Suppression of Polycrystal Nucleation by Pre-Mixed Gas of Nitrogen and Methane in the Na-Flux Method

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Gallium nitride (GaN) vertical power devices are currently not widely used, and higher quality GaN substrates are required. We have produced high-quality GaN crystals by the Na flux method [1]. In the Na flux method, carbon has been added to the solution as an additive to suppress the nucleation of polycrystals that grow at locations other than on the seed substrate. We mainly use solid graphite as an additive, while Liu *et al*, have added various solid carbon sources and studied their effects [2]. We have also tried gaseous carbon addition with methane in the past [3]. The methane addition has achieved a suppression of polycrystal nucleation and an increase in the growth rate compared to graphite. However, past studies have shown poor reproducibility of growth rate. It is estimated that there were variations in methane concentration due to poor mixing of methane and nitrogen because they were prepared separately just before the experiment. In this study, we used pre-mixed gas of nitrogen and methane to see if it would be effective in suppressing polycrystal nucleation with good reproducibility.

Four conditions were examined: 0.5 mol% graphite (conventional condition), 1.0 mol% graphite, 1.2 vol% methane, and 4.0 vol% methane. The amount of polycrystals grown at 0.5 mol% graphite and 1.2 vol% methane was over 2 g. The amount of the polycrystals was 0 g at 1 mol% graphite, and nucleation of polycrystals was successfully suppressed. LPE crystals did not grow at 4.0 vol% methane. It is considered that the methane roughened the seed crystal surface before the LPE crystal begin to grow and inhibited its growth. To see the amount of polycrystals under conditions where LPE crystal grow, it was grown in a nitrogen ambient before the LPE crystal begin to grow, and after LPE crystals began to grow, we switch to methane-mixed ambient. The result is shown in Figure 1. Polycrystals were created under the conditions of 4 MPa pressure and 880°C, and their weight was 1.6 g. Since low-pressure growth is effective in suppressing nucleation of polycrystals, the pressure was set to 3.3 MPa, but the polycrystals were created, in amount of 1.0 g. When the pressure was further reduced to 3.2 MPa, no polycrystals were created. This indicates that methane has the same suppression effect on the nucleation of polycrystals as graphite. In future, new approaches will be possible, such as controlling carbon concentration.

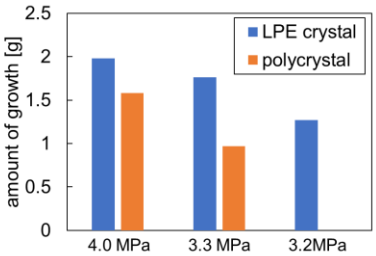

Figure. 1 Amount of LPE crystal and polycrystal in the growth with 4.0 vol% methane added from the middle of growth process.

References

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