Growth rate and its determining process for metalorganic vapor phase epitaxial InN

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Alloys of InN with GaN or AlN have recently attracted much attention as potential materials for green ～violet LDs and LEDs and high temperature high-power electronic devices. Because of the low dissociation temperature for InN, growth of InN has not been widely studied compared with GaN or AlN. Understanding of growth behavior of InN, therefore, is essentially desired in order to obtain high quality InGaN or InAlN crystals as well as high quality InN itself. Generally, growth rate of InN ever obtained in the conventional MOVPE using NH_3 as a nitrogen source is very low, less than 0.1 μm/h [1]. The reason for this and the possibility of obtaining a higher growth rate in the conventional MOVPE have not yet been clarified. Also, there have been few discussions about growth-rate determining process in the MOVPE for InN. In this paper, we report growth temperature dependence of growth rate of InN in the MOVPE and discuss growth-rate determining process in MOVPE of InN. By choosing growth conditions where the growth is rate-determined by indium source supply, the growth rate as high as 0.8 μm/h has been attained.

Using a conventional MOVPE apparatus with a horizontal reactor, InN films were grown. Trimethylindium (TMI) and ammonia (NH_3) were used as source materials and N_2 as a carrier gas. Growth temperature was varied from 500℃ to 700℃. Wafers of α-Al_2O_3 (0001) were used for substrates. Just before growth, substrate surface was nitrided in the flowing NH_3 at 900℃ for 30 min. NH_3/TMI molar ratio in the growth atmosphere was varied from $75x10^3$ to $4.5x10^3$.

Figure 1 shows growth rate of InN at a different growth temperature as a function of TMI supply for a constant NH_3 flow rate 6 SLM. It is pointed out that, in the temperature range of 500-600℃, growth rate for a constant TMI supply is increased with increasing growth temperature and shows a saturation against the increase in TMI supply. At a higher growth temperature of 650℃, on the other hand, such a saturation is not seen for TMI supply up to 28 μmol/min. The increase in growth rate at 650℃ with increasing TMI supply means that InN growth is rate-determined by TMI supply. The growth-rate saturation in the temperature range 500-600℃ shows that the growth rate is not governed by TMI supply but by NH_3 supply in this temperature range. This is because decomposition rate of NH_3 is considerably low at 500-600℃.The saturation is attributed to deficiency of active nitrogen. The increase in growth rate with temperature in this temperature range is due to the increase in NH_3 decomposition rate. By plotting saturated growth rate at each temperature against reciprocal temperature (1/T), activation energy for growth rate is obtained as about 0.8 eV. This value is in good agreement with the activation energy for thermal decomposition of NH_3 (～1 eV) [1], showing that the growth-rate saturation is due to deficiency of active nitrogen.

By increasing growth temperature up to 650℃, a high growth rate of 0.8 μm/h is attained. Figure 2 shows AFM images for InN films grown at a difference temperature for 2 hour. A very fine columnar structure is found for the film grown at 500℃ (Fig. 2(a)). The film grown at 650℃ shows a large grain size indicating that two-dimensional growth is highly enhanced (Fig. 2(c)). Thus, improvements of both growth rate and surface morphology for grown InN film are attained by employing high temperature (～650℃) growth. Figure 3 shows surface morphology for InN films grown at 600℃ with a different NH_3 supply. The TMI supply for both films is same; 2.8 μmol/min. The film grown with a 5 SLM NH_3 supply is not continuous and is composed of small islands, while that grown with a lower NH_3 supply is continuous and shows a larger grain size as a result of two-dimensional growth. This means that a lower V/III ratio, i.e., presumably In-rich condition enhances two-dimensional growth. It is also pointed out that the amount of deposited InN is markedly different for both cases; much less for the film grown with a 5 SLM NH_3 supply.

This seems to be due to the reduced driving force for the deposition reaction by an increased hydrogen partial pressure produced by the decomposition of NH_3, proposed by Koukitu et. al.[2].

In summary, we have studied temperature dependence of growth rate of InN by MOVPE and discussed growth-rate determining process in MOVPE of InN. At a lower temperature (500-600℃), growth rate is increased with increasing growth temperature and shows a saturation with an increase in TMI supply. These facts show that the growth is rate-determined by NH_3 supply in the temperature range. At a higher growth temperature of 650℃, growth rate is increased with TMI supply, showing that the rate-determining process is TMI supply. Nearly 1μm/h growth rate is obtained at a growth temperature around 650℃.

REFERENCES

[1] M. Mesrine et al., Appl. Phys. Lett. 72 (1998) 350.

[2] A.Koukitu et al., J. Crystal Growth 197 (1999) 99.

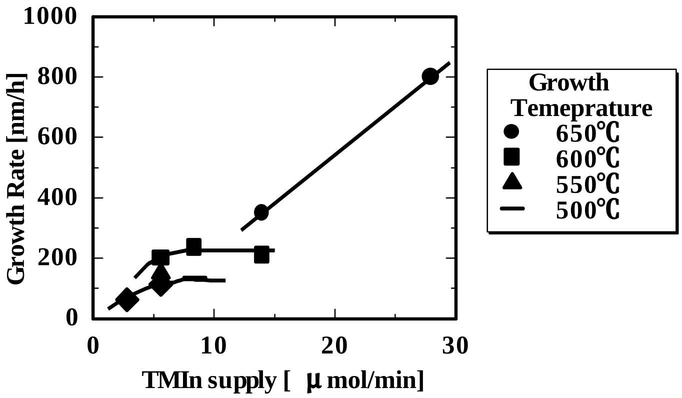
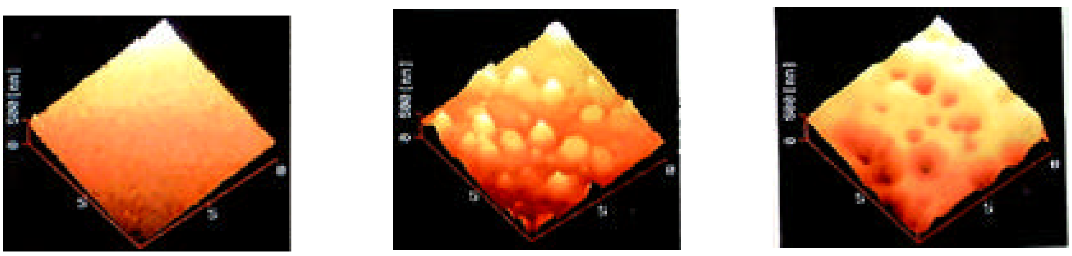
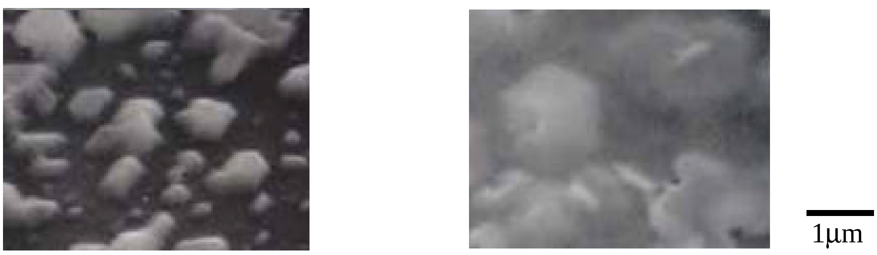


Figure 1. Growth rate of InN at a different temperature as a function of TMI supply.



(a) 500℃ (b) 600℃ (c) 650℃

Figure 2. Surface morphology for InN grown at different temperature.



NH_3 supply : 5slm NH_3 supply : 3slm

Figure 3. Surface morphology of InN grown with a different NH_3 supply.
TMI supply is 2.8umol/min for both.