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FABRICATION, TREATMENT, AND TESTING OF MATERIALS AND STRUCTURES

# Dependence of the Growth Rate of an AlN Layer on Nitrogen Pressure in a Reactor for Sublimation Growth of AlN Crystals

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**Abstract**—The dependence of the layer growth rate on nitrogen pressure in a reactor has been examined in order to analyze the conditions of growth of AlN thick layers and bulk crystals by the sublimation sandwich method. It is shown that the layer growth rate steadily increases as the pressure in the reactor is lowered within the range 1–0.02 bar. This suggests that a key role in the layer growth kinetics is played by the source-to-substrate transfer of the components (Al, N), rather than by their adsorption (desorption) on the substrate surface.

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## 1. INTRODUCTION

Obtainment high-quality thick layers and bulk crystals of AlN, suitable for use as substrates for optoelectronic and high-power microwave electronic devices, is a currently pressing problem of modern semiconductor technology. At present, it is the most successfully solved by using the sublimation sandwich method [1–3]. Even though impressive advances have been made in this way (single crystals 2″ in diameter and several millimeters thick have been grown) [3], researchers apply no less effort at determining the optimal growth conditions in which high-quality crystals can be obtained at the maximum growth rate.

One of the most important characteristics of the growth process is the state of the gas medium in the reactor (pressure, chemical composition). It is this state that largely governs the kinetics of material transfer from the source to a substrate, i.e., in the end, the crystal growth rate. It is also rather important that this characteristic profoundly affects the service life of reactor components (primarily, the heater in the resistive variant of heating).

All other conditions being the same, a decisive influence is exerted on the growth rate of the AlN layer as a function of pressure by two main factors: (i) adsorption (desorption) processes on the surface of the source and growing layer and (ii) transfer of the components (Al, $N_2$) from the source to the substrate. This problem has been theoretically considered in sufficient detail [4–7], with the experimental evidence remaining exceedingly scarce [6, 8]. An experimental analysis of the role of these factors in the dependence on the nitrogen pressure in the reactor with the aim of optimizing the growth process was the goal of this study.

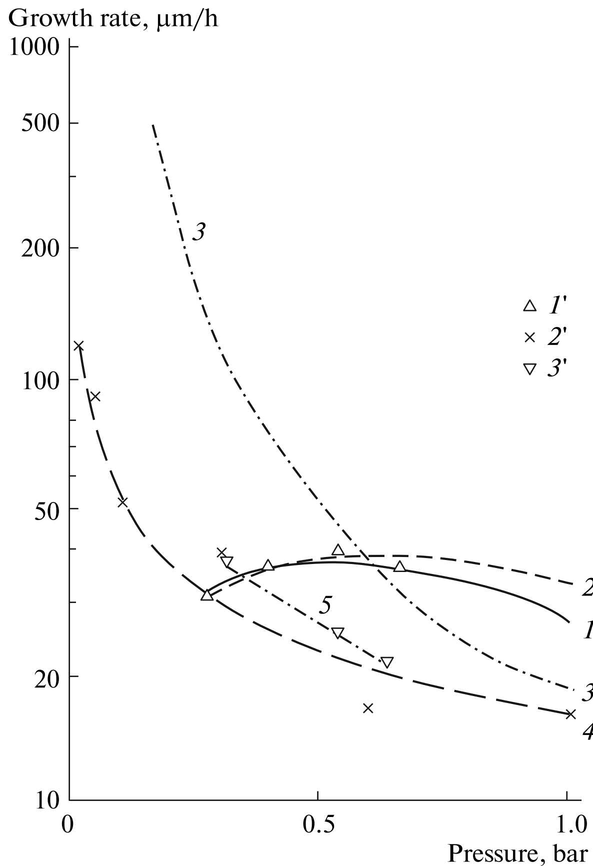
## 2. EXPERIMENTAL

The technological equipment and experimental procedure were for the most part described in [2]. As a resistive heater, we used a hollow graphite cylinder in which were placed a tantalum crucible with the source and a substrate firmly (but not vacuum tightly) adjacent to the upper rim of the crucible. Irregularly shaped 10- to 50-$mm^3$ polycrystalline blocks of an AlN powder, produced by preliminary distillation, served as the source, and a mechanically ground, polished, and etched in molten KOH plate of tantalum carbide was used as a substrate. It is undesirable to use, as it is commonly done, a SiC substrate in this case, because SiC is noticeably evaporated at high temperatures, which changes the gas composition in the growth cell and impairs the measurement accuracy of the layer growth rate. By contrast, TaC is highly stable against high temperatures. The gap between the upper edge of the source and the substrate was, on average, 3 mm (taking into account the nonplanar surface of the source).

The growth cell (crucible with the source and the substrate pressed against it) was mounted in the upper part of the heater in such a way that the temperature gradient (about 7 K/mm) was directed from the substrate to the source (i.e., the substrate was somewhat cooler). It is this circumstance that enabled the transfer of AlN and its condensation on the substrate.

In the experiments, all parameters of the growth process remained unchanged. Only the nitrogen pressure in the reactor was varied. The duration of the working phase of the process was 1 h, and the substrate temperature was about 2000°C.

We determined the layer growth rate from the increase in the weight of the substrate, rather than from that in the layer thickness, in order to preclude errors associated with the substrate deformation,




Calculated and experimental dependences of the growth rate of AlN layers on the nitrogen pressure in the reactor. (*1*) Calculated curve from [5] ($T$ = 2050°C; source–substrate gap 4 mm; $T$ difference across the gap, 7°). (*2*) Calculated curve from [6] ($T$ = 2157°C; $T$ gradient in the gap, 4.8 K/mm). Symbols *1*' represent the experimental data from the same study. (*3*) Calculated curve from [7] ($T$ = 1950°C; $T$ gradient in the gap, 5 K/mm). (*4*) Symbols *2*' show the results of our experiments ($T$ = 2000°C; grad $T$ in the gap, 7 K/mm). (*5*) Symbols *3*' represent the data from [8] ($T$ = 1700°C; source–substrate gap 1 mm).

thickness nonuniformity of the layer, irregularities of its surface, etc. The average thickness of a layer was calculated from its area and AlN density (3.25 g $cm^{-3}$).

## 3. EXPERIMENTAL RESULTS AND DISCUSSION

The figure shows the results of our experiments on determining the growth rate of an AlN layer in nitrogen atmosphere at pressures in the range 0.02–1 bar. The same figure presents the theoretically calculated curves and experimental data taken from [5–8].

Attention is immediately attracted to the difference in the nature of the calculated curves. Curves *1* [5] and *2* [6] are peaked at a pressure of 0.5 bar and decay at lower pressures, whereas curve *3* [7] exhibits a monotonic rise in the layer growth rate with decreasing pressure. A similar result was obtained in [4]. The cause of this difference is the fact that only the transport mechanism is taken into account in [4, 7]. Understandably, a decrease in the number of filler-gas atoms in the gap facilitates the transfer of the components from the source to the substrate. Two other calculations [5, 6] also take into account the contribution of adsorption (desorption) processes occurring on the AlN surface. The quantity characterizing the effect of these surface properties, the sticking coefficient, is commonly considered to be close to unity for Al and very small, on the order of $10^{-5}$ at 2200°C, for N [9]. It is the last circumstance that leads to a decrease in the calculated growth rate of the layer at low pressures, at which the deficit of nitrogen is increasingly important.

The results of our experiments (figure, curve *4*) definitely demonstrate that the layer growth rate increases with decreasing pressure. A similar result was obtained in [8] at nitrogen pressures in the range 0.3–0.6 bar. The different nature of experimental data in [6], with the maximum observed at around 0.5 bar, is possibly due to certain dissimilarities in the experimental conditions. Moreover, these data were only obtained for pressures exceeding 0.25 bar.

The similarity between the calculated curve *3* [7] and our experimental results and data of [8] apparently indicates that the role of surface adsorption (desorption) processes was overstated in the calculations made in [5, 6]. Possibly, their influence was manifested in that, as the pressure is lowered, the real growth rate increases much more slowly than does the calculated rate.

It follows from our results that considerable increase in the layer growth rate (at prescribed values of the source–substrate gap and grad $T$ in the gap) is possible because of the decrease in the nitrogen pressure in the reactor. Another, commonly used method consisting of raising the process temperature leads to an accelerated erosion of the heater, crucible, and other reactor members and to an increase in the defect concentration in the growing layer.

We successfully carried out growth processes at nitrogen pressures of 0.05 and 0.1 bar in the reactor. In the latter case, good-quality AlN layers (smooth, mirrorlike surface; half-width of the X-ray rocking curve not exceeding 10 min of arc) were obtained on 6*H*-SiC substrates at a substrate temperature of 2000°C.

## 4. CONCLUSIONS

The above results show the following.

(1) The growth rate of an AlN layer steadily increases as the nitrogen pressure in the reactor is lowered, which indicates that the component transfer across the growth cell gap predominates over adsorption (desorption) processes on the source and substrate surfaces.

(2) The growth rate of the AlN layer can be made higher by lowering the nitrogen pressure in the reactor,

rather than by raising the process temperature. This prolongs the service life of reactor members and diminishes the defect concentration in the growing layer.

## REFERENCES

1. Yu. A. Vodakov, E. N. Mokhov, M. G. Ramm, and A. D. Roenkov, Cristall Techn. **14**, 729 (1979).
2. E. N. Mokhov, O. V. Avdeev, I. S. Barash, T. Yu. Chemekova, A. D. Roenkov, A. S. Segal, A. A. Wolfson, Yu. N. Makarov, M. G. Ramm, and H. Helava, J. Cryst. Growth **281**, 93 (2005).
3. Yu. N. Makarov et al., J. Cryst. Growth **310**, 881 (2008).
4. S. Yu. Karpov, D. V. Zimina, Yu. N. Makarov, E. N. Mokhov, A. D. Reonkov, M. G. Ramm, and Yu. A. Vodakov, Phys. Stat. Solidi A **176**, 435 (1999).
5. A. S. Segal, S. Yu. Karpov, Yu. N. Makarov, E. N. Mokhov, A. D. Reonkov, M. G. Ramm, and Yu. A. Vodakov, J. Cryst. Growth **211**, 68 (2000).
6. T. K. Hossain, J. V. Lindesay, and M. G. Spenser, Mater. Sci., condmat.mtrl sci (Apr. 2002).
7. B. Wu, R. Ma, H. Zang, M. Dudley, R. Schlesser, and Z. Sitar, J. Cryst. Growth **253**, 326 (2003).
8. G. R. Yazdi, M. Syvajarvi, and R. Yakimova, Phys. Scr. T **126**, 127 (2006).
9. S. Yu. Karpov, Yu. N. Makarov, M. G. Ramm, and R. A. Talalaev, MRS Internet J. Nitride, Semicond. Res. **2**, 45 (1977).

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