a function of the input flow rates. Based on these reproducible data, thermochemical calculations were performed with thermodynamic constants evaluated from the literature or determined by fitting the measured data to the calculated equations. The results agree nicely with all the experimental data when the input flow rate ratio $f^o_{HCl}/f^o_{H_2}$ is higher than 0.01. For $f^o_{HCl}/f^o_{H_2} < 0.01$, disagreements occur and the gas-solid relationships are hence established by experimental extrapolations. The crystal composition is strongly affected by $f^o_{HCl}/f^o_{H_2}$ and $f^o_{AsH_3}/f^o_{H_2}$, in addition to $f^o_{PH_3}/f^o_{AsH_3}$. Such phenomena are briefly discussed.

The $GaAs_{1-x}P_x$ materials have been of practical importance for electronic applications. At the present time, commercial light-emitting diodes (LED's) are mainly fabricated with these materials. Red-emitting $GaAs_{1-x}P_x$ ($x \approx 0.4$) diode indicators and displays were successfully developed a few years ago. More recently, following the observations (1-3) of efficient nitrogen trap processes in indirect $GaAs_{1-x}P_x$, yellow and orange LED's with reasonable efficiency have also been made in such materials with $0.9 > x > 0.6$. Besides electroluminescent applications, high speed current limiting (4) and microwave switching (5) have been achieved in $GaAs_{0.7}P_{0.3}$ due to its velocity-field saturation characteristics. Furthermore, Blakeslee (6) has made superlattice structures in $GaAs_{1-x}P_x$ with a period as small as 110Å by vapor growth. Such structures may very well exhibit novel transport and optical properties like a negative conductance (7).

Although $GaAs_{1-x}P_x$ materials are highly desirable and the AsH_3-PH_3-$GaCl$-H_2 vapor phase epitaxy (VPE) technique has been used extensively, detailed and practical relationships between the input gas concentrations and the resulting crystal composition for such a process have not been available in the literature.

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Key words: $GaAs_{1-x}P_x$, vapor phase growth, epitaxial deposition, thermodynamics, computer calculations.

These relationships are necessary to guide experiments in order to achieve a particular composition as desired. It is the objective of the present study to establish such relationships for a crystal composition range of $0.3 < x < 0.5$ at a growth temperature of 790°C. Such a composition range is of particular interest for red LED's.

Ideally, if the growth atmosphere is close to thermodynamic equilibrium with the grown crystal, one can describe the gas-solid interface entirely by theoretical calculations. However, in practice, the growth interface can be far off thermochemical equilibria. Thus theoretical treatments alone cannot produce accurate results. On the other hand it is too time-consuming to establish the entire relationships by experiments. A semi-empirical approach was therefore necessary in order to obtain accurate gas-solid relationships. Such an approach was used in the present study.

Experimental

Procedures.—The reactor system employed in this study is shown in Fig. 1. It is similar to the one used by Tietjen and Amick (8). But, in the present system, the HCl flow can bypass the Ga source for vapor etching of the substrate and is guided to the Ga boat during the growth period to ensure nearly complete conversion of HCl into GaCl. The reactor tube is 5 cm in diameter and 91 cm in heated length. The introduced AsH_3 and HCl are 10% mixtures in H_2, and PH_3 is a 5% mixture in H_2. All the gases are of electronic grade. Ga of 99.99999 purity, loaded in a pyrolytic graphite boat, was used throughout this study. The surface area of the Ga source was around 18 cm^2. Preliminary experiments established that such a source can convert HCl into GaCl almost completely under the present experimental conditions. Chemically polished GaAs of {100} orientation was employed.

As is shown in Fig. 1, the temperatures selected for growth were 900°, 940°, and 790°C for the source, the center zone, and the substrate. Such a profile was found to yield good preliminary growth. At the beginning of a run, this profile was reached with H_2 flowing inside the reactor and the substrate in the cold zone. AsH_3 flow was then established to prevent thermal etching of the GaAs surface and the substrate moved into the deposition zone. Afterward, vapor etch was carried out with a small HCl stream introduced through the upper tubing (see Fig. 1). Three minutes later, HCl was switched into the lower tubing and set at the desired flow rate to initiate the deposition of GaAs. After GaAs was epitaxially grown for 1 hr, the PH_3 flow was started and gradually increased so as to deposit a taper region with x changing from 0 to a certain desired value x_0. And finally a layer of $GaAs_{1-x_0}P_{x_0}$ was grown with the required thickness.

The following ranges of flow rates were used for growth: H_2 (750-1000 ml/min), AsH_3 (5-20 ml/min), HCl (5-25 ml/min), and PH_3 (0.3-4.0 ml/min). Precautions were taken to eliminate deposition on the reactor upstream of the substrate so as to ascertain that all the introduced gases contribute to the growth atmosphere in the substrate region. Such precautions, together with the fact that the input HCl is almost completely converted into GaCl in the present design, allows correlation of the growth atmosphere with the input gas concentration.

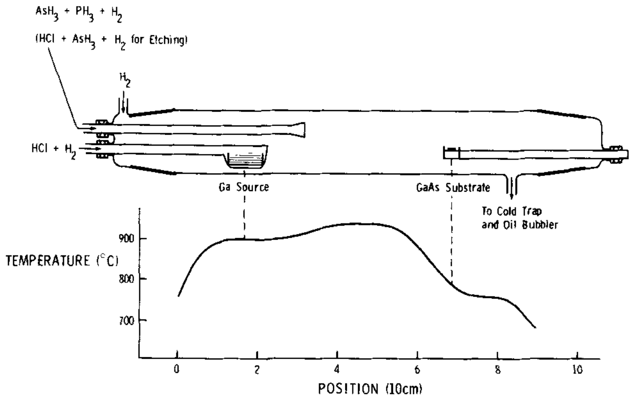

Fig. 1. Schematic diagram of the epitaxial reactor with the temperature profile used in this study.

Results.—Growth morphologies were examined and compared at different flow rate conditions. When the growth atmosphere is nonstoichiometric, pyramids or pits are developed on the as-grown surfaces depending on whether the deposition region is rich in group III element or group V elements. On the other hand, in the stoichiometric case, as-grown surfaces are generally smooth and specularly reflective. Details of such observations were reported previously (9).

Typical layers consisted of 30-50 μm of GaAs, 40-100 μm of graded $GaAs_{1-x}P_x$ and 50-200 μm of $GaAs_{1-x_0}P_{x_0}$. Electron microprobe analysis was carried out across the cross section of every layer to determine the crystal compositions. Very uniform x_0 through the final layer has been observed in all cases. The values of x_0 and their corresponding sets of flow rates at a growth temperature of 790°C are listed in Table I. Such data were checked and found to be reproducible. It was estimated that all the values of x_0 should be accurate within ±0.01 and the various flow rate ratios to be within ±5%. Extreme care was taken to check the reproducibilities and to keep the results within the above accuracies. It should also be pointed out that such correspondence can be used to describe the gas-solid relationships at the growth interface, since, as mentioned above, the input flow rate conditions in the present setup can be related accurately to the growth atmosphere. Part of the data in Table I was therefore adopted in the following calculation to adjust theoretical parameters.

Thermodynamic Calculations

Theoretical model.—The theoretical approach used in this study is formulated with the following assumptions: (*i*) Dynamic thermochemical equilibria are established in a deposition region of uniform temperature and pressure. Such equilibria at the gas-solid interface control the growth of $GaAs_{1-x}P_x$. (*ii*) All the gas species obey the ideal gas law and the GaAs-GaP solid can be described by regular solution behavior. (*iii*)

Table I. Relationships between the input flow rates and the grown crystal composition obtained in the present experiments

$f^o_{HCl}/f^o_{H_2}$ ($\times 10^{-3}$)	$f^o_{AsH_3}/f^o_{H_2}$ ($\times 10^{-3}$)	$f^o_{PH_3}/f^o_{H_2}$ ($\times 10^{-3}$)	$f^o_{PH_3}/f^o_{AsH_3}$ ($\times 10^{-2}$)	x ($\times 10^{-2}$)
5.0	15.0	1.5	10	34
11.0	15.0	1.5	10	36
10.0	20.0	2.0	10	44
15.0	20.0	2.0	10	36
17.5	30.0	3.0	10	40
27.5	40.0	4.0	10	33
9.0	8.0	1.6	20	44
12.5	10.0	2.0	20	42
16.5	12.0	2.4	20	40
20.0	15.0	3.0	20	39
19.0	20.0	4.0	20	50
25.0	20.0	4.0	20	43
33.0	20.0	4.0	20	33
33.0	30.0	6.0	20	46
15.0	5.0	1.5	30	37
17.0	10.0	3.0	30	49
20.0	10.0	3.0	30	43
30.0	10.0	3.0	30	34
30.0	20.0	6.0	30	49
11.0	2.5	1.0	40	38
2.5	5.0	2.0	40	50
7.5	5.0	2.0	40	57
11.0	2.5	1.0	40	40
20.0	4.5	1.8	40	40
30.0	5.0	2.0	40	33
8.0	7.0	2.8	40	62
25.0	7.0	2.8	40	40
30.0	10.0	4.0	40	42
30.0	15.0	6.0	40	51
5.0	15.0	3.75	25	46
6.3	11.0	2.75	25	44
11.0	12.5	3.1	25	59
11.0	11.0	1.65	15	40
13.0	13.0	1.95	15	41
18.0	17.0	2.6	15	36
23.0	22.0	3.3	15	34

HCl is completely converted into GaCl by reaction with Ga in the source region. (*iv*) No deposition occurs upstream of the substrate. (*v*) M_2, M_4, and MH_3 (M = As or P) contribute equally to the deposition of $GaAs_{1-x}P_x$, while various mixed species of the type As_yP_z can be neglected.

Assumptions (*iii*) and (*iv*) were found to be reasonably true in the present case. A mass-spectrometric study by Ban (10) has determined the relative abundances and deposition reactivities of various M-containing species at 680°, 780°, and 880°C. Such results indicate that assumption (*v*) should also be a good approximation. However, on the other hand, assumptions (*i*) and (*ii*) deviate to a good degree from the actual cases. These deviations make it impossible to obtain accurate relationships between the gas concentrations and the solid composition based on a theoretical approach alone. Incorporation of experimental data is therefore necessary. Indeed, it was found here that the equations derived with the above theoretical assumptions could be used to describe the vapor growth of $GaAs_{1-x}P_x$, only if the thermodynamic constants were properly adjusted to fit experimental data. A similar technique has been applied successfully by Wu and Pearson (11) to the liquid phase epitaxy of $In_xGa_{1-x}As$.

Based on the assumptions (*i*-*v*), one can write the following reactions to illustrate the equilibria existing around the gas-solid interface

$$GaCl + \frac{1}{3}\,AsH_3 + \frac{1}{6}\,As_2 + \frac{1}{12}\,As_4 \rightleftarrows GaAs + HCl \qquad [1]$$

$$GaCl + \frac{1}{3}\,PH_3 + \frac{1}{6}\,P_2 + \frac{1}{12}\,P_4 \rightleftarrows GaP + HCl \qquad [2]$$

$$As_4 \rightleftarrows 2\,As_2 \qquad [3]$$

$$P_4 \rightleftarrows 2\,P_2 \qquad [4]$$

$$2\,AsH_3 \rightleftarrows As_2 + 3\,H_2 \qquad [5]$$

$$2\,PH_3 \rightleftarrows P_2 + 3\,H_2 \qquad [6]$$

The equilibrium constant K_j of reaction (j) can be expressed as follows

$$K_1 = \frac{(1-x)\,\gamma_{GaAs}\,p_{HCl}}{p_{GaCl}\,(p_{AsH_3})^{1/3}\,(p_{As_2})^{1/6}\,(p_{As_4})^{1/12}} \qquad [7]$$

$$K_2 = \frac{x\,\gamma_{GaP}\,p_{HCl}}{p_{GaCl}\,(p_{PH_3})^{1/3}\,(p_{P_2})^{1/6}\,(p_{P_4})^{1/12}} \qquad [8]$$

$$K_3 = \frac{(p_{As_2})^2}{p_{As_4}} \qquad [9]$$

$$K_4 = \frac{(p_{P_2})^2}{p_{P_4}} \qquad [10]$$

$$K_5 = \frac{p_{As_2}\,(p_{H_2})^3}{(p_{AsH_3})^2} \qquad [11]$$

$$K_6 = \frac{p_{P_2}\,(p_{H_2})^3}{(p_{PH_3})^2} \qquad [12]$$

where p and γ designate the partial pressure and the activity coefficient, respectively. Since we are dealing with an open-flow system with H_2 as the major gas specie and the GaAs-GaP solid is assumed to be regular, we have

$$p_{H_2} \approx 1 \text{ atm} \qquad [13]$$

and

$$\gamma_{GaAs} = \exp\left[\frac{\alpha x^2}{RT}\right] \qquad [14]$$

$$\gamma_{GaP} = \exp\left[\frac{\alpha(1-x)^2}{RT}\right] \qquad [15]$$

where α is the interaction parameter between GaAs and GaP.

Thus we have established relationships between the solid composition and the partial pressures of various gas species in the deposition region. These partial pressures can be further related to the input flow rates through consideration of mass conservation due to the validity of assumptions (*iii*) and (*iv*). The conservation of chlorine, arsenic, and phosphorus can be described, respectively, as

$$p_{HCl} + p_{GaCl} = p^o_{HCl} \approx \frac{p^o_{HCl}}{p^o_{H_2}} = \frac{f^o_{HCl}}{f^o_{H_2}} \qquad [16]$$

$$p_{AsH_3} + 2p_{As_2} + 4p_{As_4} + (1-x)\,p_{HCl} = p^o_{AsH_3} \approx \frac{f^o_{AsH_3}}{f^o_{H_2}} \qquad [17]$$

and

$$p_{PH_3} + 2p_{P_2} + 4p_{P_4} + x\,p_{HCl} = p^o_{PH_3} \approx \frac{f^o_{PH_3}}{f^o_{H_2}} \qquad [18]$$

where f^o is the flow rate of the incoming gas and p^o represents the partial pressure of this input gas in the growth region prior to deposition.

With Eq. [7]-[18], one can solve for the crystal composition and the partial pressures at any given incoming gas concentrations, once all the equilibrium constants and the interaction parameter are determined.

Determination of constants.—The above approach can be used for any growth temperature. But the present calculation was performed with 790°C, which was found to be satisfactory for $0.3 < x < 0.5$.

The equilibrium constants, *i.e.*, K_1, K_2, K_3, K_4, K_5, and K_6 were evaluated from Kirwan's results (12) to be 10^4, 80, 10^{-5}, 9×10^{-5}, 3×10^{17}, and 3×10^2 atm units, respectively, at 790°C. Panish and Ilegems (13) have reported a value of 0.4 kcal/mole for α. Based on these parameters, preliminary calculations resulted in significant deviations from the experimental data obtained in this study. This can be expected due to the fact that complete thermochemical equilibria do not exist and kinetic effects such as adsorption, surface reaction, and desorption cannot be neglected in actual cases, as were reported by Shaw (14, 15). In order to obtain accurate results with the above-described equations, one thus has to adjust appropriate parameters to fit experimental data. Such adjustments will invalidate some of the above assumptions and thus result in inconsistency in the theoretical model. However, the main purpose of this paper is to establish correct gas-solid relationships for practical use. The adoption of simple, unrealistic assumptions was merely to set up calculation equations to start with.

Since we are mainly interested in calculation of the crystal composition at given gas concentrations, such adjustments should be made with parameters which have major influences on the depositions of GaAs and GaP. Adjustable parameters should therefore be selected among K_1, K_2, and α, which can affect the deposition reactions directly.

By following the same techniques used in Ref. (11), it was found that, with $K_2 = 3 \times 10^2$ atm units, $\alpha = (4.2\text{-}3.8\,x)$ kcal/mole, and other equilibrium constants given above, the calculation resulted in good agreement with the experimental data.

Calculation results.—Such a calculation was carried out to determine the crystal composition x at any given $f^o_{HCl}/f^o_{H_2}$, $f^o_{AsH_3}/f^o_{H_2}$, and $f^o_{PH_3}/f^o_{AsH_3}$. The results are shown in Fig. 2 and 3 together with the experimental data obtained in the study. The solid lines are calculated results and the dashed lines are experimental extrapolations. Good agreement was obtained for $f^o_{HCl}/f^o_{H_2} > 0.01$. However, when $f^o_{HCl}/f^o_{H_2}$ is smaller than 0.01, the calculation fails and the relationships were established entirely by experiments. The failure of the calculation in this region indicates that such a semi-empirical approach can only be applied in limited regions since the actual deposition process can be far more complicated.

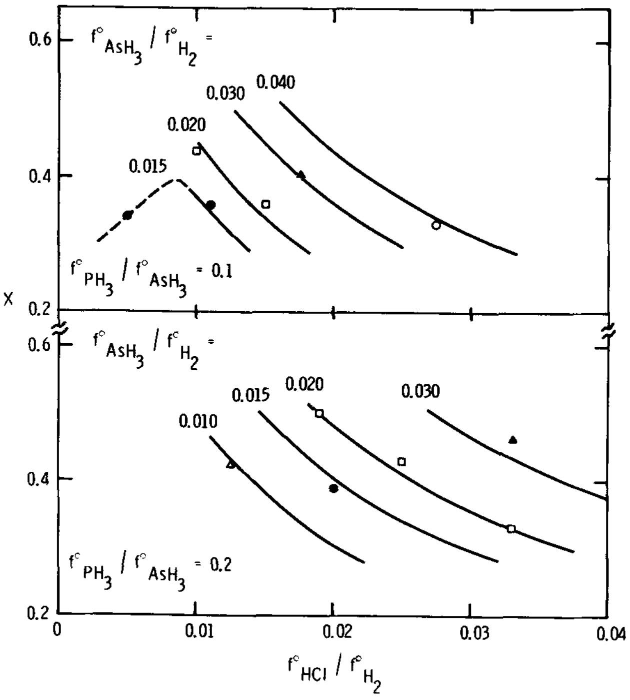

Fig. 2. Crystal composition x as a function of input flow rate ratios $f^o_{HCl}/f^o_{H_2}$, $f^o_{AsH_3}/f^o_{H_2}$, and $f^o_{PH_3}/f^o_{AsH_3}$ (= 0.1 and 0.2) at a growth temperature of 790°C. Solid lines are calculated results. The dashed line is experimental extrapolation. The values of $f^o_{AsH_3}/f^o_{H_2}$ for various measured data are: 0.010 (△), 0.015 (●), 0.020 (□), 0.030 (▲), and 0.040 (○).

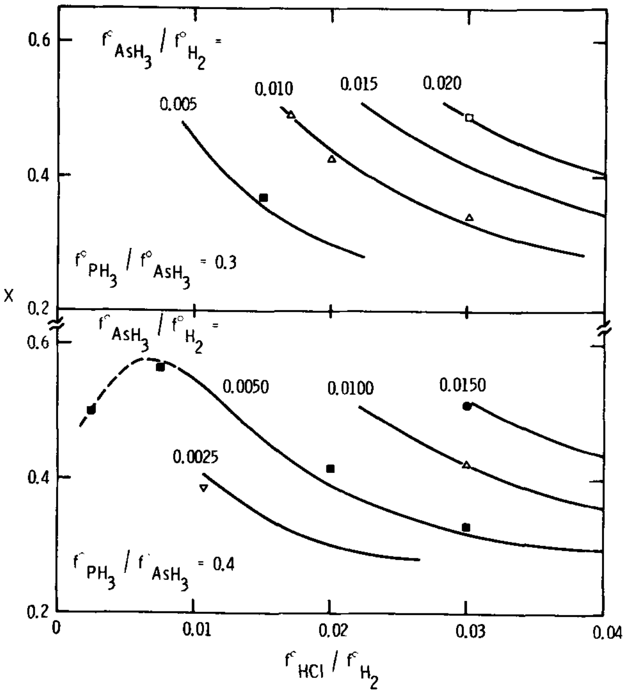

Fig. 3. Crystal composition x as a function of input flow rate ratios $f^o_{HCl}/f^o_{H_2}$, $f^o_{AsH_3}/f^o_{H_2}$, and $f^o_{PH_3}/f^o_{AsH_3}$ (= 0.3 and 0.4) at a growth temperature of 790°C. Solid lines are calculated results. Dashed line is experimental extrapolation. The values of $f^o_{AsH_3}/f^o_{H_2}$ for various measured data are: 0.0025 (▽), 0.0050 (■), 0.0100 (△), 0.0150 (●), and 0.020 (□).

Figure 4 gives the relationships among the incoming flow rates required to grow $GaAs_{0.6}P_{0.4}$ under the present experimental conditions. All the lines are drawn from Fig. 2 and 3, together with the corresponding experimental data in Table I. Similar plots can be obtained for other x and serve as guides to achieve specific compositions.

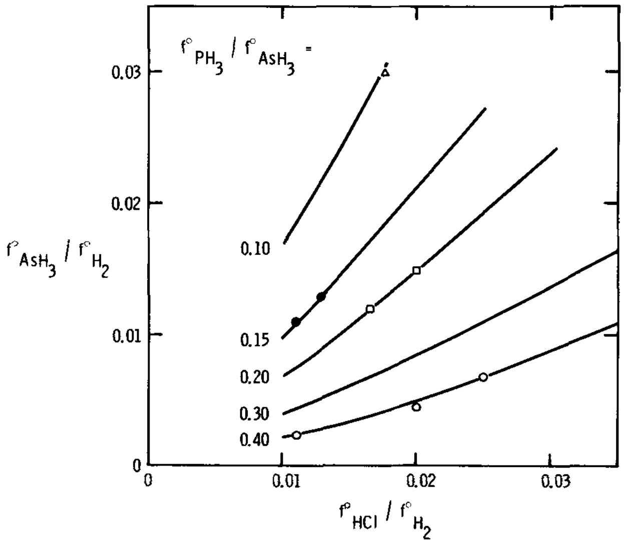

Fig. 4. Input flow rate ratio $f^o_{AsH_3}/f^o_{H_2}$ required to achieve x = 0.4 ± 0.02 with various fixed $f^o_{HCl}/f^o_{H_2}$ and $f^o_{PH_3}/f^o_{AsH_3}$ at 790°C. All the lines are calculated results. The values of $f^o_{PH_3}/f^o_{AsH_3}$ for various measured data are: 0.10 (△), 0.15 (●), 0.20 (□), and 0.40 (○).

Discussion

From the above results, the crystal composition is not only determined by $f^o_{PH_3}/f^o_{AsH_3}$, but is also affected to a great extent by $f^o_{HCl}/f^o_{H_2}$ and $f^o_{AsH_3}/f^o_{H_2}$. In particular, at constant $f^o_{AsH_3}/f^o_{H_2}$ and $f^o_{PH_3}/f^o_{H_2}$, x increases along with $f^o_{HCl}/f^o_{H_2}$ when $f^o_{HCl}/f^o_{H_2} < 0.01$ but decreases as $f^o_{HCl}/f^o_{H_2}$ is increased further. It is difficult to explain such behavior in much detail since precise thermochemistry at a real growth interface is not known. Nevertheless the following interpretations can be made here.

The crystal composition is influenced by two factors: (*i*) the reactant partial pressures and (*ii*) the reactivities of various M-containing species with GaCl. When f^o_{HCl} is much greater than $f^o_{AsH_3} + f^o_{PH_3}$, there is more than sufficient GaCl in the deposition region. In that case, the reactivity influence is overruled by the pressure influence and the crystal composition x is mainly determined by $f^o_{PH_3}/(f^o_{PH_3} + f^o_{AsH_3})$. As $f^o_{HCl}/f^o_{H_2}$ is reduced while keeping $f^o_{PH_3}/f^o_{AsH_3}$ and $f^o_{AsH_3}/f^o_{H_2}$ constant, the relative reactivities of M-containing species will then become increasingly important. The fact that x increases with decreasing $f^o_{HCl}/f^o_{H_2}$ (>0.01) can hence be explained by the over-all higher reactivity of P-containing reactants in this range. The same explanation also holds for the increasing x when $f^o_{AsH_3}/f^o_{H_2}$ is increased with constant $f^o_{PH_3}/f^o_{AsH_3}$ and $f^o_{HCl}/f^o_{H_2}$.

Sedgwick (16) proposed a "quasi-equilibrium" model to describe the H_2 reduction of $SiCl_4$. In this model, only a fraction β of the incoming gases equilibrates with the condensed phase. Shaw (14) has found such a model can be successfully applied to calculate GaAs deposition rates for certain limited growth conditions if a constant, empirical β is assumed. The present results agree with such previous observations. The adjustment of K_2, in the present case, is equivalent to the quasi-equilibrium treatment. It was also found here that such a treatment can be successfully used to calculate x for certain growth conditions, namely, $f^o_{HCl}/f^o_{H_2} > 0.01$ in the present case.

Although the use of a composition-dependent α is inconsistent with the assumption of regular solution behavior between GaAs and GaP, it yields some accurate and practical results in the present calculations. After all, the GaAs-GaP solid solution is by no means strictly regular. Rao and Tiller (17) have found composition-dependent α's can be used to describe similar systems.

It should also be pointed out here that the empirical adjustments of K_2 and α were performed based on part of the reproducible data in Table I only. But the remaining data were also found to agree with the calculated results nicely. This suggests such a semiempirical approach is a good technique for interpolation and reasonable extrapolation with a limited amount of data.

In a previous paper (9), an optimum growth condition was reported for the process used in this study, namely, $f^o_{HCl}/f^o_{H_2} = (f^o_{AsH_3} + f^o_{PH_3})/f^o_{H_2} = 0.02$. Such a condition together with the relationships given in Fig. 4 indicate that $GaAs_{0.6}P_{0.4}$ can be best grown with $f^o_{HCl}/f^o_{H_2} = 0.02$, $f^o_{AsH_3}/f^o_{H_2} = 0.0165$, and $f^o_{PH_3}/f^o_{AsH_3} = 0.19$ at a temperature of 790°C.

Although the present relationships are established for the AsH_3 and PH_3 process, they can be applied equally well to the $AsCl_3$ and PCl_3 process since the thermochemistry at the gas-solid interface is the same for both processes. As a matter of fact, the trichloride process can be considered as a special case of the hydride process, which satisfies $p^o_{HCl} = 3(p^o_{AsH_3} + p^o_{PH_3})$ with $p^o_{AsH_3}$ and $p^o_{PH_3}$ equal to $p^o_{AsCl_3}$ and $p^o_{PCl_3}$, respectively. Thus, the present relationships can be reduced for use in the trichloride case by introducing a restriction that $f^o_{HCl}/f^o_{H_2}$ is equal to $3(f^o_{AsH_3} + f^o_{PH_3})/f^o_{H_2}$. It was indeed found that the results so obtained agree well with the calculated relationships by Bleicher (18) for the trichloride process.

Conclusions

A thermochemical investigation on the VPE of $GaAs_{1-x}P_x$ $(0.3 < x < 0.5)$ has been carried out and the following conclusions can be made:

1. The crystal composition x $(0.3 < x < 0.5)$ as a function of the input flow rate ratios $f^o_{PH_3}/f^o_{AsH_3}$, $f^o_{AsH_3}/f^o_{H_2}$, and $f^o_{HCl}/f^o_{H_2}$ at 790°C has been obtained for the hydride process. A semiempirical approach was used to establish such relationships. Such an approach demolishes the theoretical consistency, but is certainly a simple way to establish accurate, practical relationships. Similar techniques can be used for other ranges of x or other materials.

2. The flow rate dependences of x for $f^o_{HCl}/f^o_{H_2} > 0.01$ can be explained by the relative abundances and reactivities of various vapor sources.

3. It is concluded that $GaAs_{0.6}P_{0.4}$ vapor epitaxial layers can be best grown with $f^o_{HCl}/f^o_{H_2} = 0.02$, $f^o_{AsH_3}/f^o_{H_2} = 0.0165$, and $f^o_{PH_3}/f^o_{AsH_3} = 0.19$ at 790°C. Such information is particularly important for red-emitting diode application.

4. The above relationships can be reduced and used for the trichloride process which is actually a special case of the hydride process.

5. The results established here are applicable to any such reactors where the input flow rates can be correlated with the incoming gas concentrations in the substrate region.

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