Growth of GaN, AlGaN and AlN Layers for LED Manufacturing: Investigations on Growth Conditions using a "Hotwall" MOCVD System

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Abstract

We report on a new planetary hotwall research system independently from each other heating the ceiling and the susceptor. Therefore, the influence of an actively controlled ceiling during an MOCVD process can be investigated. At maintained constant susceptor surface temperature, it was found that the temperature profile across the substrate becomes more uniform by increasing the ceiling temperature. Furthermore, the deposition profiles for GaN-related materials and AlN demonstrate a higher maximum where the gas flow hits the wafer area as well as the coating on the ceiling changes from a solid to a flaky deposition when raising the ceiling temperature above the susceptor temperature.

INTRODUCTION

The deposition of GaN in an MOCVD (Metal-Organic Chemical Vapour Phase Deposition) system is based on the injection of Trimethylgallium (TMGa), trimethylaluminium (TMAl) and ammonia (NH_3) into the reaction chamber. After dissociation of the source materials in the gas phase, chemical reactions take place leading to a crystalline layer being formed on a substrate, mainly sapphire (Al_2O_3) or, to an increasing extent, silicon. Besides other factors, the density of nitrogen vacancies is a critical parameter in GaN material due to insufficient supply of atomic nitrogen at the growth surface. The low cracking probability of NH_3 [1] at typical temperatures accounts for the observed nitrogen deficiency [2-3]. In a typical planetary MOCVD system, the upper wall (in the following referred to as "ceiling") is passively heated by radiation and heat transport from the actively heated susceptor. As a consequence of such a setup, the gas phase is significantly colder than the surface temperature of the substrate. Another critical aspect for growth of GaN on sapphire or silicon as substrate is related to the different thermal expansion coefficients, leading to a wafer bow due to the compressive or tensile stress of the grown layer which increases by thickness. As a result, the contact of the wafer to the heated susceptor will be non-uniform causing an unwanted temperature profile across the wafer area. This will further be pronounced for layers grown at higher temperature like aluminium nitride (AlN). By reducing or even inverting the vertical thermal gradient between growth surface and ceiling, the impact of wafer bow on its temperature profile is reduced. In this study, we will compare experimental results obtained in a hotwall system with simulation.

SYSTEM SETUP

For this work, a modified AIXTRON multiple wafer planetary hotwall research system was used (see cross section in Figure 1). This system was equipped with a specially coated graphite susceptor in a 6 times 4" configuration, inductively heated by an RF coil. The ceiling, also made of coated graphite, was independently heated by a second RF coil which allowed control of the ceiling temperature. Both coils were powered by 80 kW generators enabling high-temperature operation up to 1650°C for susceptor and ceiling. This setup enables to apply the same surface temperature during growth while varying the ceiling temperature independently. As a reference figure, the ceiling to susceptor power ratio is introduced, indicating the main heat source in the reactor. The reactor was set up with an in-situ pyrometer to measure the surface temperature and monitor the growth process by reflectrometry at 950 nm and 633 nm wavelengths, respectively through the same view port hole in the ceiling. The setpoints for susceptor and ceiling temperature were measured at a radius of r = 110 mm from the backsides and the in-situ surface temperature at r = 123 mm from the top.

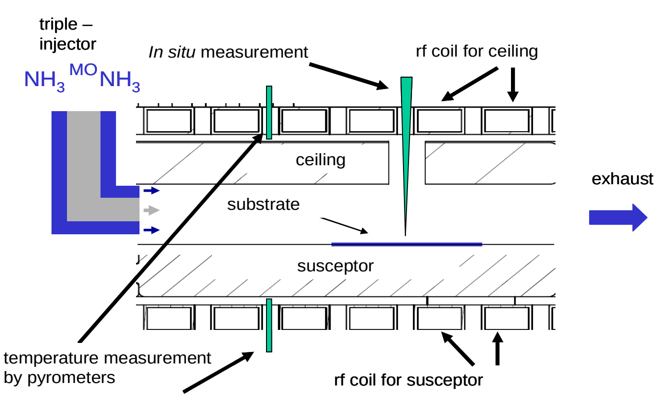

Figure 1: Cross section of the planetary research hotwall system

The reactor featured a water-cooled three-fold injection nozzle (the triple "injector"), by which the metal-organic (MO) precursors were introduced into the reaction chamber via the center inlet, while ammonia feeding in through the top and bottom zone. It was possible to mix H_2/ N_2 carrier gases in any ratios for all injector zones to influence the gas velocities and momentums.

EXPERIMENTAL

All growth experiments for GaN were carried out at a reactor pressure of 400 mbar. For all samples, a thin GaN nucleation layer at 540°C surface temperature was grown followed by an annealing step and growth of 1 µm GaN at 1050°C surface temperature and a ceiling temperature setpoint of 830°C. On top of this first layer, 3 µm of GaN were grown with different ceiling temperatures between 830°C and 1100°C without growth interruption and at the same constant V/III ratio of 1500. X-ray diffraction (XRD) was carried out with a Phillips X'Pert PRO four-circle diffractometer. XRD full width at half maximum (FWHM) values for the (0002) and (10-12) reflections were measured with an open-detector configuration. For the depletion profile measurements, at first, sapphire wafers were used to grow a thick epitaxial GaN layer. The thickness profile of this pre-deposited layer was mapped by a Nanometrics white light interferometer with a lateral resolution of 1 mm. Second, these templates were overgrown on a stalled disc and measured after growth again. By subtracting the thickness profile of the templates, the growth rate distribution (depletion profile) in the reactor as a function of the reactor radius was obtained. Sheet carrier concentration and electron mobility were measured by Hall measurement using a van-der-Pauw geometry. Sheet carrier concentration was additionally measured by a WEP ECV profiler.
Similar experiments were carried out for deposition of AlN to measure the thickness profile while those for AlGaN were used for investigations on the uniformity of Al content for the ternary compound using the hotwall set-up.

RESULTS AND DISCUSSION

Increasing the ceiling temperature from 830°C up to 1100°C while reducing the susceptor setpoint temperature from 1050°C to 916°C at the same time in a manner keeping the wafer surface temperature constant at 1050°C, resulted in a change for the power ratio "ceiling to susceptor" from 0.12 to 4.6 (Figure 2). For the maximum value, almost all power is transferred into the ceiling and the substrate is mainly heated from above. Evaluation of the growth experiments as described above provided XRD FWHM values of 210 arcsec for the (0002) reflex and 360 arcsec for the (10-12). These FWHM values did not change with increasing ceiling temperature with the assumption that this may be related to the same initial growth conditions. AFM measurements show a step-flow growth mode and a low root mean square roughness (RMS) between 0.4 nm and 0.5 nm for 5x5 µm² scans for all samples, indicating that the same surface temperature was obtained for all samples. It was found that the parasitic ceiling coating changes when increasing the ceiling temperature setpoint from 1000°C to 1050°C. The coating altered from a solid to a more loose coating and became snowflake-like when the ceiling temperature was further raised to 1100°C, causing particles falling off during growth or when opening the reactor. Therefore, the ceiling temperature was limited for the following experiments to a maximum of 1000°C. Two temperature settings of $T_{Susceptor}/T_{Ceiling}$ at 1050°C/830°C and 995°C/1000°C were simulated and compared to experimental data.

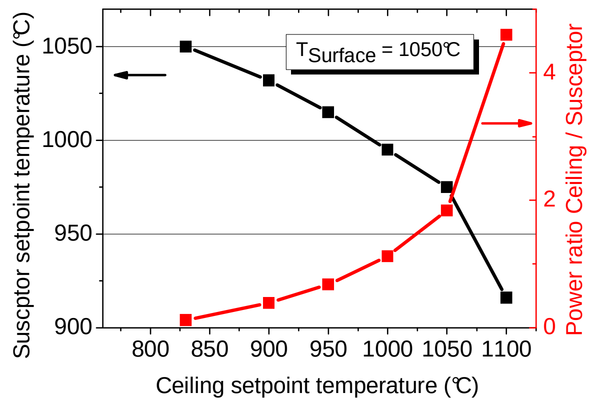

Figure 2: Temperature settings and power ratio for ceiling temperatures between 830°C and 1100°C as well as a constant surface temperature of 1050°C

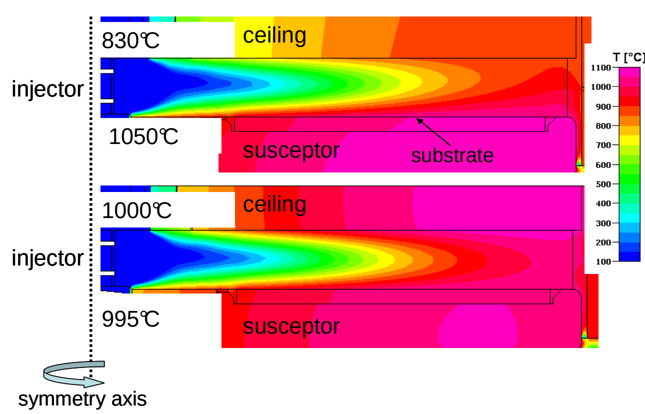

Figure 3 Thermal simulation for a susceptor and ceiling temperature of 1050°C/830°C (picture at the top) and 995°C/1000°C (picture at the bottom)

Figure 3 shows the thermal simulation for a ceiling temperature setpoint of 830°C (top) and 1000°C (bottom) and typical flow conditions. As one can see the "cold finger" caused by the process gases injected at lower temperature from the water-cooled injector is significantly reduced and the gas phase temperature above the rotated substrate rises with higher ceiling temperature. Also, the on-disc temperature becomes more uniform, which has been confirmed by in-situ measurement as shown in Figure 4, in which the on-satellite gradient was reduced by 4.8 K when the ceiling temperature was set to 1000°C.

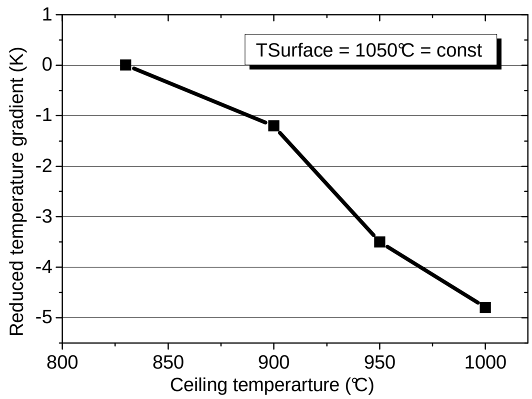

Figure 4: Reduction of the on-satellite disc temperature gradient for a constant surface temperature (reference $T_{Ceiling}$ = 830°C)

The influence of the hot ceiling on the deposition profile was evaluated by taking depletion curves experimentally and comparing them with the simulated results. Although one would expect that parasitic growth on the ceiling is enhanced due to the higher temperature, which would be indicated by a steeper decline of the depletion curve, no change in the tail was observed when comparing the growth profile over reactor radius (Figure 5). These results go well along with simulation, which predicted exactly the same behaviour. It is worth mentioning that a higher ceiling temperature did not caused shift in the depletion peak position, but a higher maximum peak growth rate, increasing the efficiency for the TMGa precursor in this configuration.

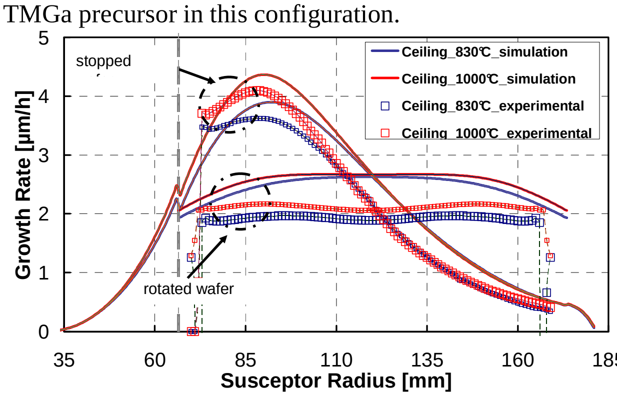

Figure 5: Experimental and simulated growth profiles for a ceiling temperature of 830°C and 1000°C at $T_{Surface}$ = 1050°C

These observations lead to the conclusion that even for 830°C, the parasitic deposition rate on the ceiling is purely transport-limited and no additional losses occur. The higher peak growth rate is caused by enhanced diffusion due to the hotter ambient. Further optimization of the initial growth resulted in a high crystal quality, indicated by improved XRD-RC with FWHM of 195 arcsec for (0002) reflex and 295 for (10-12) reflex. To judge the crystal quality by electrical properties, 2 µm of n-doped GaN was grown on a 3 µm GaN Buffer. As one can see in Figure 6, a very high mobility of 695 cm²/Vs was obtained for a carrier concentration of 1.3E17 cm^{-3}, may possibly be related to the low density of nitrogen vacancies as mentioned earlier. The sheet carrier concentration measured by Hall and ECV was nearly identical and showed a low background concentration of 7E15 for the undoped buffer.

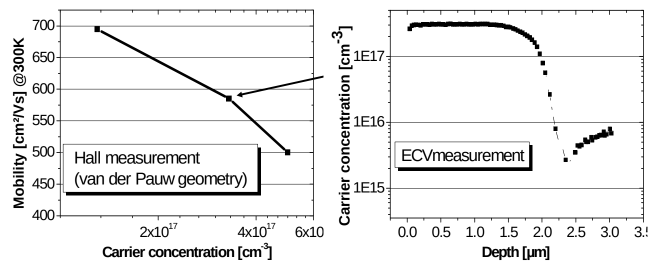

Figure 6: Carrier mobility vs. carrier concentration (left picture) and ECV depth profile for the n = 3E17 cm^{-3} sample (right picture)

FIRST INVESTIGATIONS FOR ALN AND ALGAN

To investigate the feasibility to grow Al alloys with a hotwall system, AlGaN was deposited on a GaN template. The Al content in film was determined by PL. The reactor pressure was set to 75 mbar and the surface temperature to 1060°C. The fraction of Al to total group III content in gas phase was set to 31.8% and a growth rate of 0.8 µm/h at a V/III ratio of 4240 was obtained. With these conditions, Al incorporation of 29.5% with a very uniform distribution (σ=0.85%) was achieved (Figure 7), indicating no sign of parasitic gas phase losses and a high Al incorporation efficiency.

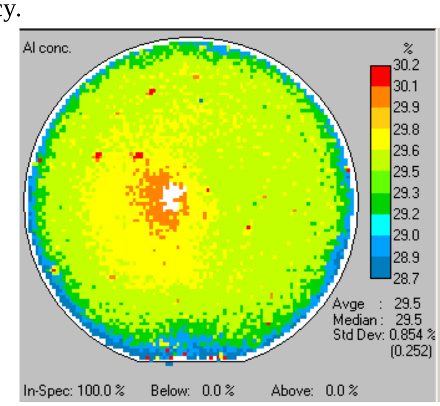

Figure 7: Al content distribution in film

The influence of increased growth temperature on the AlN growth profile along flow direction was investigated as well. For this approach, growth of AlN at 1050°C surface temperature ($T_{Susceptor}/T_{Ceiling}$: 1050°C/830°C) was compared with high-temperature growth conditions at a susceptor/ceiling temperature of 1200°C/1100°C, as a result providing a wafer surface temperature above 1200°C. AlN was deposited on AlN templates and the deposition profile was measured. As one can see in Figure 8, the depletion profile is significantly influenced by the temperature, which can be explained by enhanced diffusion as well as faster decomposition of the TMAl precursor for the higher susceptor surface temperature.

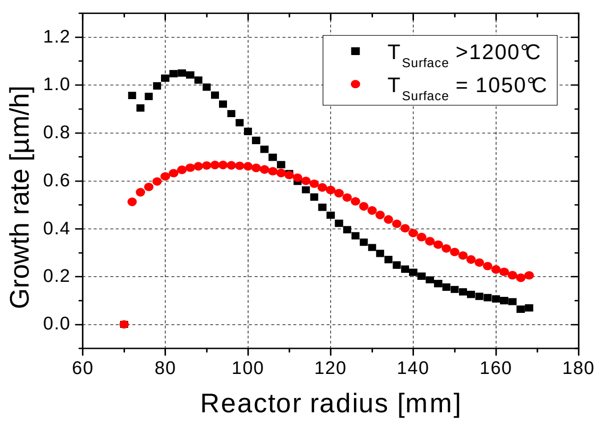

Figure 8: Growth rate distribution for AlN at $T_{Surface}$ = 1050°C and $T_{Surface}$ > 1200°C

CONCLUSIONS

Achieving almost isothermal conditions within the process chamber is established setting the ceiling temperature to match the actual growth temperature which optimizes the on-satellite temperature uniformity. Furthermore, the substrate temperature homogeneity becomes independent of the wafer bow as a significant improvement. Using a hot ceiling does not cause additional losses through parasitic growth, as one may expect, but rather increases the growth rate in consequence of the enhanced diffusion to the substrate in conjunction with the improved atomic nitrogen supply mentioned before. Additional increase in ceiling temperature equal or above 1100°C is not beneficial due to degradation of the parasitic coating on the ceiling leading to formation of particles. We conclude using the same temperature for the ceiling and susceptor demonstrates ideal conditions for the growth of GaN enabled through the specific design of the Planetary Hotwall set-up. Matching results between simulation and experiments confirm our understanding of the parasitic loss mechanism.

First investigations for $Al_xGa_{1-x}N$ show a very homogeneous Al distribution and an efficient Al incorporation at least up to a film composition of x = 29.5%. The high temperature capability of the tool and the homogenous deposition of $Al_xGa_{1-x}N$ address the needs for development of deep UV LEDs. Further investigations for higher Al contents in AlGaN as well as high-temperature AlN will follow.

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ACRONYMS

AFM: Atomic Force Microscopy
FWHM: Full Width Half Maximum
NH_3: Ammonia
MO: Metal-Organic
MOCVD: Metal-Organic Chemical Vapor Deposition
PL: Photoluminescence
TMAl: Trimethylaluminum
TMGa: Trimethylgallium
XRD:X-RayDiffraction