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Numerical simulations of epitaxial growth in MOVPE reactor as a tool for aluminum nitride growth optimization

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Abstract

The present study concerns numerical simulations and experimental measurements on the influence of inlet gas mass flow rate on the growth rate of aluminum nitride crystals in Metalorganic Vapor Phase Epitaxy reactor model AIX-200/4RF-S. The aim of this study was to design the optimal process conditions for obtaining the most homogeneous product. Since there are many agents influencing reactions relating to crystal growth such as temperature, pressure, gas composition and reactor geometry, it is difficult to design an optimal process. Variations of process pressure and hydrogen mass flow rates have been considered. Since it is impossible to experimentally determine the exact distribution of heat and mass transfer inside the reactor during crystal growth, detailed 3D modeling has been used to gain insight into the process conditions. Numerical simulations increase the understanding of the epitaxial process by calculating heat and mass transfer distribution during the growth of aluminum nitride crystals. Including chemical reactions in the numerical model enables the growth rate of the substrate to be calculated. The present approach has been applied to optimize homogeneity of AlN film thickness and its growth rate.

Keywords: metalorganic vapor phase epitaxy, Finite Volume Method, semiconductors, aluminum nitride

1. Introduction

Semiconductors based on silicon carbide are commonly used in electronic applications. Silicon carbide-based nitride semiconductors are used in applications such as High Electron Mobility Transistors (HEMT), Insulated Gate Bipolar Transistors (IGBT), Metal and Metal-Oxide Semiconductor Field Effect Transistors (MESFET, MOSFET), high power blue diodes, Light Emitting Diodes and Laser Diodes [1]. LEDs have recently attracted more attention as a potential alternative for incandescent bulbs and fluorescent lamps. Epitaxial growth is a widely used method for manufacturing such semiconductors, because the epitaxial layer can be created as a single crystal, without grain boundaries and other defects, so it produces crystal of the highest quality [2].

Since there are many agents influencing reactions relating to crystal growth such as temperature, pressure and volumetric flow rate of inlet gases, it is difficult to design an optimal process. Experimental analysis of the process conditions is very limited, mainly due to high temperature and the flow disturbance caused by introducing measuring probes [3]. Therefore it is impossible to determine experimentally the exact distribution of heat and mass transfer inside the reactor during the process. Numerical simulations lend insight to the process by calculating heat and mass transfer distribution during the growth process of silicon carbide, gallium nitride and aluminum nitride crystals [4–6]. Including the gas-phase and surface chemical reactions occurring inside the reactor chamber in the numerical model enables the growth rate distribution of the crystal on the substrate tobe determined. This model can be used to study the influence of reagents volumetric flow rate and process pressure on the thickness homogeneity of the obtained crystal [7–9].

Commercial reactors are commonly used in production lines and laboratories worldwide and have become a common tool for the design of new materials and devices. As a result, the development of predictive models of commercial systems have attracted much research in recent years. Yakovlev et al. [8] studied deposit behavior of gallium nitride in the AIX-200/4RF-S reactor. Pawlowski et al. [10] performed modeling analysis of GaN MOVPE in the Thomas Swan close-coupled shower head 3 x 2 inch reactor. Optimization of the growth rate uniformity for nitrogen and hydrogen carrier gases was performed in [11] for the growth of AlGaAs in the AIX 200 horizontal single wafer reactor.

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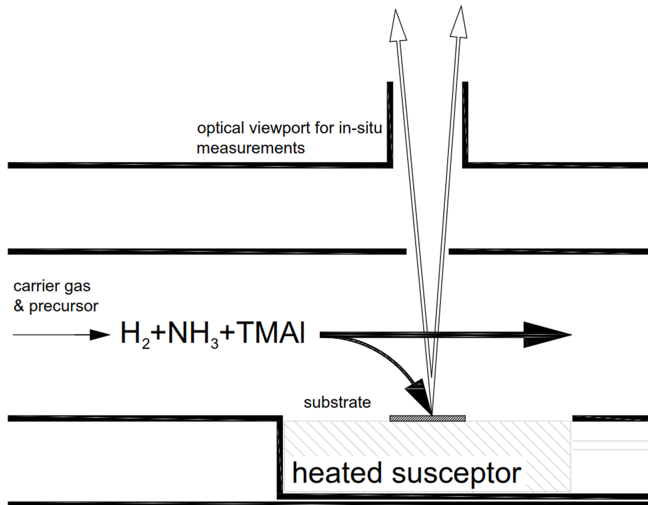

Figure 1: Schematic of the experimental procedure

Two different gas inlet configurations were considered in [12] to analyze the homogeneity of AlAs/GaAs structures in the AIX 200 reactor. The horizontal MOVPE reactor SR6000 numerical model was developed and studied in [13] together with planetary and vertical reactors. Modeling results of GaN MOVPE in the AIX 200/4 HT reactor with separated supply of group-III and group-V precursors were reported in [14] for two configurations of the separation plate, and the computations matched well the character of the growth rate distribution along the flow direction.

In this work a modeling and experimental study of aluminum nitride crystal growth in Low Pressure Metal Organic Vapor Phase Epitaxy reactor model AIX 200/4 RF-S was performed as a follow-up to previous research [15]. The effect of process pressure and hydrogen volumetric flow rate on the GaN growth rate were studied and a comparison of numerical and experimental results was performed.

2. Experimental procedure

This study concerns the influence of pressure inside the reactor chamber on the process of AlN crystal growth. Aluminum nitride epitaxial layers were grown on a silicon carbide substrate in an AIX 200/4 RF-S low-pressure metalorganic vapor phase epitaxy reactor (LP MOVPE). A schematic diagram of the process is shown in Fig. 1.

The inlet gases used for creating the aluminum nitride layer were: trimethylaluminium (TMAl, 20 sccm), ammonia (NH_3, 1200 sccm) and hydrogen (H_2, 9900sccm). 4H-SiC substrates were used for epitaxial processes. A quartz plate separated the inlet of gases, resulting in TMAl with H_2 being introduced through the upper inlet and NH_3 with H_2 carrier gases through the lower inlet. The temperature of the experimental process was 1023 K. Experimental data regarding the influence of process pressure on the crystal growth was used to verify the modeling results.

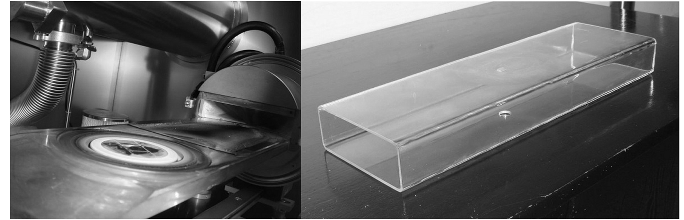

Figure 2: MOVPE reactor chamber

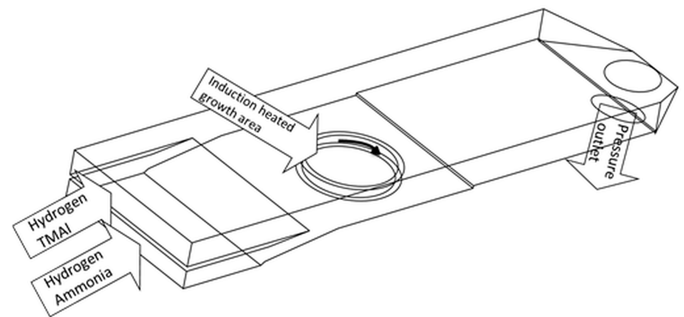

Figure 3: Schematic model of MOVPE reactor

3. Numerical model

To study the influence of process parameters on crystal growth, a model of the MOVPE reactor AIX-200/4RF-S was created. The MOVPE reactor and its model are shown in Figures 2 and 3.

The disk on which the silicon carbide susceptor was placed is made of graphite and was induction heated during the process. Experimental measurements show that the graphite disk has constant temperature during the process, however the temperature on the surface of the plate is significantly lower than the heating temperature. The temperature difference is caused by a thin gap below the disk filled with hydrogen, which provides the rotation of the susceptor disk. The temperature on the surface of the modeled susceptor has been adjusted to fit experimental measurements at the reference point for each process. Such fitting makes it possible to neglect the details of induction heating and saves computational time. The other walls of the reactor are made of quartz. The inlet gases were introduced to the reactor at room temperature through two inlets separated by a quartz plate, and mixing took place just before the crystal growth area.

Mesh was created to represent the geometry of the reactor. The mesh is refined over the chemical reactions area, since it is the area of highest importance, and mesh should be the most accurate here.

Table 1: Process parameters for model verification

Pressure, mbar	Temperature, K	Trimethylaluminum volumetric flow rate, sccm	Hydrogen volumetric flow rate , sccm	Ammonia volumetric flow rate, sccm
50	1023	20	9900	1200

Computational fluid dynamics simulations taking into account gas-phase chemical reactions and surface chemistry were carried out using ANSYS Fluent software. The governing equations were three dimensional Navier-Stokes equations with vapor-phase chemical reactions. Radiative heat transfer was described by the Discrete Ordinates model and took into account wavelength-dependent optical properties of quartz (non-gray radiation).

The ANSYS Fluent software used for calculations solved a set of differential equations describing mass and heat transfer. The governing equations in Computational Fluid Dynamics problems are mass, momentum and energy conservation equations. The energy equation takes the following form (Eq. 1).

$$\frac{\sigma}{\sigma t}(\rho E) + \nabla \cdot (\vec{v}(\rho E + p)) = \nabla \cdot \left(k_{eff} \nabla T - \sum_j h_j \vec{J}_j + \left(\bar{\bar{\tau}}_{eff} \cdot \vec{v} \right) + S_h \right) \quad (1)$$

where: energy E and temperature T are mass-averaged variables, k_{eff} is the effective thermal conductivity, J_j is the diffusion flux of species j, h is enthalpy, v is fluid velocity, ρ and p are density and pressure respectively. The first three terms on the right-hand side of the equation represent energy transfer due to conduction, species diffusion, and viscous dissipation, respectively. The source term S_h contains contributions from radiation and reactions, which is crucial in the MOVPE process, where both radiative heat transfer and reactions as a heat source have to be taken into account. The radiative heat transfer is calculated as in Equation 2.

$$\frac{dI(\vec{r}, \vec{s})}{ds} + (a + \sigma_s) I(\vec{r}, \vec{s}) = an^2 \frac{\sigma T^4}{\Pi} + \frac{\sigma_s}{4\Pi} \int_0^{4\Pi} I\left(\vec{r}, \vec{s}'\right) \Phi\left(\vec{s} \cdot \vec{s}'\right) d\Omega' \quad (2)$$

where: $\vec{r}$ is the position vector, $\vec{s}$ is the direction vector, $\vec{s}'$ is scattering direction, s is the path length, a is absorption coefficient, n is refractive index, σ_s is scattering coefficient, σ is Stefan-Boltzmann constant ($5.672 \times 10^{-8} W/m^2K^4$), I represents radiation intensity, which depends on position and direction vectors, T is the local temperature, Φ and Π are phase function and solid angle, respectively.

The mixture of gases in the numerical model was simulated as an ideal gas. The density for such case is described by the equation of state (Eq. 3).

$$\rho = \frac{pM}{RT} \quad (3)$$

where: M is molecular weight of the gaseous phase, R is universal gas constant, and T is temperature.

The flow in the reactor is considered as compressible, so the pressure in the Clapeyron equation is calculated as a sum of the local relative pressure predicted by Fluent and the operating pressure. The main heat transfer mechanisms for the epitaxial growth process are convection and radiation. Outer walls thermal boundary conditions were set as mixed and reflect the fact that the reactor is constantly cooled during the process with hydrogen [16] at room temperature. Mathematical and numerical representation of the chemical reactions in gas-phase chemical and on the plate surface was included in ANSYS Fluent using dedicated modeling software package CVDSim Nitride Edition from STR GMBH.

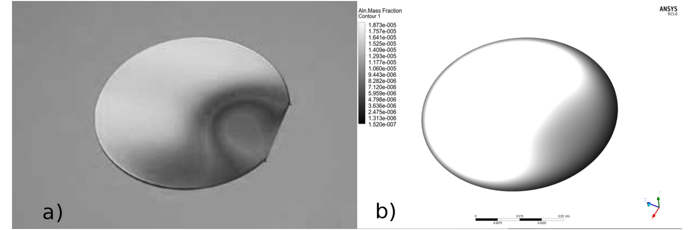

Figure 4: Comparison of experimental and numerical results: a) thickness distribution of obtained crystal b) calculated aluminum nitride mass fraction distribution

The 3D model developed here took into account precursor mixing, transport and decomposition. The gas-phase chemistry model includes the unimolecular gas-phase decomposition of TMAl. Deposition of trimethylaluminum which occurs on the reactor walls was described by the surface chemistry model that covers deposition conditions such as low temperature kinetically limited growth, mass transport limited growth at moderate temperatures and growth limited by aluminum desorption at elevated temperatures.

4. Results and discussion

In the first step, experimental verification of the validity of the developed model was performed. The process parameters of the experimental setup for the verification purpose are listed in Table 1.

The thickness distribution of the obtained aluminum nitride crystal was examined and compared with AlN mass fraction distribution calculated from numerical simulations. The comparison is presented in Fig. 4. Thickness distribution is presented as a photograph of the aluminum nitride crystal. Brighter color on the picture designates higher crystal thickness. Modeling results exhibit good agreement with the experimental data.

To investigate the influence of hydrogen volumetric flow rate on the growth rate of aluminum nitride a series of simulations with hydrogen volumetric flow rates of 1100, 2000, 4800 and 7700 standard cubic centimeters per minute was performed. Process parameters are described in Table 2.

Heat and mass transfer and crystal growth rate in the aluminum nitride MOVPE process were calculated. Distribution of aluminum nitride mass fraction on the crystal growth area is shown in Fig. 5 and the calculated AlN crystal growth rate is shown in Fig. 6.

The growth rate of aluminum nitride crystal is the highest for the hydrogen volumetric flow rate of 2000 sccm. However,

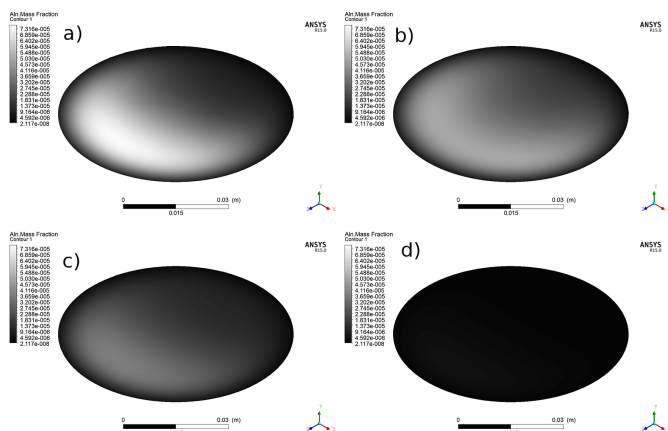

Figure 5: Calculated AlN mass fraction distribution for hydrogen volumetric flow rates of: a) 7700 sccm, b) 4800 sccm, c) 2000 sccm, d) 1100 sccm

Table 2: Process parameters for surface deposition rate calculations

Pressure, mbar	Temperature, K	Trimethylaluminum volumetric flow rate, sccm	Hydrogen volumetric flow rate , sccm	Ammonia volumetric flow rate, sccm
50	1023	20	1100	1200
50	1023	20	2000	1200
50	1023	20	4800	1200
50	1023	20	7700	1200

for this flow rate we obtained the least homogenous product, the least promising for the obtained structures. The aim of this study was to optimize crystal thickness distribution, hence the main conclusion from the simulations performed is that we can obtain better thickness distribution by increasing the hydrogen volumetric flow rate. To obtain similar layer thickness as for higher growth rates, it suffices to extend the time of the growth process.

5. Conclusions

Heat and mass transfer distribution and crystal growth rate in the gallium nitride MOVPE process were studied. The developed numerical model was verified using experimental data from AIX 200/4 RF-S. The aluminum nitride growth rate for different hydrogen volumetric flow rates was studied to obtain the most homogeneous monocrystalline layer. Based on the modeling results, optimal process conditions were chosen. Determining a valid range of temperature and mass fraction of gases during epitaxial crystal growth made it possible to optimize process parameters and obtain crystals of the best quality.

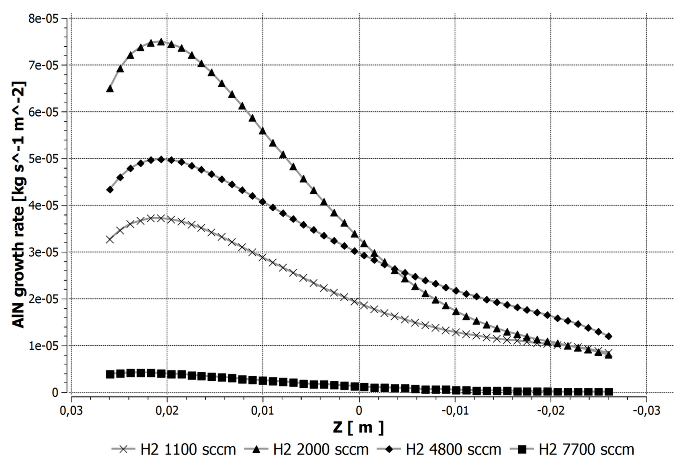

Figure 6: Calculated AlN crystal growth rate for studied set of H_2 volumetric flow rates

Acknowledgments

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