

MOCVD route to In_2O_3 thin films on SiO_2 substrates

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Indium oxide (In_2O_3) film is a technologically important transparent semiconducting oxide material and thus used in various fields such as photovoltaic devices, transparent windows in liquid crystal displays, sensors, and antireflection coatings [1–3].

In_2O_3 films can be prepared by a wide range of deposition techniques such as evaporation [4, 5], sputtering [6–8], spray pyrolysis [9], and ultrasonic spray chemical vapor deposition (CVD) [10]. Additionally, for the metal organic CVD (MOCVD) growth, various precursors such as trimethylindium ($\text{In}(\text{CH}_3)_3$) [11, 12], indium 2,2,6,6-tetramethyl-3,5-heptanedione ($\text{In}(\text{thd})_3$) [11, 13], and indium acetylacetonates ($\text{In}(\text{acac})_3$) [14, 15] have been employed. In this letter, we report the growth of In_2O_3 films on SiO_2 substrates using a simple reaction of a triethylindium (TEI) and oxygen (O_2) mixture. We investigate the effect of deposition temperature on the structural properties of thin films.

The SiO_2 layer was thermally grown on the Si (0 0 1) with a thickness of 60 nm. The SiO_2/Si substrate was cleaned in acetone and then rinsed by deionized water. A schematic diagram of the MOCVD reactor used in our experiments was previously reported [16]. High-purity Ar (99.999% purity) passed through the TEI bubbler and delivered TEI vapor to the reactor. The TEI bubbler was maintained at a temperature of 35 °C. The In_2O_3 film was synthesized by supplying O_2 and Ar carrier gases, respectively, with the flow rate of 5 sccm (standard cubic centimeters per minute) and 20 sccm. The deposition was carried out for 20 min and the substrate temperatures used in the present work were 200, 250, and 300 °C. The crystalline structure of the films was examined by X-ray diffraction (XRD) analysis on the θ - 2θ geometry using the $\text{CuK}\alpha$ line. The structural morphology was characterized by scanning electron microscopy (SEM).

Fig. 1 shows the XRD spectra of the as-deposited film at substrate temperatures in the range of 200–300 °C. Apart from the Si(0 0 4) diffraction peak, the θ - 2θ scan data from the deposits at 300 °C exhibit a strong and sharp peak at 30.58 °, corresponding to (2 2 2) diffraction peaks of cubic bixbyite In_2O_3 phase (JCPDS 44-1087). Fig. 1a indicates that although we observe a very weak (2 2 2) diffraction peak corresponding to the In_2O_3 structure, the film prepared at 200 °C is close to the amorphous phase due to the absence of a strong In_2O_3 diffraction peak. The (2 2 2) diffraction peak of

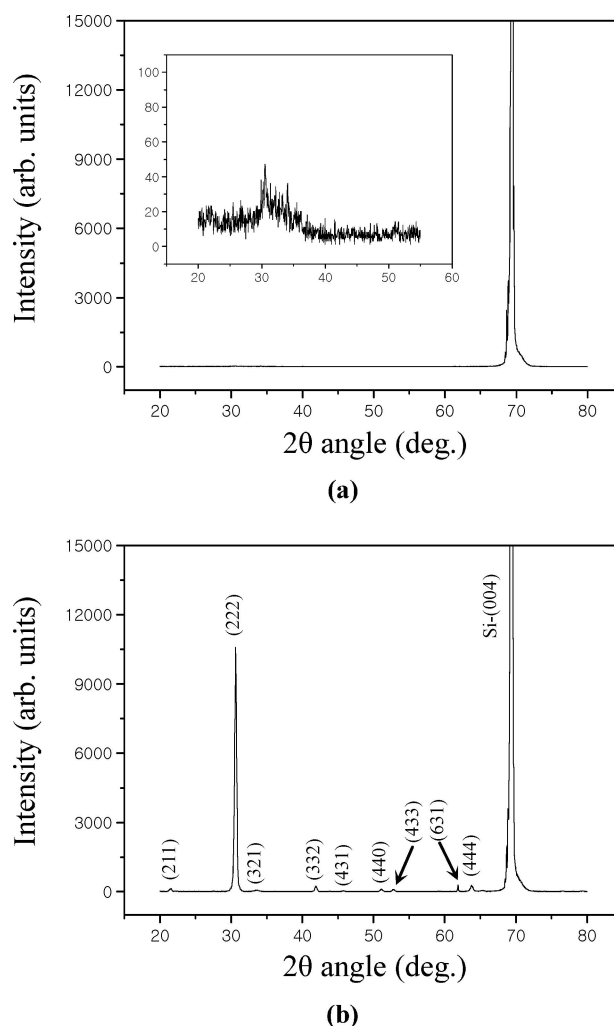
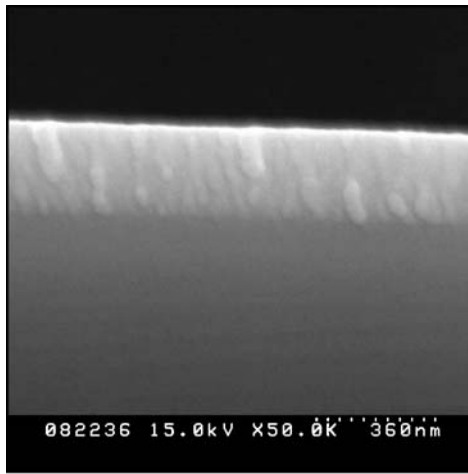


Figure 1 XRD spectra of as-deposited In_2O_3 films on SiO_2 substrates at a substrate temperature of (a) 200 °C and (b) 300 °C. The insets show the enlargement of the XRD patterns.

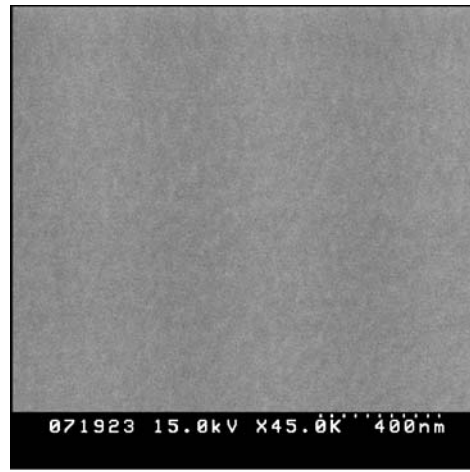
the films deposited at 300 °C is relatively strong compared to the neighbouring diffraction peaks (Fig. 1b), revealing a high degree of texturing in the (2 2 2) direction. The XRD spectrum of the 250 °C-deposited film is very similar to that of 300 °C-deposited film (not shown here).

Fig. 2 shows the cross-sectional SEM images, indicating that the thicknesses of the films deposited at 200, 250, and 300 °C, respectively, are 300, 595, and 1660 nm. Accordingly, the growth rate increases with increasing the substrate temperature, from 15 nm/min

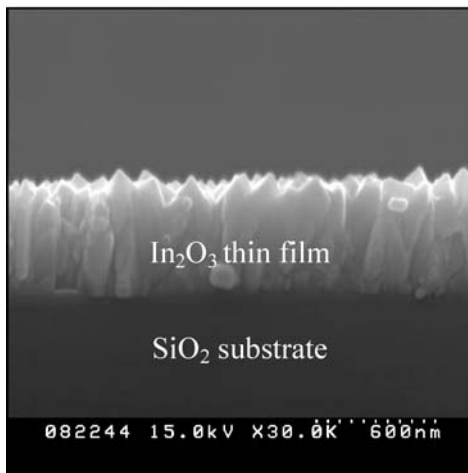
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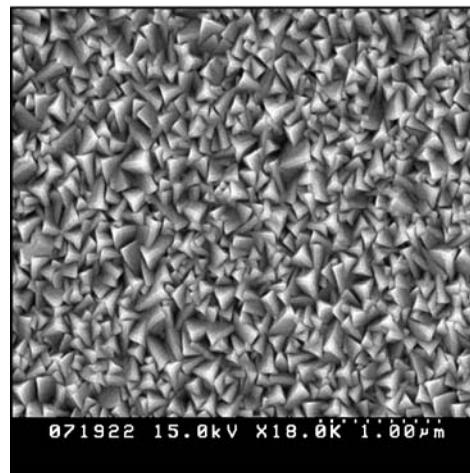
(a)



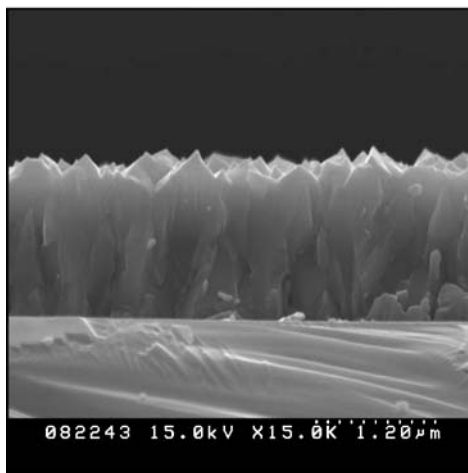
(a)



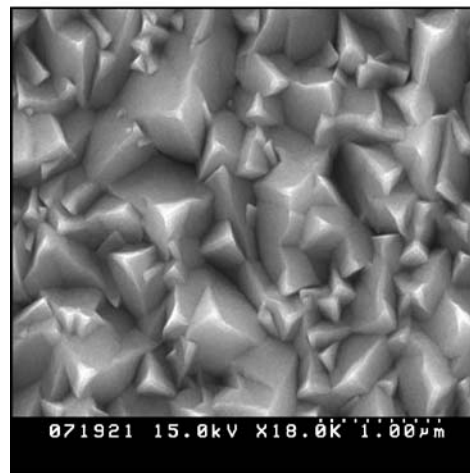
(b)



(b)



(c)



(c)

Figure 2 Cross-sectional SEM images of In_2O_3 films at a substrate temperature of (a) 200 °C, (b) 250 °C, and (c) 300 °C.

at 200 °C to 83 nm/min at 300 °C. Fig. 3 shows the plan-view SEM images of In_2O_3 thin films, revealing that the grain structures appear at substrate temperatures of 250–300 °C, while no grain structures are clearly observed on top of the film deposited at 200 °C. We surmise that at higher temperature, the atoms have enough diffusion activation energy to occupy the correct site in the crystal lattice and thus crystallization can be proceeded. In Figs 3b and c, the grain struc-

Figure 3 Plan-view SEM images of In_2O_3 films at a substrate temperature of (a) 200 °C, (b) 250 °C, and (c) 300 °C.

tures with smooth facets and a sharp tip are shown on the surface of the films. From their geometrical shapes and the XRD analysis, we surmise that the tip direction is along the [2 2 2] direction. The average grain size increases with increasing the substrate temperature in the range of 250–300 °C.

In summary, we have demonstrated the deposition of highly-oriented In_2O_3 thin films on SiO_2 substrates using the TEI as a precursor in the presence of

oxygen. The growth rate of In_2O_3 thin films increases with increasing substrate temperature at 200–300 °C. We reveal that the film deposited at 300 °C is a cubic structure with the predominant (2 2 2) orientation.

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References

1. K. L. CHOPRA, S. MAJOR and D. K. PANDYA, *Thin Solid Films* **102** (1983) 1.
2. K. HARA, T. HORIGUCHI, T. KINOSHITA, K. SAYAMA, H. SOGIHARA and H. ARAKAWA, *Sol. Energy Mater. Sol. Cells* **64** (2000) 115.
3. H. YAMAURA, T. JINKAWA, J. TAMAKI, K. MORIGA, N. MIURA, N. YAMAZOE, *Sensor. Actuat. B* **36** (1996) 325.
4. S. NASEEM, I. A. RAUF and K. HUSSAIN, *Thin Solid Films* **156** (1988) 161.
5. S. MURANAKA, *Thin Solid Films* **221** (1992) 1.
6. S. KASIVISWANATHAN and G. RANGARAJAN, *J. Appl. Phys.* **75** (1994) 2572.
7. A. S. RYZHIKOV, R. B. VASILIEV, M. N. RUMYANTSEVA, L. I. RYABOVA, G. A. DOSOVITSKY, A. M. GILMUTDINOV, V. F. KOZLOVSKY and A. M. GASKOV, *Mater. Sci. Eng. B* **96** (2002) 268.
8. C. GRIVAS, D. S. GILL, S. MAILIS, L. BOUTSIKARIS and N. A. VAINOS, *Appl. Phys. A* **66** (1998) 201.
9. J. J. PRINCE, S. RAMAMURTHY, B. SUBRAMANIAN, C. SANJEEVIRAJA and M. JAYACHANDRAN, *J. Cryst. Growth* **240** (2002) 142.
10. M. GIRTAN, *Surf. Coat. Technol.* **184** (2004) 219.
11. S. REICH, H. SUHR and B. WAIMER, *Thin Solid Films* **189** (1990) 293.
12. T. MARUYAMA and T. KITAMURA, *Jpn. J. Appl. Phys.* **28** (1989) L1096.
13. W. KERN, *Thin Solid Films* **29** (1975) 155.
14. T. MARUYAMA and K. FUKUI, *J. Appl. Phys.* **70** (1991) 3848.
15. L. A. RYBOVA, V. S. SALUN and I. A. SERBINOV, *Thin Solid Films* **92** (1982) 327.
16. H. W. KIM and N. H. KIM, *Appl. Surf. Sci.* **230** (2004) 301.

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