We have studied the effects of growth temperature, film thickness, and Si doping on the phase separation in InGaN films grown by metalorganic chemical vapor deposition. As the growth temperature decreased, the band-edge photoluminescence peak became splitted due to the In composition fluctuation in the InGaN film and finally separated into two discrete peaks corresponding to the epitaxial InGaN and InN-rich phase. The size and the In content of the spinodally-formed InN-rich regions increased with increasing InGaN film thickness. The Si doping was found to suppress the composition fluctuation and the In incorporation in InGaN films.

1. Introduction

InGaN alloys have been used as active layer materials of III-nitride-based light-emitting diodes (LEDs) and laser diodes (LDs) [1]. Many researchers have reported the growth and characterization of InGaN films [2 to 7] and the phenomena of InGaN phase separation [8 to 11] since it has been reported that InN-rich regions play a significant role in the emission mechanism of nitride-based blue or green LEDs and LDs [12, 13]. However, it is still necessary to further investigate the optical and structural properties of phase-separated InGaN films in order to control the InGaN phase separation for the device application. In this work, we investigated the effects of growth temperature, film thickness, and Si-doping on the phase separation of InGaN films grown by low pressure MOCVD by taking the photoluminescence (PL), the synchrotron X-ray diffraction (XRD), and the conventional XRD of InGaN films.

2. Experimental

InGaN films were grown in a low pressure MOCVD system. The c-plane sapphire was used as a substrate. A 30 nm thick GaN buffer layer was grown at 500 °C followed by GaN epitaxial films at the elevated temperature of 1020 °C and then InGaN films were grown in the temperature range of 700 to 800 °C. PL measurements were carried out at room temperature using a He–Cd laser operating at 325 nm. The synchrotron XRD ($\lambda = 1.38$ Å) measurements were performed at the beamline 5C2 at Pohang Light Source in Korea and the conventional XRD ($\lambda = 1.54$ Å) measurements were carried out using a Cu-Kα X-ray source. The transmission electron diffraction (TED) measure-
ments were performed using a JEOL 2010 transmission electron microscope (TEM) operating at 200 kV.

3. Results and Discussion

Fig. 1a shows PL spectra measured at room temperature for 100 nm InGaN epilayers grown with 5 sccm of SiH₄. The decrease of growth temperature increased the FWHM of the PL peak and splitted the PL peak as shown in Fig. 1a. It was reported that the decrease of growth temperature increases the In-composition fluctuation in the InGaN epilayer [3, 4]. The In-rich regions may have a broad composition distribution or size distribution, resulting in an increase in the FWHM of the PL peak. To investigate the thermal properties of InN-rich regions, we carried out rapid thermal annealing (RTA) treatments for the films at a temperature range of 800 to 1050 °C for 5 min. For the samples treated by RTA, the PL peaks from InN-rich regions became narrower and shifted to the higher energy which is related to the band edge of epitaxial InGaN. This indicates that InN-rich regions are not thermally stable compared to the In-poor epitaxial regions. The separated low energy PL peak for the sample grown at 700 °C may come from the thermodynamically stable InN-rich phase [9]. However, even though the

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Fig. 1

**Fig. 1.** a) Room temperature PL and b) synchrotron XRD of 100 nm thick InGaN films grown at different growth temperatures

Fig. 2

**Fig. 2.** a) Room temperature PL and b) conventional XRD of undoped InGaN films with different film thicknesses grown at 760 °C
PL peak of In-rich regions is completely separated from that of epitaxial InGaN grown at 700 °C, the synchrotron XRD curve does not show the In-rich regions probably due to the detection limit of synchrotron XRD as shown in Fig. 1b. This result implies that the size of InN-rich regions may be of the order of nm under our growth conditions.

Fig. 2a shows the room-temperature PL of InGaN films grown at 760 °C with different film thicknesses. For a 30 nm thick InGaN, a sharp single PL peak corresponding to near-band edge transition in the epitaxially-grown InGaN phase appeared at 420 nm. However, for a 100 nm thick film, two additional peaks appeared in the lower energy side. When the film thickness increased to 300 nm, the additional PL peak was completely separated from the 420 nm PL peak as shown in the inset of Fig. 2a. The separated PL peak seems to originate from the InN-rich regions. This result agrees with the XRD curve shown in Fig. 2b. The broad FWHM of 30 nm thick InGaN in Fig. 2b can be caused by the thickness of a very thin film [14] and two small peaks on both sides of the InGaN peak are originated by the thickness fringe effect. For the 100 nm thick film, a 2θ peak corresponding to an epitaxial InGaN appeared. For 300 nm thick InGaN, however, an InN-rich phase was formed in the film as clearly shown in the XRD curve. Applying the Scherrer formula [14], we obtained the domain size of 26 nm for the InN-rich regions. To confirm the formation of InN-rich regions, we carried out TED measurements. The TED pattern for the 300 nm thick film showed splitted diffraction spots, indicating the existence of two phases with different In compositions [5]. However, we could not find splitted spots for the film of 100 nm. From these results, it can be con-

![Fig. 3. Room temperature PL spectra of a) 100 nm and b) 300 nm thick Si-doped InGaN films grown with different SiH4 fluxes](image-url)
cluded that the spinodally-formed InN-rich regions grow in size and the In content in these regions increases as the film thickness increases under our growth conditions.

Fig. 3a shows the PL of 100 nm thick InGaN films grown at 750 °C with different amount of SiH₄ fluxes. As the SiH₄ flux increases, the intensity of the 421 nm peak increases more predominantly than that of 465 nm peak [1, 7]. Fig. 3b shows the PL of phase-separated 0.3 μm InGaN films grown with various SiH₄ fluxes at 760 °C. An NH₃ flux was decreased during the growth of InGaN films to enhance the In composition fluctuation in the InGaN film [3]. As the SiH₄ flux increases, the PL peak corresponding to the InN-rich region shifts toward the higher energy side and its line width decreases as shown in Fig. 3b. From the conventional XRD measurements, we estimated the In contents in the InGaN films by using the ideal Vegard’s law assuming that the films are fully relaxed. The In contents of InN-rich regions grown using SiH₄ flux of 0, 5, and 30 sccm were thus calculated to be 28.1, 27.3, and 26.6%, respectively. It is believed that the Si doping in the InGaN film increases the coherency strain in the InGaN film since the atomic radii of Ga and In are larger than that of Si. Therefore, the Si incorporation in InGaN films may suppress the formation of InN-rich regions and also the In incorporation in InGaN films.

4. Conclusions
We investigated the effects of growth parameters on the phase separation of InGaN films grown by MOCVD. The spinodally-formed InN-rich regions grew in size and the In content of the regions increased when the InGaN film thickness increased. The Si incorporation in the InGaN film was found to suppress the composition fluctuation and also to decrease the In incorporation in InGaN films.

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References