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Influences of elevated thermal decomposition of ammonia gas on indium nitride grown by sol–gel spin coating method

Zhi Yin Lee[a,b,*], Sha Shiong Ng[a], Fong Kwong Yam[b], Zainuriah Hassan[a]

[a] Institute of Nano Optoelectronics Research and Technology (INOR), Universiti Sains Malaysia, 11800 USM, Penang, Malaysia
[b] School of Physics, Universiti Sains Malaysia, 11800 USM, Penang, Malaysia

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ABSTRACT

Indium nitride (InN) thin films grown on aluminum nitride on p-type silicon (111) [AlN/p-Si(111)] substrates are prepared via sol–gel spin coating method followed by nitridation. The thermal decomposition effects of ammonia (NH_3) gas on the structural properties and surface morphologies of the deposited films are investigated. X-ray diffraction results reveal that the crystalline quality of InN degrades markedly as thermal decomposition of NH_3 gas increases from 700 to 850 °C, at which indium oxide (In_2O_3) forms in the deposited films. The thermal etching effect and formation of indium droplet on the film are observed at 850 °C. These findings are consistent with those of elementary and cross-sectional analysis obtained through energy dispersive spectroscopy and field-emission scanning electron microscopy. The findings deduced that InN thin films with densely packed grains can be grown at NH_3 decomposition temperature of 700 °C.

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1. Introduction

Group III-nitride compounds, specifically indium nitride (InN), have received substantial research interest primarily because of its unique properties, such as high electron mobility, small electron effective mass, and low carrier concentration [1]. The breakthrough of achieving the narrow energy band gap of 0.7–1.0 eV has further sparked the attention of researchers and manufacturing industry [2,3]. Since then, InN has been examined extensively, and the optical and electronic devices, such as light-emitting diodes, laser diodes, high-efficiency solar cells, and high-electron-mobility transistors, have been developed. These compounds can also be a potential candidate for water splitting applications because of their good electrochemical behavior with the incorporation of zinc oxide [4,5].

Despite the rapid development of InN-based semiconductor devices, their growth mechanism is associated with various limitations, due to stoichiometric instability, large lattice mismatch between the films and substrates, and low dissociation temperature [6,7]. To alleviate the lattice mismatch issues, researchers introduced various buffer layers, such as GaN, AlN, GZO and In_2O_3. The used of buffer layers have demonstrated buffer nucleation and highly *c*-preferred orientation InN [8,9]. Further studies have been conducted to produce InN below its dissociation temperature. However, low growth temperatures have resulted in the deficiency of active nitrogen atoms and reduction of the kinetic energies of ablated indium atoms in forming InN bonds [10].

Various sophisticated deposition techniques, such as metal–organic chemical vapor deposition, molecular beam epitaxy, and reactive sputtering, have been applied to synthesize InN thin films [11,12]. Although these conventional methods are beneficial for large area deposition, excellent composition control, and film uniformity, these methods require the availability of suitable precursors and ultrahigh vacuum systems. These methods also involve a toxic precursor and a relatively expensive and complicated setup. Hence, simple, safe, non-toxic, and low-cost deposition methods to produce InN thin films is highly desirable.

In the present work, we propose an easier and cost-effective alternative method for synthesizing InN, namely, sol–gel spin coating followed by nitridation [13]. In this method, the effects of the thermal decomposition of ammonia (NH_3) gas on the growth of InN thin films were investigated. The structural properties, surface morphologies, and elemental analysis of the deposited thin films were characterized through X-ray diffraction (XRD), field-emission scanning electron microscopy (FESEM), and energy dispersive spectroscopy (EDS).

* Corresponding author at: Institute of Nano Optoelectronics Research and Technology (INOR), Universiti Sains Malaysia, 11800 USM, Penang, Malaysia.
E-mail address: lee_zhiyin2003@yahoo.com (Z.Y. Lee).

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2. Experimental details

In this work, commercial AlN/p-Si(111) templates were used as a substrate for the sol–gel spin coating growth of the InN thin films. The substrates were cleaned by a chemical solution of hydrofluoric acid and distilled water with a ratio of 1:50 for approximately 1 min; then, rinsed with distilled water, and dried with nitrogen gas. A homogeneous solution was prepared by dissolving indium nitrate hydrate [$In(NO_3)_3 \cdot xH_2O$, purity 99.999%] powder in ethanol solvent (purity 99.7%) through ultrasonic agitation. The resulting precursor was placed onto the substrates dropwise and rotated for 30 s at 3000 rpm. This process was repeated for several times to form a thin layer with a certain thickness. Post-thermal annealing at 300 °C under nitrogen atmosphere was performed for 2 h to dry and refine the film structure. Subsequently, InN thin film was synthesized through nitridation by using two tube-furnaces connected in parallel under NH_3 atmosphere. The first furnace was adjusted at 700–850 °C to decompose NH_3 gas, and generate active nitrogen radicals, while the second furnace was set at 600 °C to induce a chemical reaction. A complete reaction process was conducted for 45 min at a constant NH_3 gas flow rate of 300 sccm. After the completion of the reaction, the sample was cooled down naturally to room temperature. Various characterization techniques were adopted to examine the properties of the deposited film. The crystalline structures were identified through XRD (PANalytical X'Pert PRO MRD) with $Cuk_{\alpha1}$ radiation source of 0.154 nm. Surface morphology and elemental analysis were examined by FESEM (FEI Nova NanoSEM 450) and EDS (which is attached to FESEM).

3. Results and discussion

Fig. 1 shows the XRD patterns of the deposited thin films at NH_3 decomposition temperature ranging from 700 to 850 °C. The samples show two common diffraction peaks at 28.0° and 36.0°, which originate from the silicon substrate and AlN buffer layer, respectively. Apart from that, the diffraction peaks associated with InN and indium oxide (In_2O_3) can also be detected, and the transformation mechanisms are explained as below. At 700 °C, the supplied NH_3 gas gradually decomposes into the reactive species of N_2 and H_2. Radicals, such as H, NH, and NH_2, with a relatively shorter lifetime are also produced. During the post-treatment at 300 °C, the coated film decomposes to form its metastable phase $In(OH)_2(NO_3)$ and release water vapor (H_2O) and dinitrogen pentoxide (N_2H_5).

The metastable phase further disintegrates to form In_2O_3 and H_2O. The reactive nitrogen atoms are transported through the surface diffuse into the film to produce InN and H_2O. The quasi-equilibrium of diffusion is achieved at the end of the reaction. The chemical routes can be expressed as follows [14]:

$$2NH_{3(g)} \rightarrow N_2 + 3H_{2(g)} \quad (1)$$

$$NH_{3(g)} \rightarrow H + NH_2 \quad (2)$$

$$In_2O_{3(s)} + 2NH_{3(g)} \rightarrow 2InN_{(g,s)} + 3H_2O_{(g)} \quad (3)$$

As a result, three intense diffraction peaks correspond to InN (100) at 29.1°, InN(002) at 31.3°, and InN(101) at 33.1° can be observed in Fig. 1(a).

However, further increment in thermal decomposition of NH_3 has degraded the crystallization of InN. At 750 °C, most of the InN diffraction peaks are diminished, as shown in Fig. 1(b), except the InN(100) and $In_2O_3(222)$ at 30.6°. By referring to Eq. (1), the forward reaction forming the product under the effect of temperature changes is endothermic, that is, the yield increases as temperature increases [15]. Under this condition, the partial pressure of H_2 in the system is higher than that of N_2. The lack of reactive nitrogen atoms leads to an inefficient diffusion to the coated film. Consequently, the conversion process of In_2O_3 into InN

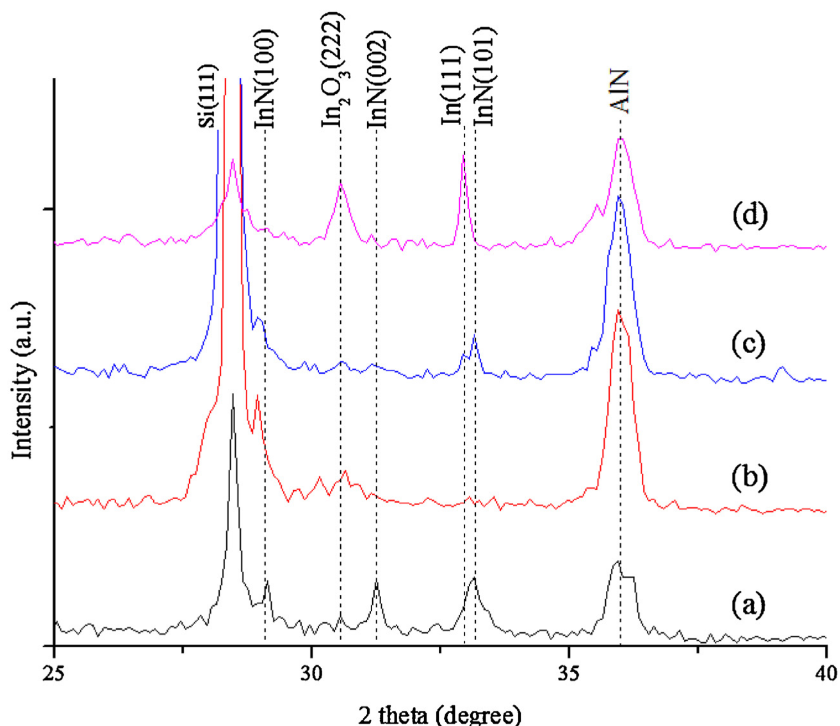

Fig 1. XRD patterns of the deposited thin films at thermal decomposition of NH_3 gas: (a) 700, (b) 750, (c) 800, and (d) 850 °C.

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