

## High-temperature H<sub>2</sub>O beam generated by a catalytic reaction for the growth of metal oxide thin films

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Metal oxide thin films such as ZnO, MgZnO, Ga<sub>2</sub>O<sub>3</sub>, indium zinc oxide (IZO), and indium gallium oxide (IGO) have received extensive attention due to their excellent properties, which has stimulated intensive research for optoelectronic and electronic device applications, such as light-emitting diodes, transparent conductive thin films for flat panel displays and solar cells, and thin film transistors. Although metal-organic chemical vapor deposition (MOCVD) has many advantages for industrial applications over methods such as molecular beam epitaxy and pulsed laser deposition, metal oxide thin films produced by MOCVD tend to be low quality, due to incomplete reaction between the metal and oxygen source gases in the gas phase. A new CVD method has been developed for metal oxide film growth using the hydrolytic reaction between metal-organic gas and high-temperature H<sub>2</sub>O generated by a Pt-catalyzed exothermic H<sub>2</sub>-O<sub>2</sub> reaction [1]. The resulting thin films, such as ZnO films grown on a-plane (11-20) sapphire (a-Al<sub>2</sub>O<sub>3</sub>) substrates, exhibited excellent optical and electronic properties [1]. The high-temperature H<sub>2</sub>O is generated in a catalytic cell and is effused through a de Laval nozzle to the reaction zone in the CVD chamber. The temperature and kinetic energy of the H<sub>2</sub>O beam are dependent on the nozzle structure and may therefore influence the film properties. In this study, the temperature of the high-energy H<sub>2</sub>O beam in the nozzle was estimated according to the theory of compressible flow for various opening angles of the de Laval nozzle, i.e., 50°, 60° and 90°, and the cluster size in the H<sub>2</sub>O beam was also estimated. The H<sub>2</sub>O beam temperature decreased with the opening angle, due to the difference in the cooling rates of the H<sub>2</sub>O beam through the adiabatic expansion process. The H<sub>2</sub>O beam temperature in the nozzle was simultaneously measured using a thermocouple. The measured temperatures were also low for the nozzle with a large opening angle, but were significantly higher than the theoretical values. The reasons for the difference were considered to be the vertical shock by insertion of the thermocouple and thermal radiation from the de Laval nozzle to the thermocouple. The H<sub>2</sub>O cluster size was estimated using Hagen's empirical formula [2]. For all H<sub>2</sub>O nozzles, the reduced scaling parameters ( $I^*$ ) were smaller than 200 and the mean cluster sizes ( $\langle n \rangle$ ) were estimated to be less than 1 with an initial gas temperature and pressure of 1273 K and  $1 \times 10^3$ – $2 \times 10^5$  Pa, respectively. Therefore, it was speculated that H<sub>2</sub>O clusters were not formed by the de Laval nozzle under the conditions used with our CVD method.

### References

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- [2] O. F. Hagen, Rev. Sci. Instrum. **63** (1992) 2374.

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