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Enhanced preferential orientation and electrical property of fluorine-doped SnO_2 thin films via barrier layer

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Polycrystalline fluorine-doped SnO_2 thin films with SiC_xO_y or $Si_xSn_yO_2$ barrier layer are deposited on glass substrates by atmospheric pressure chemical vapor deposition (APCVD) method. The effect of barrier layer on structure and electrical property of FTO films was investigated. Results show that the inserting of barrier layer, especially the SiC_xO_y layer, has led to the improved crystallinity and the enhanced preferential orientation along the (2 0 0) crystallographic plane. SnO_2:F/SiC_xO_y/Glass films with larger grain size and a columnar growth structure exhibited lower resistivity ($\sim 4.9 \times 10^{-4}$), higher reflectance in the mid-far-infrared region ($\sim$80%) and lower emissivity (0.16), while maintaining high transmittance in the visible range. The SiC_xO_y film has therefore been considered as a more ideal potential barrier layer for FTO thin film production.

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1. Introduction

Polycrystalline fluorine-doped SnO_2 (FTO) thin films are of increasing importance for modern information and energy applications, such as displays, photovoltaics, and architectural, due to its outstanding optical transparency and high electrical conductivity [1–5]. Atmospheric pressure chemical vapor deposition (APCVD) has been widely considered to be a promising process method for large-scale production of the FTO film due to its advantages, such as reliable reproducibility and low cost [2,6]. The electrical and optical properties of FTO thin film strongly depend on its underlying microstructure, preferential orientation (i.e. texture), and morphology [7]. It has been reported that the enhanced (2 0 0) preferential crystallographic orientation decreases the resistivity of SnO_2 thin films to $\sim 10^{-3}\ \Omega$ cm [8]. At present, the control of film structural ordering and morphology has been regarded as a crucial step toward the realization of high performance FTO films.

The structure and morphology of FTO films have been successfully tailored via adjusting the growth conditions, such as precursors, temperature, substrate, and barrier layer, etc [7–9]. Particularly, the functional performance of FTO films at high temperature is often hampered by the lattice mismatch between the glass substrate and FTO layer, and the diffusion of alkali metal ions from hot glass substrate into the FTO layer [10]. To reduce such unexpected influence, a barrier layer, such as TiO_2 [11] and SiO_2 [12], has been introduced for the FTO production. However, a scientific understanding of the mechanism for the intrinsic effect of barrier layer on the film morphology and functional properties are rarely studied.

This study therefore focuses on the enhanced preferential orientation and the electro-optical properties of FTO thin films with barrier layer deposited via the APCVD method on an industrial production line. SiC_xO_y and $Si_xSn_yO_2$ barrier layers were selected due to their similar major constituent to the glass substrate. The modification of the structure and surface morphology was also investigated.

2. Experimental

SnO_2:F, SnO_2:F/SiC_xO_y and SnO_2:F/$Si_xSn_yO_2$ films were deposited on glass substrate by APCVD method on an industrial production line. The temperature of the glass surface was maintained at $\sim$690 °C, and the deposition time was set at 13 s. Silane (SiH_4) and ethane (C_2H_4) were used as precursors of SiC_xO_y layer, $\sim$60 nm in thickness, deposited in the tin bath. The atomic ratio of the SiC_xO_y layer was $\sim$1:2:5 as reported before [13]. Metal organic monobutyltin trichloride (MBTC, $C_4H_9SnCl_3$, 99.0 wt%) and

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tetraethoxysilane (TEOS, $C_8H_{20}O_4Si$, 99.0 wt%) were used as precursors of the $Si_xSn_yO_2$ layer, ~60 nm in thickness, deposited in the annealing kiln. The atomic ratio of the $Si_xSn_yO_2$ layer was ~2:3:10. Subsequently, SnO_2:F thin film, thickness of ~350 nm, was deposited using MBTC and trifluoro acetic acid (TFA, CF_3COOH) as precursors.

The crystalline phase of the films prepared was determined by a X-ray diffraction (XRD, Thermo electron ARL X'TRA). Seven main diffraction peaks, (1 1 0), (1 0 1), (2 0 0), (2 1 1), (2 2 0), (3 1 0) and (3 0 1), were taken into account. The quantitative texture analysis was carried out from the $K_{\alpha 1}$ component of each diffraction peak in the framework of the Harris method [14]: the texture coefficients C_{hkl} for each (hkl) crystallographic direction and the degree of preferred orientation σ were defined respectively as follows [12,14]:

$$C_{hkl} = \frac{(I_{hkl}/I_{0,hkl})}{(1/N)\sum^N (I_{hkl}/I_{0,hkl})} \quad \text{and} \quad \sigma = \frac{\sqrt{\sum^N (C_{hkl}-1)^2}}{\sqrt{N}}$$

where $N=7$ is the peak number, I_{hkl} and $I_{0,hkl}$ are the intensity of the (hkl) Bragg reflection for the studied sample and randomly oriented sample respectively, as given from the International Centre for Diffraction Data (ICDD). In principle, for films with randomly oriented grains, the texture coefficients and degree of preferred orientation equal 1 and 0, respectively. In contrast, for the films with perfectly oriented grains along the (hkl) direction, the texture coefficient equals N (for (hkl) planes) or 0 (for other planes), and the degree of preferred orientation is $\sqrt{N-1}$. The film surface morphology was examined using a scanning electron microscope (SEM, SU-70, Hitachi). The cross-sectional samples of the as-deposited films were prepared by focused ion beam (FIB, Quanta 3D, FEI) technique. The atomic scale micrographs of thin films were obtained by field emission transmission electron microscopy (TEM, FEI tecnai G2 F20). The carrier concentration, Hall mobility and resistivity, were examined by Hall-effect measurement (BIO-RAD, MODEL HL5500). FTIR spectrophotometer (Bruker Tensor 27) was employed to obtain the reflectance in the middle-far-range infrared region, while the transmittance in the visible range was obtained via ellipsometer (GES_5E Semilab).

3. Results and discussion

3.1. Structure and morphology

As shown in XRD patterns (Fig. 1a–c), all films prepared in this work exhibit a polycrystalline rutile structure consistent with the characteristics of the SnO_2 structure (P42/mnm (1 3 6)) in JCPDS card 41–1445. For FTO films with barrier layer, the intensity of diffraction peaks increase, and the peak width decreases, indicating the enhanced crystallinity. In addition, the degree of preferred orientation for SnO_2:F, SnO_2:F/SiC_xO_y and SnO_2:F/$Si_xSn_yO_2$ films are calculated as 0.93, 1.31 and 1.21, respectively (Fig. 1d). Therefore, the preferred orientation increases by ~40% due to the SiC_xO_y barrier layer. Furthermore, the calculated texture coefficients suggest that the (2 0 0) crystallographic orientation is dominant for all three films, and a dramatic enhancement is observed for the films with barrier layer. In contrast, (3 0 1) and (2 1 1) crystallographic orientations are weakened. The strong texture along the (2 0 0) orientation has been reported to correspond to the best balance between the electrical resistivity and optical transmittance [8]. The texture coefficient for SnO_2:F/SiC_xO_y films varies with a higher magnitude, in accord with the degree of preferred

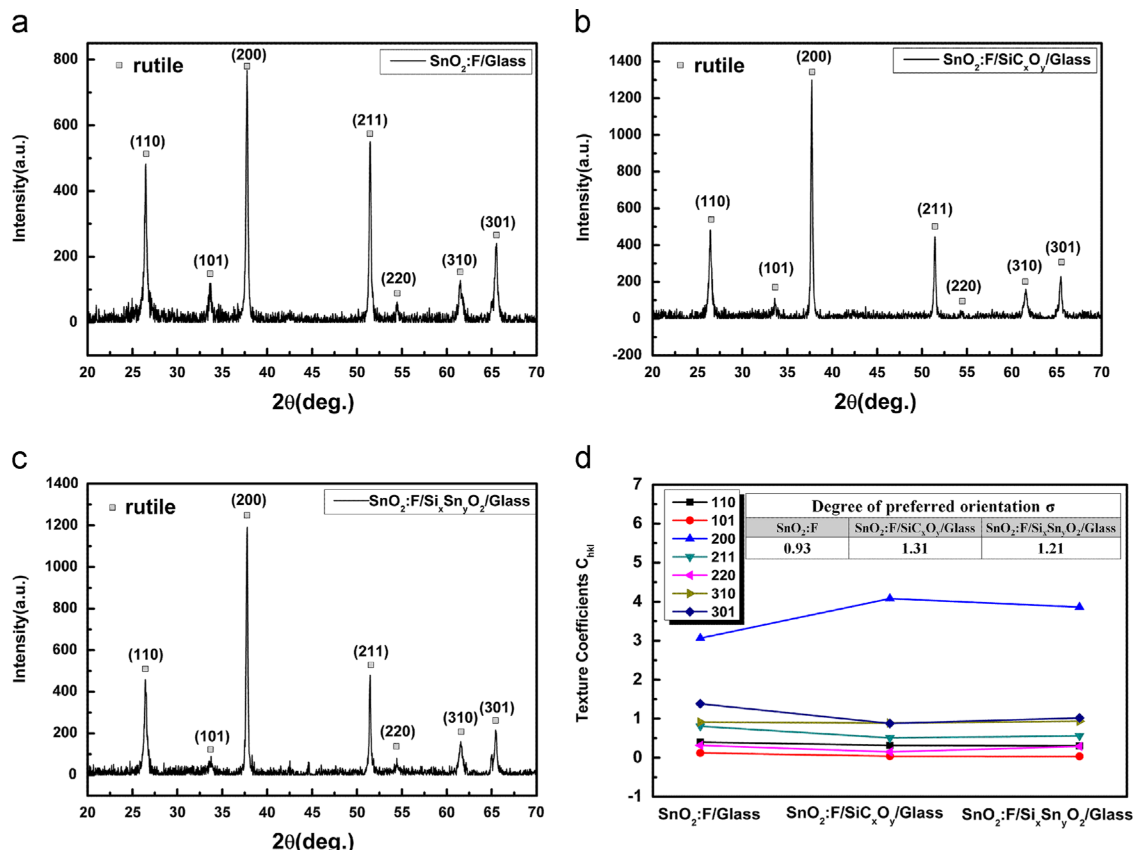

Fig. 1. XRD diffraction patterns of (a) SnO_2:F, (b) SnO_2:F/SiC_xO_y and (c) SnO_2:F/$Si_xSn_yO_2$ films on glass substrate, (d) the texture coefficients variation of the three films with the inset table showing the degree of preferred orientation.

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