Thin Solid Films 551 (2014) 142–145

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Influence of Pre-trimethylindium flow treatment on blue light emitting diode

Bing Xu [a], Jun Liang Zhao [a], Hai Tao Dai [a,*], Shu Guo Wang [a], Ray-Ming Lin [b,**], Fu-Chuan Chu [b], Chou-Hsiung Huang [b], Sheng-Fu Yu [c], Xiao Wei Sun [d,***]

[a] Tianjin Key Laboratory of Low Dimensional Materials Physics and Preparing Technology, School of Science, Tianjin University, Tianjin 300072, China
[b] Graduate Institute of Electronic Engineering and Green Technology Research Center, Chang Gung University, Taoyuan 333, Taiwan
[c] Institute of Microelectronics and Department of Electrical Engineering, Center for Micro/Nano Science and Technology, Advanced Optoelectronic Technology Center, National Cheng Kung University, Tainan 70101, Taiwan
[d] South University of Science and Technology of China, Shenzhen, Guangdong, China

ARTICLE INFO

Article history:
Received 31 May 2013
Received in revised form 25 November 2013
Accepted 27 November 2013
Available online 4 December 2013

Keywords:
Light emitting diodes
Trimethylindium treatment
Opto-electronics

ABSTRACT

The effects of Pre-trimethylindium (TMIn) flow treatment prior to quantum well growth on blue light emitting diode properties were investigated. High-resolution X-ray diffraction indicated that Pre-TMIn flow treatment did not change the composition of indium in quantum wells, but influenced electrical and optical properties of blue light emitting diode. Electroluminescence exhibited redshift with increasing TMIn treatment time. Though, the forward voltage became a little larger with longer Pre-TMIn treatment time due to the slight phase separation and indium aggregation, the efficiency droop of the device was improved effectively.

1. Introduction

III–V compound semiconductors are considered as key materials for the next generation lighting source. Especially, indium gallium nitride (InGaN) is proposed as the most promising material for high-efficiency light emitting diode (LED) devices due to its attractive features, such as long theoretical lifetime, direct band gap, widely spanned emission spectrum, and design flexibility. GaN-based LEDs and violet laser diodes have been commercially available in the past decade [1,2]. Until now, the emission efficiency of LED has been improved drastically, but it is still highly desirable to achieve higher luminescence at larger current densities for high power LED devices. Whereas, efficiency droop [3,4], which refer to the dramatically decreasing of external quantum efficiency (EQE) at high injection current density, inhabits the development of ultra-high power GaN-based LED. Many physical mechanisms have been modeled to interpret this behavior, such as Auger recombination [5], current roll-off [6], carrier delocalization [7–9], and polarization field [10,11]. Nevertheless, the main reason is still under discussion. On the other hand, the internal quantum efficiency of GaN-based LED can also be further improved by means of optimizing the quality and thickness of InGaN/GaN quantum well (QW) layers to reduce the non-radiative centers, especially V-shaped defects [12,13]. Considering the easily generating of V-shaped defects in low temperature and high indium composition, there are two methods to prevent the formation of V-shaped defects. One is to use growth interruption at high growth temperature to obtain flat InGaN surface by means of strain relaxation and indium desorption [14], the other growth technique – only trimethylindium (TMIn) and ammonia (NH_3) were allowed to flow into the reactor – called Pre-TMIn treatment, which can enhance the quantum efficiency and light output power of LEDs. These improvements were attributed to the reduced V-shaped defect density, surface smoothing process, and better QW structures. TMIn treatment can suppress InGaN decomposition, and increase the indium aggregation that acted as localized states to trap carriers for radiative recombination, similar to the effect of quantum dots. Consequently, LEDs can perform stronger photoluminescence intensity and higher light output power.

In this study, the effect of Pre-TMIn treatment time on the performance of InGaN-based LEDs was investigated experimentally. For comparison, four samples were prepared in experiment with 0 s, 60 s, 120 s, and 180 s Pre-TMIn treatment. According to the experimental results, the efficiency droop can be improved by increasing Pre-TMIn treatment time. Electroluminescence spectra exhibited redshift with longer Pre-TMIn treatment though the indium composition kept

* Corresponding author. Tel.: +86 15620398753.
** Corresponding author.
*** Corresponding author.
E-mail addresses: htdai@tju.edu.cn (H.T. Dai), rmlin@mail.cgu.edu.tw (R.-M. Lin), xwsun@sustc.edu.cn (X.W. Sun).

0040-6090/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.tsf.2013.11.108

constant. The forward voltage increased with the prolonged Pre-TMIn treatment time, which is attributed to the carriers trapping induced by the enhanced phase separation and indium aggregation during Pre-TMIn treatment. Furthermore, it was found that external quantum efficiency can be improved by using Pre-TMIn treatment during the growth of quantum wells.

2. Experiments

InGaN/GaN multiple quantum well samples were grown on (0001) sapphire substrate using atmospheric-pressure metalorganic chemical vapor deposition system. Trimethylgallium, trimethylindium, trimethylaluminum, and ammonia are used as precursors of Ga, In, Al and N respectively. Bicyclopentadienyl and silane are used as the p-type and n-type dopant precursors. Prior to growth, the substrate was first heated in H_2 ambient to remove any contaminants. Fig. 1 shows the schematic of the LED sample in experiment, included a 25 nm low temperature GaN nucleation layer, a 1 m undoped GaN buffer layer, a 3 m n-GaN layer, seven periods of multiple quantum well layers containing 2.7 nm InGaN well layers and 8 nm GaN barrier layers (in our experiment, there are three set of QWs. The first set: two pairs of InGaN QWs and GaN barriers were grown at 860° C and TMIn proflow temperature is 860° C; the second set: three pairs of InGaN QWs and GaN barriers were grown at 810° C and 860° C, respectively, and TMIn preflow temperature is 810° C; the third set: three pairs of InGaN QWs and GaN barriers were grown at 810° C and the TMIn preflow temperature is 810° C), a 10 nm AlGaN electron blocking layer and a 100 nm p-GaN layer. Note that, at each InGaN-to-GaN interface before InGaN layer, only TMIn and NH_3 were allowed to flow into the reactor. LEDs had a mesa structure with an area of 1×1 mm^2 were formed using conventional photolithography.

Four samples were prepared, marked as reference, TMIn-60 s, TMIn-120 s, TMIn-180 s, in order to analyze the effect of the Pre-TMIn treatment. High-resolution X-ray diffraction (HR-XRD) system equipped with a Göbel mirror and a Bartels-4-crystal monochromator at the incident beam side and a xenon point detector at the diffracted beam side have been used. Measurements were carried out using Cu $K_{\alpha 1}$ radiation ($\lambda = 0.1542$ nm). A typical 2θ scan in the range of 32–37° and a step size of 0.02° were employed. Current versus applied voltage character was measured using an Agilent B1500A semiconductor parameter analyzer. Electroluminescence and the light output power were measured by USB4000 optical spectrometer and an integrated sphere detector system. To avoid thermally induced degradation of the LED output power, pulsed mode measurement was employed, to analyze current effect on the external quantum efficiency. For pulsed measurement, a pulse width of 1 ms and a duty cycle of 0.1% was used with a maximum injection current up to 1 A.

Fig. 1. The structure of LEDs devices.

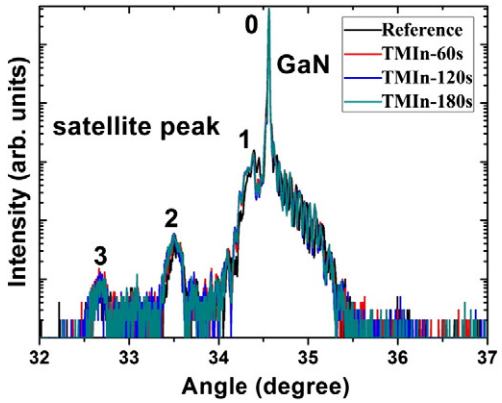

Fig. 2. (002) ω-2θ HR-XRD data for all the samples.

3. Results and discussion

Fig. 2 shows (002) ω-2θ HR-XRD data for all the samples. The diffraction peak corresponding to GaN can be verified near 34.5° labeled as peak 0, which is mainly originated from the GaN in buffer and contact layers. And the indium composition was calculated using Vegard's law from lattice parameters determined by XRD. In our experiment, indium content is estimated about 16.8% for the reference sample and 17% for the other samples, and the sum thickness of well and barrier layers was calculated by the constant distances between the periodic fringes (peak 2 and peak 3). And according to the growth rate of InGaN and GaN, well lay and barrier lay thickness can be calculated and the result shows that the well thickness was 2.7 nm for all the samples. In consequence, TMIn treatment cannot apparently affect the indium content or the thickness of the quantum well. This result leaded us to postulate that TMIn treatment may suppress the out diffusion of indium rather than form InN film.

Room temperature photoluminescence spectra of the four samples are shown in Fig. 3. Photoluminescence peak is obtained at 444 nm, 456 nm, 466 nm, and 467 nm, respectively for reference, TMIn-60 s, TMIn-120 s, and TMIn-180 s. It is obvious that the redshift occurred with increasing the treatment time. For the TMIn-180 s sample, the redshift was nearly the same as the TMIn-120 s sample, which is owed to the solubility limit of indium in GaN. In general, the redshift is responded to the large friction of In composition, however HR-XRD

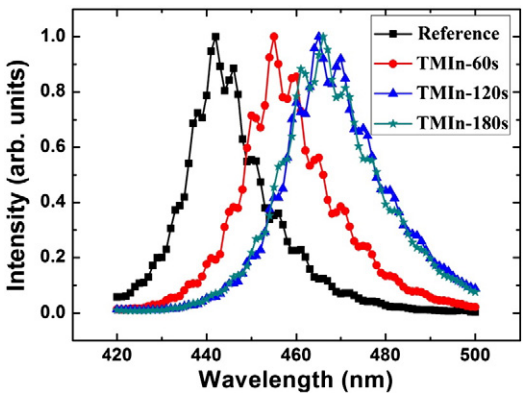

Fig. 3. Room temperature photoluminescence spectra of all the samples.