Influence of hydrogen addition on electric and optical properties of sputter-deposited aluminum-doped zinc oxide thin films

Ryoji Kan¹, Tadashi Iwata¹, Takashi Shibasaki¹, Masanori Shinohara², and Yoshinobu Matsuda²*

¹Graduate School of Science and Technology,
²Department of Electrical and Electronic Engineering,
Nagasaki University, Bunkyo-machi 1-14, Nagasaki 852-8521, Japan
*Tel: +81-95-819-2540, Fax: +81-95-819-2540, E-mail: ymat@nagasaki-u.ac.jp

Abstract

We have investigated the influence of hydrogen addition on AZO thin films. The electric conductivity of the film was improved when a very small amount of hydrogen (0.25%) was added, but was decreased by adding excessive amount of hydrogen. The optical transmittance and the deposition rate were almost constant for a small amount of hydrogen addition of not more than 0.5%, but decreased by adding excessive amount of hydrogen.

Introduction

For the last ten years, aluminum-doped zinc oxide (AZO) has been a focus of constant attention as a transparent conducting material that may take the place of tin-doped indium oxide (ITO). The inherent electric conductivity of AZO is lower than that of ITO, but the former has advantages over ITO in environment resistance and resource cost. To completely replace the ITO, a reproducible and highly-reliable fabrication process of good quality AZO thin films has to be developed. Recently, it has been reported that the electric conductivity of the AZO film was improved when hydrogen gas was added to working argon (Ar) gas.[1] Thus, we have investigated the effect of hydrogen addition on the electric conductivity, optical transmittance and deposition rate of the AZO thin films deposited by ICP assisted sputter-deposition.

Experimental Setup and Procedure

A 2 wt % aluminum-doped ZnO target was used in the experiment. The distance between the target and the substrate was set 80mm. An internal coil antenna of 10cm in diameter covered with insulator was arranged between the substrate and the target. Argon mixed with a small amount of hydrogen was used for the working gas. The total gas pressure was fixed at 30 mTorr, and the amount of hydrogen addition was changed by setting the preset partial pressure of hydrogen before each deposition. The fraction of hydrogen defined by $C_H = ([H_2]/([Ar]+[H_2]))$ was varied in steps 0, 0.25, 0.5 and 1%.
The ultimate pressure in the chamber was $3 \times 10^{-6}$ Torr or less. The flow rate of Ar was adjusted to 50 sccm by using a mass flow controller. The sputtering power (target power) and the ICP assist power (ICP-RF power) were fixed at 44 W and 200 W, respectively. Deposition time was 30 minutes for all samples. The film thickness, the electric conductivity and the optical transmittance of the deposited AZO thin films were measured by a stylus profiler (Mitsutoyo, SV-400), a four-point probe and an optical fiber spectrometer (ocean optics, HR4000CG) for each piece, respectively.

**Results**

Figures 1 and 2 show the change in resistivity, transmittance and deposition rate of ZnO films against the hydrogen mixture fraction. From Fig.1, it is found that the film conductivity was slightly improved; the minimum resistivity of $1.8 \times 10^{-3} \Omega \cdot \text{cm}$ was obtained at $C_{\text{H}}=0.25\%$. Excess addition of hydrogen more than 0.5\%, however, decreased the film conductivity. Next, the transmittance and the deposition rate is shown in Fig. 2. The optical transmittance and the deposition rate decrease at $C_{\text{H}}=1.0\%$.

**Conclusions**

By adding a small amount of hydrogen into the working argon gas, a slight improvement of film conductivity was confirmed in the ICP assisted sputter-deposition of aluminum doped ZnO. The lowest resistivity of $1.8 \times 10^{-3} \Omega \cdot \text{cm}$ was obtained at 0.25\% hydrogen addition. However, adding excessive amount of hydrogen of more than 0.5\% deteriorated all the conductivity, the transmittance and the deposition rate of deposited AZO thin films.

![Fig1 Variations of resistivity of AZO films deposited under Ar + H₂ Plasma for different H₂ fraction](image1)

![Fig2 Variations of optical transmittance and deposition rate of AZO films deposited under Ar + H₂ Plasma for different H₂ fraction](image2)

**References**