Effect of Substrate on Growth Mechanism of Flower Structured InN Fabricated by APHCVD

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Abstract. InN belongs to the III-group nitride materials and is known to have a low decomposition temperature which causes intractable grain growth compared to the other nitrides, GaN, AlN, etc. We prepared InNs with a flower-like shape as well as film structure by Atmospheric Pressure Halide CVD process, in which InN is synthesized by CVD under atmospheric pressure. In the present study, growth mechanisms of the flower structured InN prepared on Si(100) and a-plane sapphire substrates is reported.

Introduction. The III-group nitride materials attract attentions due to their electroluminescence properties. The GaN based nitride materials, e.g. AlGaInN, AlGaN etc. are under intense investigation[*1-4*] and widely applied for the blue and UV Light Emitting Diode since it has a band gap energy of the UV regions, which can be designed merely by controlling the composition of the solid solution between them. On the other hand, InN, which has a band gap energy of the IR region ~0.65eV, is far from practical application even though it has a unique properties of the high mobility of electrons and a promising material for the IR emitting/receiving optical devices. One of a key issue for application of the InN is its instability against high temperature. Unlike the other nitride materials, the InN is known to be unstable at high temperature and therefore it is quite difficult to control crystal growth mechanisms of the InNs. On this regard, many efforts has been made for investigating crystal growth mechanisms and for producing structure designed InN nano structures. L-Weber et al., reported InN layers and InN nanorods grown on the c- and r-planes of sapphire, where InNs were epitaxially grown on those substrates by the MOVPE method[*5*]. Hu et al., reported fabrication technique for InN nanobelts using a guided-stream thermal CVD [*6*]. They also reported the nanobelt structures with 40 to 250 nm width and 10 to 35 nm thickness showed infrared lasing. As an example of a two dimensional growth of InN nanostructure, a heteroepitaxial growth of the InN was reported by Harui et al[*7*]. They reported position controlled InN nanocolumns grown by MOVPE method on a patterned GaN epitaxial layers grown on sapphire substrate, on which reticular patterns of holes were intentionally fabricated by a focused ion beam (FIB) before the InN growth. These reports involves interesting results, however, they still have grown the InN in one or two dimensional growth direction, i.e. the nanocolumns or films. On the other hand in our previous reports, InN flower structures were successfully fabricated by APHCVD method[*8-10*]. Interestingly enough, it has three dimensionally uniform microstructures. In the present reports, we have fabricated the flower structured InN on both Si(100) and a-plane sapphire substrate in order to investigate the effect of the substrate on crystallization behavior of the InN flowers.

Experimental. Flower structured InN was prepared by APHCVD (Atmospheric Pressure Halide CVD) method. The APHCVD system is simple since the reaction tube can be composed of silica glass tube with several inlet/outlet. Experimental setup for the APHCVD method was shown in the fig. 1. All of the reaction steps occurred inside of a silica glass tube, which was attached with a rubber heater, electric furnace, and rubber tubes for nitrogen and ammonia gas inlet/outlet. Indium source, InCl3, placed on an alumina boat was placed at the center of the heating area surrounded by the rubber heater. Vaporized $InCl_3$ by the heater was transferred by N_2 carrier gas to a crystal growth region, which consisted of electric furnace and NH_3 gas inlet. At the crystal growth region, the transferred $InCl_3$ and NH_3 mixed and reacted on a substrate at a reaction temperature controlled by the furnace. Experimental condition for the flower structured InN growth was indicated at the table 1. The substrate used for growing InN flowers were Si(100) and sapphire (11-20) single crystals.
Crystallographic analysis of the flower structured InN prepared was measured by X ray pole figure (ATX-G, Rigaku Corporation, Japan) in order to observe in-plane orientation of the InN on the Si and sapphire substrates as well as observing a crystal structure of the InN products. Morphological study was made by the SEM (JSM-5510LV, JEOL co. ltd.).

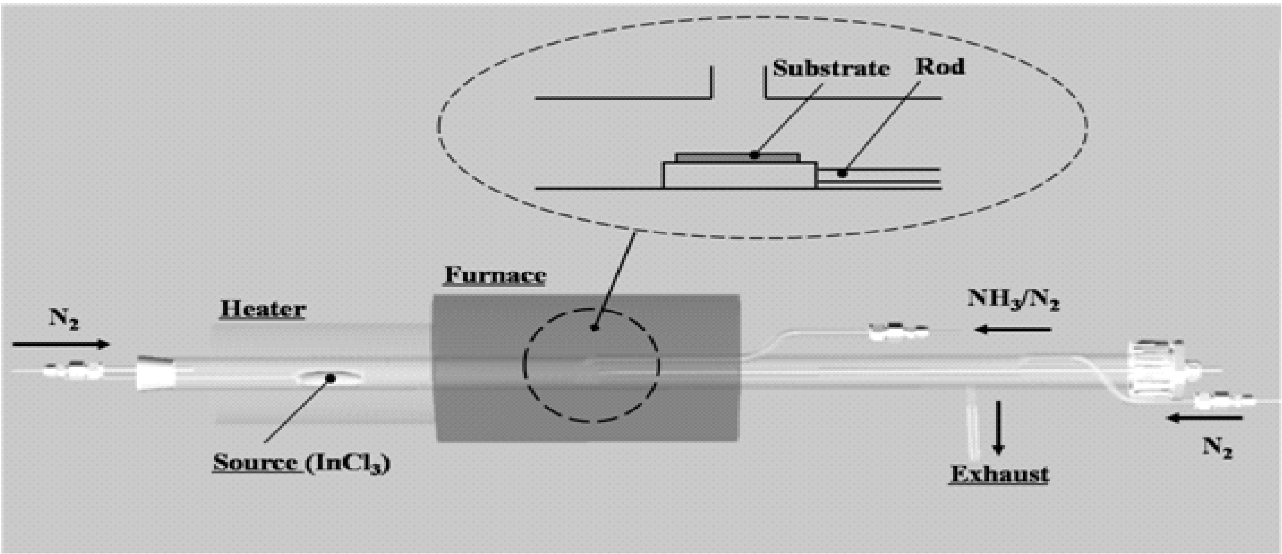

Fig. 1. Experimental setup for the APHCVD method. Inset surrounded by a circle shows location of the substrate, ammonia gas inlet, and silica glass rod supporting the substrate.

Table 1. Typical growth condition of the flower structured InN

Substrate	Si(100) or Sapphire (11-20)
V/III ratio	50
Growth temperature	550℃
$InCl_3$ heating temperature	410℃
Growth time	60min
$InCl_3$ partial pressure	4.6×10^{-3} atm
NH_3 partial pressure	2.3×10^{-1} atm
Total gas pressure	1.0 atm
Carrier gas	N_2

Results and Discussion.

1. Flower structured InN on Si(100) substrate

As shown in fig. 2(a), flower structured InN was successfully fabricated by the APHCVD method on Si(100) substrate. One can recognize that the flower was mostly composed of 7 parts of InN single crystals. As indicated in the SEM image, one single crystal grown perpendicular to the substrates was called as “Style” and the other six parts surrounding the style was called as “Petal” due to the similarity of the natural flower. Fig. 2(b) shows an X ray pole figure of the InN flowers on Si(100) substrate. A broad ring pattern indicates no orientation of the InN on the silicon substrate; it was not the epitaxial growth. Figures 2(a) and (b) are SEM image of the flower structured InN grown on the Si(100) substrate and a-sapphire substrate, respectively. Figures 2(c) and (d) shows X-ray pole figures for the InN (10-11) plane of the flower grown on Si (100) substrate and a-sapphire substrate, respectively. The pole figures indicate the InN flower on the Si substrate has no orientation in plane. On the other hand, the pole figure of the InN grown on a-sapphire substrate showed six isolated spots appeared which indicated that the InN has orientation in plane.

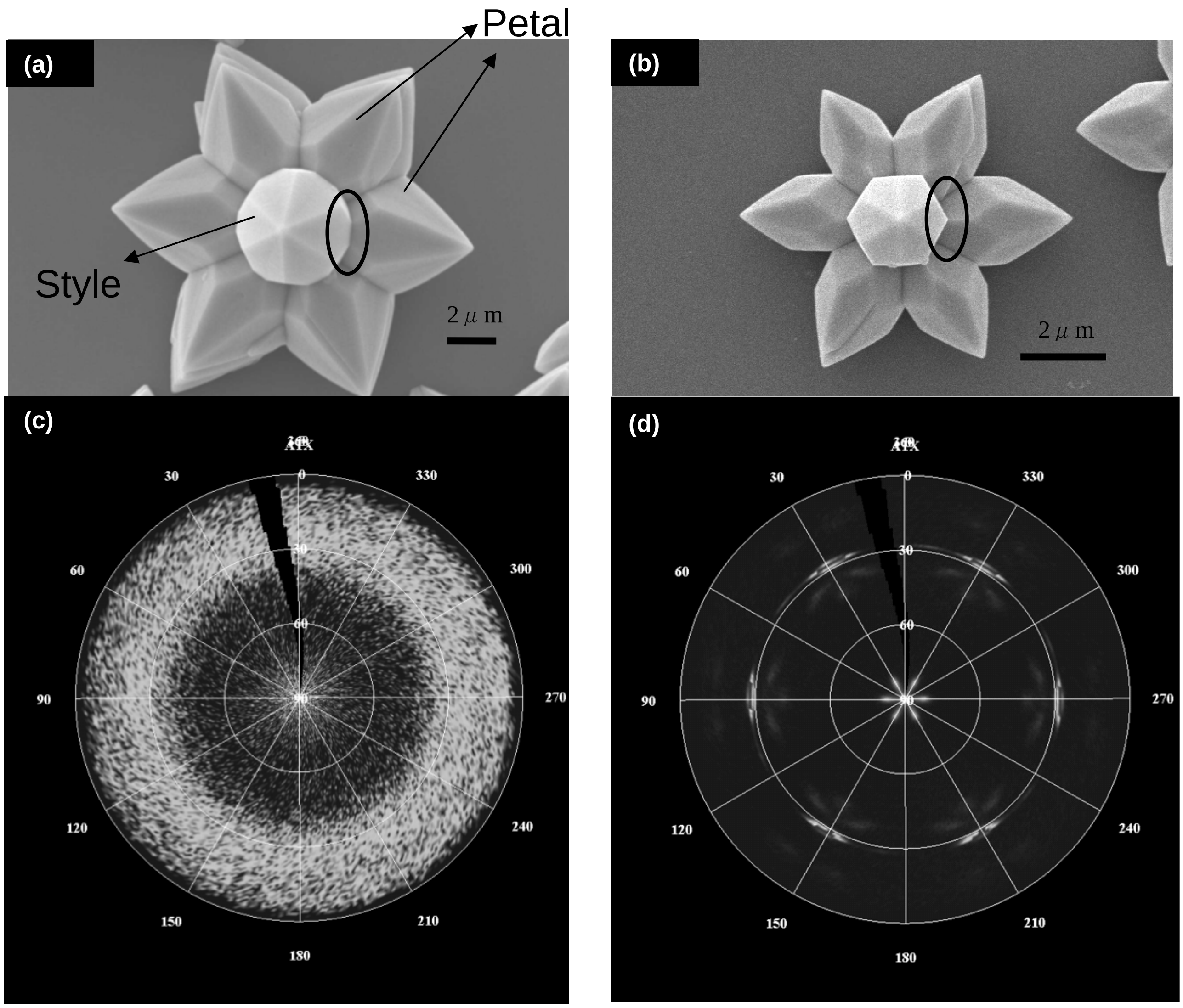

Fig. 2. SEM image of the flower structured InN grown on the Si(100) substrate(a) and on the a-sapphire substrate(b), and X-ray pole figure of the InN grown on Si (100) substrate(c), and on the a-sapphire substrate(d).

2. Flower structured InN on a-sapphire substrate

The morphology of the flowers grown on the a-sapphire substrate was similar to that seen on the silicon substrate except for the location of the petals surrounding the style as indicated with circles in the figs. 2(a) and (b). In the case of silicon substrate, the bottom of petals existed on the plane of the style edge, i.e. the style and the petal share the plane between them. On the other hand, in the InN flower grown on the a-sappire substrate the bottom of the petals existed on the edge of the style, i.e. the style and the petals share the corner of them. These difference maybe caused by the growth mechanisms difference of the flower structured InN on different substrates. Figure 2(c) and (d) also show a different crystallographic behavior between the two. When the InN flower grows on the a-sapphire substrate, it grows epitaxially as indicated by the isolated six spots appeared in the pole figure. The crystallographic difference also indicates the different crystallization mechanisms between these two substrates. As a reason for the epitaxial growth, we consider that the sapphire substrate may hold the InN more strongly than the silicon substrate due to lattice matching between the substrate and InN, although exact epitaxial relationship between the InN flowers and a-sapphire substrate is under investigation. Further investigation is progressing for understanding the nucleation / growth mechanisms of the flower like structured InN on different substrate using TEM observation.

Summary

Flower structured InN was prepared on the Si(100) substrate and a-sapphire substrate. Both of the InN flowers showed similar structures composed of 1 style and 6 petals. Morphological difference between them was that the contact point of the style and the petal; they share plane in the case of silicon substrate, and they share edge in the case of a-sapphire substrate. Crystallographic difference between them was the in-plane orientation; no particular orientation in the case of silicon substrate, and uniformly oriented in the case of a-sapphire substrate. These results indicated that the InN flower grown on the a-sapphire substrate was more strongly held from the substrate than the silicon substrate.

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