9th Asia-Pacific Conference on Plasma Physics, 21-26 Sep, 2025 at Fukuoka

Incorporation of nitrogen to gallium oxide film on mist chemical vapor deposition

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1. Introduction

Incorporation of nitrogen into gallium oxide (Ga_2O_3) films can modify its electrical and optical properties. A gallium oxynitride (GaON) is a semiconductor with intermediate properties between gallium oxide (Ga_2O_3) and gallium nitride (GaN). The bandgap of GaON can be tuned from 2.2 to 2.8 eV by controlling the O/N ratio and it is expected to be used as a photocatalytic material for solar water splitting or photoelectrochemical (PEC) cells due to visible light responsiveness. GaON combines the chemical stability of GaN with the wide bandgap of Ga_2O_3, offering both durability and light absorption capability.

Mist chemical vapor deposition (Mist CVD) enables the formation of highly-oriented metal oxide films with high precursor utilization efficiency [1][2]. So far, there is few reports on the incorporation of nitrogen in to Ga_2O_3 film. The nitrogen gas is decomposed by the plasma and the nitrogen species are transported to the reaction area.

In this study, the incorporation of nitrogen to Ga_2O_3 film on Mist CVD was conducted by using the atmospheric plasma. The crystal structure and chemical composition of GaON were evaluated.

2. Experimental

A hot-wall-type Mist CVD equipment was used. A 0.05 mol/L solution of $Ga(acac)_3$ was prepared by dissolving it in ultrapure water containing 0.28 mol/L hydrochloric acid. The solution was atomized using an ultrasonic transducer operating at 2.4 MHz. Nitrogen gas as a carrier gas was decomposed by using an atmospheric pressure packed bed plasma to supply nitrogen species. The decomposed nitrogen gas species was transported to the mist generation unit. The films were deposited on C-plane sapphire substrate positioned at a 45 ° angle to the gas flow direction. The growth temperature was at 450 ℃.

3. Results and Discussion

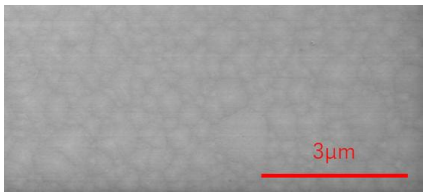
Table 1 shows the chemical composition of the deposited film measured by energy dispersive X-ray spectroscopy. The nitrogen was incorporated into the Ga_2O_3 film by supplying the plasma-decomposed nitrogen gas to the mist. The composition of nitrogen increased with increasing the applied voltage to atmospheric pressure packed bed plasma because the decomposition of nitrogen gas was promoted.

Figure 1 shows the SEM image of the deposited film. The spherical shape was observed for the film deposited without plasma decomposition, as shown in Fig. 1(a). The XRD peak of α-Ga_2O_3 (006) at 450℃ was observed for all samples. The structure of nitrogen-incorporated Ga_2O_3 film with plasma decomposition at 13.2kV was different from that of α-Ga_2O_3 without plasma decomposition. The size of structure deposited at 15.5 kV was slightly smaller than that at 13.2 kV. The incorporated nitrogen induced lattice strain and suppressed the surface diffusion of adatoms, resulting in a higher nucleation density. Accordingly, the structure was changed and the size was decreased because the crystal growth was inhibited by the nitrogen incorporation.

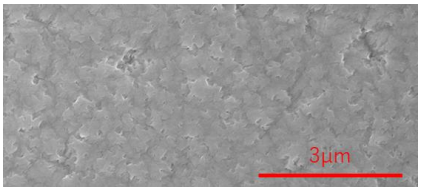
References
[1] T. Kawaharamura *et al.*, J.Appl. Phys., 53, 05FF08 (2014).
[2] C. Zhang *et al.*, Material, 12, 3670 (2019).

Table 1. Composition of deposited film.

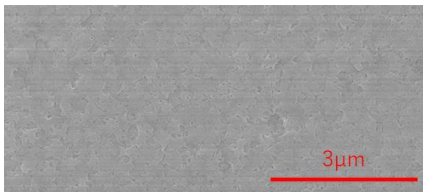
	Ga [%]	O [%]	N [%]
15.5kV	57.2	35.0	7.7
13.2kV	56.7	37.1	6.2
0kV	55.7	42.2	2<



(a) without plasma decomposition



(b) with plasma decomposition at 13.2 kV



(c) with plasma decomposition at 15.5 kV

Fig 1. Plane view of SEM images.