主 論 文 の 要 約

論文題目　Synthesis of GaN Film from Aqueous Solution
(水溶液中からの窒化ガリウム膜の合成)

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論 文 内 容 の 要 約

Gallium nitride (GaN) is a direct bandgap semiconductor and one of the most interesting materials in the optoelectronic device and high-power electronic device field. GaN has a wide bandgap energy of 3.39 eV at room temperature which can cover the entire UV visible range, thus it emits blue light efficiently and has been successfully employed in optoelectronics industry. In addition, GaN having a stable wurtzite structure has attracted attention as an alternative to silicon semiconductors since its high electron transfer speed and stability in high frequency region. Thus, for decades, many researchers have been trying various researches for high quality GaN, and have recently succeeded in producing single crystal GaN using by various dry methods such as MOCVD, MBE, HVPE, etc.

Nevertheless, high-cost of GaN manufacturing technologies are proposed as a challenge to overcome in its wide application and commercialization. Expensive equipment for high vacuum and high temperature environments and ultrapure source materials requiring complex processing are the main challenges in the commercialization of GaN. The main key for GaN commercialization is to reduce the costs used for high vacuum, high temperature environments and ultrapure sources. Due to these demands, electrochemical methods are again attracting attention, and electrodeposition has been proposed as one of the possible methods for GaN fabrication by its low-temperature and low-cost characteristics. Therefore, I tried to synthesize GaN film by applying electrochemical deposition method.

GaN films were synthesized on n-type-silicon (111) substrates using a low-cost and low-temperature technique of electrochemical deposition. The electrochemical behavior of gallium, ammonium, and nitrate ions in the aqueous solutions used as sources of GaN were confirmed by cyclic voltammetry. The scanning electron microscopy images showed that the films deposited at a current density of 3.5 mA cm-2 or greater have plate-like surface morphologies on the silicon substrate. The energy dispersive X-ray spectroscopy results showed that oxygen, gallium, and nitrogen coexist in these plate-like films. In the X-ray diffraction patterns, the sample synthesized at a current density of 3.5 mA cm-2 for 24 h exhibited peaks of cubic-GaN (c-GaN) and hexagonal-GaN (h-GaN) phases. Also, the peaks corresponding to gallium and gallium oxide phase were observed. Photoluminescence analysis revealed the peaks at 3.2 and 3.39 eV, which corresponds to the band gap energy of GaN, as well as a broad peak at around 2.5 eV at room temperature.

Next, the electrodeposition of GaN film has been attempted on various substrate materials. And electrochemical behavior of gallium and nitrogen ions on different electrodes. A cathodic current corresponding to the reduction reaction of each ion on different four substrates was confirmed. The hydrogen overpotential of electrodes influenced the reduction reactions of gallium and nitrogen species ions, and it was confirmed that the formation of adsorbed nitrogen on the substrate with high hydrogen overvoltage was easy. Reduction reaction of gallium occurred along with hydrogen evolution, and CV analysis confirmed the peaks of the reduction reaction of each ion separated on gallium and aluminum electrode. Nitrogen content of 20 mol% or more was observed from the films deposited on gallium and aluminum electrodes, and a peak due to reflection of GaN was detected by X-ray diffraction. In particular, a sharp peak corresponding to GaN was observed on Al substrate.

As previous research has confirmed that the properties of substrates affect the formation of GaN film, and the presence of high content of gallium and nitrogen in the film deposited on the aluminum substrate was confirmed. Therefore, the films were synthesized on aluminum substrates heat-treated at various temperatures to investigate the effect of aluminum. Deposited films showed that the plate-like polygonal structure and irregular cauliflower-like morphology. The presence of h-GaN and gallium oxide phases was confirmed by X-ray diffraction. Composition analysis revealed that more than 35 mol% of gallium and nitrogen are present with oxygen in the film. It was confirmed that the amount of oxygen in the electrodeposited films was significantly reduced by the effect of boric acid added into the solution as a buffer. The X-ray photoelectron spectroscopy and Raman analysis revealed that the signals due to h-GaN,

and the stronger signals appeared for the films formed on heat-treated aluminum substrate. In particular, the GaN film is more easily grown in the presence of alumina formed by heat treatment. The growth characteristics of electrodeposited GaN are strongly influenced by the composition and orientation of Al substrate. It is confirmed that the GaN film grows in a direction perpendicular to the preferred orientation of Al substrate and the GaN grows in the direction of making lattice mismatch small. In other words, it has been found that GaN growth can be controlled depending on substrate conditions.

Although the synthesis of single crystal GaN was not achieved, the possibility of synthesis of GaN from room temperature aqueous solution by electrodeposition was confirmed. Therefore, this is a simple one-step method for the synthesis of GaN, and this is possible to apply as the economical method for preparation of next generation materials.