

## Supporting information

### Acidic ammonothermal growth of GaN crystal using GaN powder as nutrient

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Figure 1. Ammonothermal crystal growth of GaN using polycrystalline GaN at 150 MPa

Figure 2. Top-view (left) and cross-sectional (right) photographs of GaN crystals grown using polycrystalline GaN with different thickness. (a) 50 $\mu$ m; (b) 200 $\mu$ m; (c) 374 $\mu$ m

Figure 3. XRC from 002 reflections of GaN crystals grown using polycrystalline GaN with different thickness.

Figure 4. SEM images of GaN crystals grown using grown using polycrystalline GaN with different thickness.

Figure 5. Images of HVPE seed and different nutrients

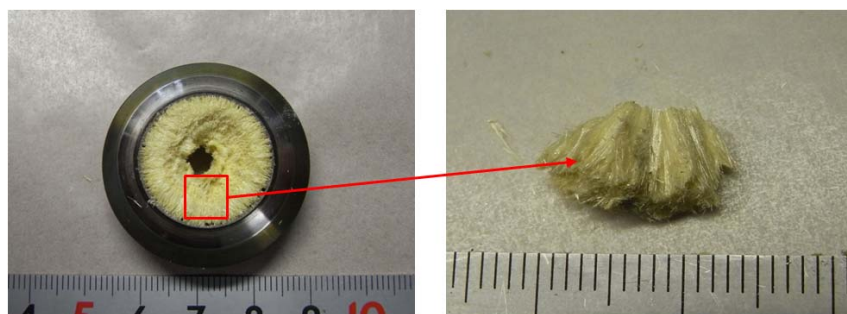


Figure 1. Ammonothermal crystal growth of GaN using polycrystalline GaN at 150 MPa

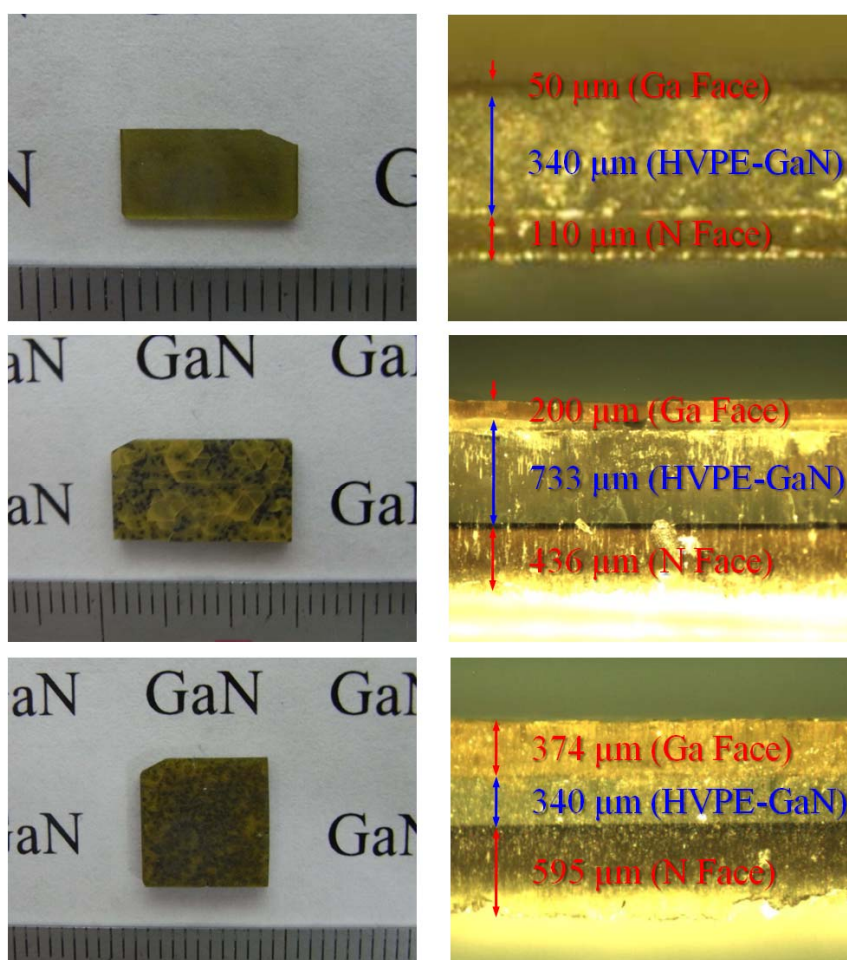


Figure 2. Top-view (left) and cross-sectional (right) photographs of GaN crystals grown using polycrystalline GaN with different thickness. (a) 50 μm; (b) 200 μm; (c) 374 μm.

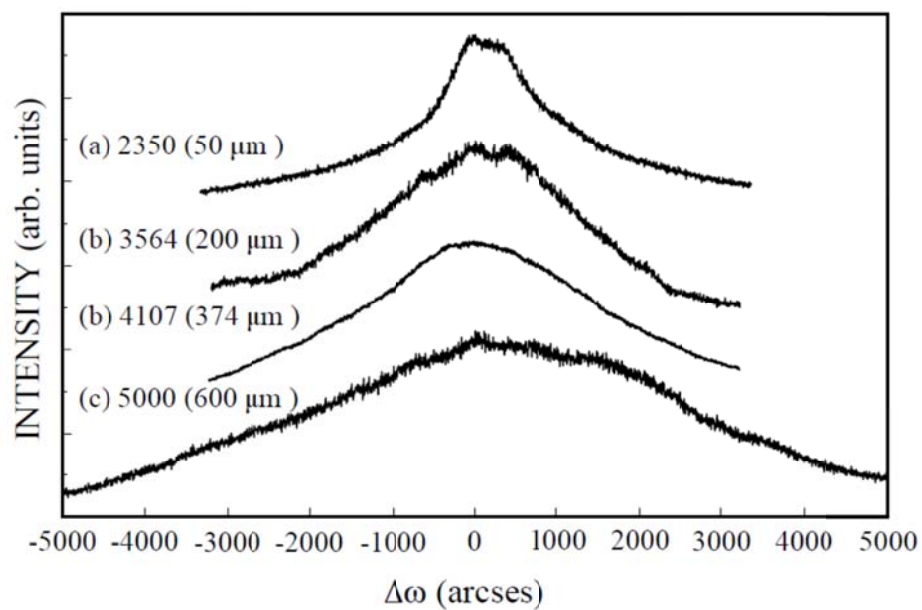


Figure 3. XRC from 002 reflections of GaN crystals grown using polycrystalline GaN with different thickness.

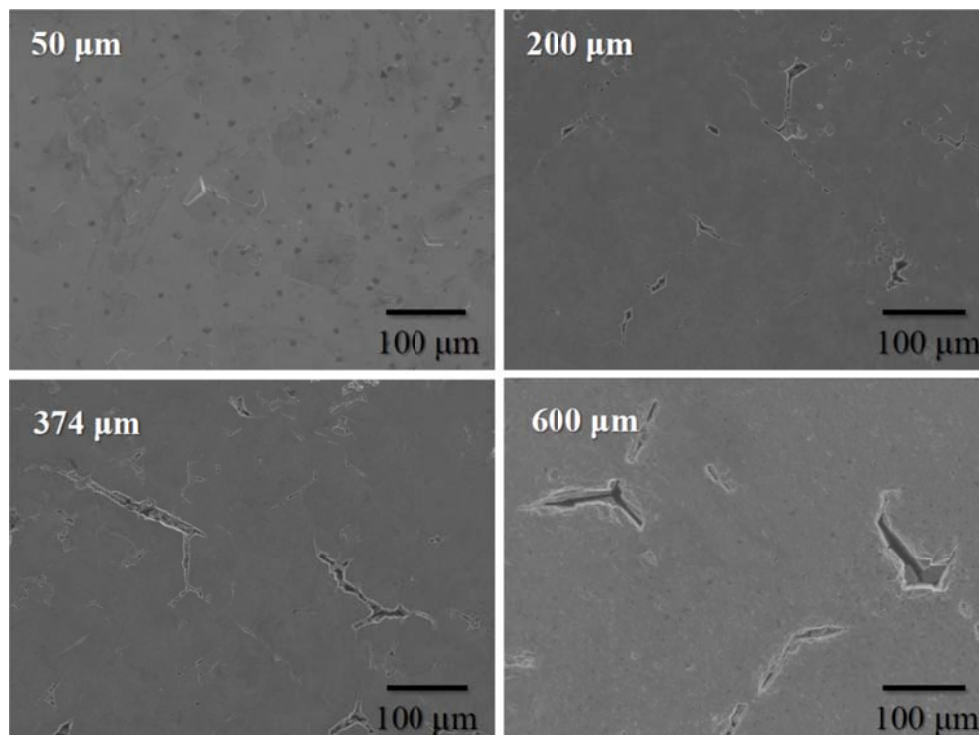


Figure 4. Representative SEM images of GaN crystals grown using polycrystalline GaN with different thickness.



Figure 5. Images of HVPE seed and different nutrients. (a) HVPE seed; (b) polycrystalline GaN; (c) Ga metal; (d) GaN powder.

As can be seen from Fig. 5, the HVPE seed is color-less and transparent before growth experiment. Unfortunately, however, resultant crystals usually suffered from the coloring, especially in the case of polycrystalline GaN, which tend to give darker color than Ga metal or GaN powder.

On the basis of our experiences with the acidic ammonothermal crystal growth of GaN, the coloring of GaN films is a very complex issue. Possible origins include the following:

- 1) Concentration of O species in the growth environment. O can come from the residual air in the autoclave, nutrient and mineralizer.
- 2) Concentrations of Ga vacancies and N vacancies in GaN films. This depends on the growth parameters such as temperature, pressure, nutrient and mineralizer.
- 3) Other impurities such as Fe, Ni coming from the autoclave.