

Influence of nitrogen oxide gas supply during ZnO film growth using high-temperature H₂O generated by a catalytic reaction

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Zinc oxide (ZnO) is highly useful for various applications. Recently, because of its large bandgap (3.37 eV at RT) and large exciton binding energy (60 meV)¹⁾, its application to optoelectronic devices such as light emitting diodes and laser diodes operating in the ultraviolet region has been intensively investigated²⁻⁹⁾. A wide variety of growth techniques, including molecular beam epitaxy (MBE)³⁻⁵⁾, pulsed laser deposition (PLD)⁶⁻⁷⁾, magnetron sputtering⁸⁾, and metal-organic chemical vapor deposition (MOCVD)⁹⁾, have been used for preparing ZnO films. We have developed a new CVD method for ZnO film growth using a reaction between an alkylzinc (DMZn) and high-energy H₂O generated by the Pt-catalyzed exothermic H₂-O₂ reaction. The resulting ZnO films grown on a-plane sapphire (a-Al₂O₃) substrates exhibited excellent electronic properties, that is, electron mobility was greater than 180 cm²/Vs¹⁰⁾. In addition, it became clear that the nitrogen oxide gas addition during the ZnO film growth influenced the film properties.¹¹⁾ In this study, we report the influence of the N₂O gas addition on the ZnO film properties.

The structure of the CVD apparatus and the growth procedure used in this study are the same as those shown in a previous paper¹⁰⁾, except for the addition of N₂O gas to the reaction zone. The ZnO films were directly grown on a-Al₂O₃ substrates at 773K for 60 min without any buffer layer. The thickness of the ZnO films characterized was 6–8 μm. Although the N₂O gas pressure varied from 3.2×10⁻³ Pa to 9.7×10⁻² Pa, all films showed an n-type character. The ZnO films grown with N₂O gas addition showed large crystalline facets greater than 10 μm in AFM images (Fig. 1 (B)-(D)), while that without N₂O showed small crystalline facets less than 1 μm (Fig. 1 (A)). The electron mobility of ZnO with N₂O (3.2×10⁻³ Pa) film at RT (300 K) was 217 cm²/Vs, while that of the ZnO without N₂O was 197 cm²/Vs. The mobility increases significantly with decreasing temperature to 100 K for all films. The mobility of the film with N₂O (3.2×10⁻³ Pa) increased to 1100 cm²/Vs at 100 K. The FWHM value of the ZnO (0002) ω-rocking curve of the film with N₂O (3.2×10⁻³ Pa) was 142 arcsec, while that of the film without N₂O was larger than 194 arcsec. The FWHM value of PL peak (3.36 eV) at 10K for the films with N₂O was 0.6-0.7 meV, while that of the peak for the film without N₂O was 0.9 meV. The nitrogen concentrations in all films with N₂O, however, were lower than the detection limit of SIMS (<2×10¹⁷ cm⁻³). From the improvement of the crystal orientation and the size of crystalline facets, nitrogen precursors generated by the reaction between the high-energy H₂O and the N₂O gas were speculated to play a role of surfactants on the growing-film surface.

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References

- 1) B. K. Meyer et al., *phys. stat. sol. (b)*, **241** (2004) 231.
- 2) A. Tsukazaki et al., *Nature Materials*, **4** (2005) 42.
- 3) M. Sano et al., *Jpn. J. Appl. Phys.*, **42** (2003) L1050.
- 4) H. Tampo et al., *Appl. Phys. Lett.*, **84** (2004) 4412.
- 5) K. Miyamoto et al., *J. Cryst. Growth*, **265** (2004) 34.
- 6) E. M. Kaidashev et al., *Appl. Phys. Lett.*, **82** (2003) 3901.
- 7) A. Ohtomo et al., *Semicond. Sci. Technol.*, **20** (2005) S1.
- 8) T. Minami, *Semicond. Sci. Technol.*, **20** (2005) S35.
- 9) J. Dai et al., *J. Cryst. Growth*, **290** (2006) 426.
- 10) K. Yasui et al., *MRS Symp. Proc.*, **1494** (2013) 127.
- 11) N. Yamaguchi et al., *MRS Symp. Proc.*, **1633** (2014) 61.

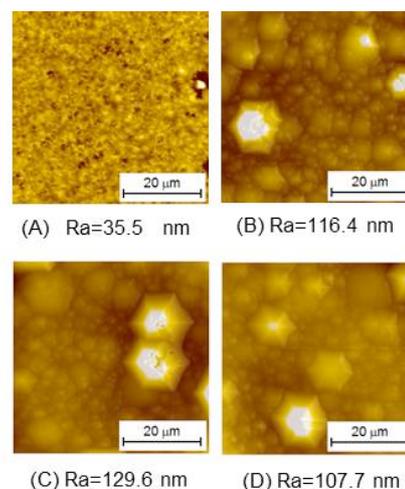


Fig. 1 AFM images of the ZnO films grown with (B)-(D) and without N₂O (A)