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EFFECTS OF DEPOSITION PARAMETERS AND OXYGEN ADDITION ON PROPERTIES OF SPUTTERED INDIUM TIN OXIDE FILMS

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Abstract

Indium tin oxide (ITO) films were sputtered on corning glass substrate. Oxygen admixture and sputtering deposition parameters were optimized to obtain the highest transparency as well as lowest resistivity. Structural, electrical and optical properties of the films were then examined. Increasing deposition rate and film thickness changed the crystallographic orientation from (222) to (400) and (440), as well as higher surface roughness. It was necessary to apply substrate heating during reposition to get films with better crystallinity. The lowest resistivity of 5.36 x 10^{-4} Ω•cm was obtained at 750 nm film thickness. The films' resistivity was increased by addition of oxygen up to 2% in the argon sputtering gas. All films showed over 85% transmittance in the visible wavelength range, possible for applications in photovoltaic and display devices.

Abstrak

Efek Parameter Deposisi dan Penambahan Oksigen terhadap Sifat-sifat Lapisan Tipis Indium Timah Oksida Hasil *Sputtering*. Lapisan tipis indium timah oksida (ITO) dideposisikan pada substrat gelas corning dengan metode *sputtering* menggunakan gas argon. Parameter deposisi dan penambahan oksigen dalam gas *sputtering* dioptimasi untuk mendapatkan tingkat transparansi lapisan tertinggi dan resistivitas listrik terendah melalui pengamatan struktur, sifat listrik dan sifat optik. Peningkatan laju deposisi dan ketebalan lapisan menghasilkan perubahan orientasi kristalografi dari (222) ke (400) dan (440), serta peningkatan kekasaran permukaan lapisan. Pemanasan substrat sangat diperlukan untuk mendapatkan lapisan tipis dengan kristalinitas yang lebih baik. Nilai resistivitas lapisan cenderung naik dengan penambahan oksigen hingga 2% dalam gas *sputtering*, dengan nilai resistivitas terendah sebesar 5.36 x 10^{-4} Ω•cm dapat dicapai pada ketebalan lapisan 750 nm. Semua lapisan tipis yang dideposisi pada penelitian ini menunjukkan transparansi lebih dari 85% sehingga memungkinkan untuk diaplikasikan pada divais fotovoltaik dan *display*.

Keywords: characterization, indium semiconductor, sputtering, thin films, tin oxide

1. Introduction

Indium tin oxide (ITO) thin films are by far the most popular transparent conducting oxide (TCO) materials, widely used as electrodes in display devices and photovoltaic cells [1]. It has many advantageous properties such as low resistivity, high transparency in the visible light wavelength, and long-term stability [2]. Several methods have been employed successfully to deposit ITO films. Amongst the methods, sputtering is the most widely used due to its ability to produce a large area of films while maintaining excellent properties and low production cost [3-5].

Although many good properties are attributed with ITO, the growth mechanisms leading to low resistivity and high homogeneity ITO films during deposition with magnetron sputtering are still not fully understood [6]. Conductivity of ITO films is believed to be mainly controlled by the presence of dopant (SnO_2), carrier density, and concentration of oxygen vacancy in the film, which are highly dependent on deposition and post-deposition conditions [5,7]. Variation of deposition parameters and the presence of oxygen admixture during sputtering deposition may affect the properties of resulted films.

In this paper, we report the deposition of ITO films using RF magnetron sputtering from ITO ceramic target. Parameters for deposition conditions were varied and optimized in order to achieve low resistivity and high transparency at an optimum thickness level for application in display panels and photovoltaic cells.

2. Experiment

ITO thin films were deposited using RF magnetron sputtering (13.56 MHz) on corning glass 7509 substrate from a Ø 70 mm ITO ceramic target composed of In_2O_3–10% SnO_2. The substrate-target distance was kept at 50 mm. Base pressure for the deposition was 0.01 mTorr while the working pressure was 25 mTorr using 99.999% Ar as sputtering gas. Corning glass substrates, sliced into 10x10mm, were cleaned in organic solvent before deposition. Among the varied deposition parameters were RF power, deposition time, substrate temperature, and oxygen admixture in the argon sputtering gas. To observe the effects of oxygen admixture, the film thickness was kept at 150 nm by optimizing deposition parameters at 200 ^oC substrate temperature. 0-2% oxygen was introduced into the sputtering chamber through a mass flow controller.

X-ray diffractometer (XRD, RINT-Rigaku) with a Cu-Kα radiation was used for phase identification and analyzing crystallographic structure. The microstructure was studied by Scanning Electron Microscope (SEM Hitachi S-4100) while the thickness was measured using α-step surface profilometer (Sloan Dektak III). The electrical resistance of the thin films was measured using a four-point probe (CMT-SR1000N, Changmin Co.) while the optical transmittance of the thin films was measured with a UV-Vis-NIR Spectrophotometer (CARY 5G, Varian) in the wavelength range of 300–1000 nm. Surface texture and roughness (root mean square-RMS) value were examined by atomic force microscope (AFM).

3. Results and Discussion

Film thickness is mainly controlled by deposition time and RF power (Figure 1). When deposited using pure Ar sputtering gas, the deposition rate was measured in the range of 18–39 nm/min. There is no thickness restriction for application of TCO in photovoltaic cells, but it has been a common requirement to limit the thickness to 150–200 nm for application in display devices.

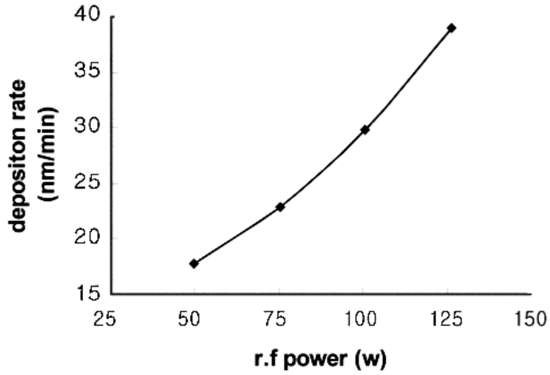

Figure 1. Deposition Rate vs RF Power of Sputtered ITO Films

Structural properties. The XRD analysis shows the films were mostly dominated by (222) (400) (440) (622) orientations, which reflects the crystalline structure of cubic In_2O_3. There was also (211) peak appeared in the spectra, but we could not confirm the origin of the peak as it can be either from In_2O_3 or silicon rubber used in the specimen holder. It also shows that substrate temperature plays an important role to produce films with higher crystallinity. Films deposited at substrate without intentional heating showed poor crystallinity, especially at low RF power and shorter deposition time (50 W-5 min), which resulted in almost completely amorphous films. A similar result has been observed by Park, *et al.*, which is attributed to lack of substrate temperature for crystallization as well as required film thickness for proper XRD analysis [8].

Figure 2(a) shows the XRD pattern of films deposited at 100W RF power and 200^oC substrate temperature. As deposition time and RF power increased, the crystal orientation shifted from (222) to (400), (440) and (622), with (400) and (440) peaks getting stronger as film thickness increased except at 125 W RF power and 20 min deposition time where those peaks slightly decreased again. It is necessary to apply substrate heating during deposition to obtain films with better crystallinity.

The same trend was found when the deposition parameters were optimized to obtain films with 150 nm thickness. Figure 2(b) shows that with 150 nm film thickness, as RF power increased, the (222) peak decreased and the crystal orientation shifted towards (440). Figure 2(c) summarizes the XRD spectra at various oxygen admixtures. The addition of small amount of oxygen in the sputtering gas did not affect the deposition rate and structure of the films in general. The crystal orientation shifted from (222) towards (440) plane as was also observed at films with the same thickness deposited using pure argon. Baia *et al.* reported that higher percentage oxygen admixture (more than 10%) during sputtering promotes the growth of (440) peak and reduce the intensity of (400) peaks [9].

Observation by SEM, as given in Figure 3 (a) and (b), reveals that deposition at higher deposition rate and films thickness as well as higher substrate temperature

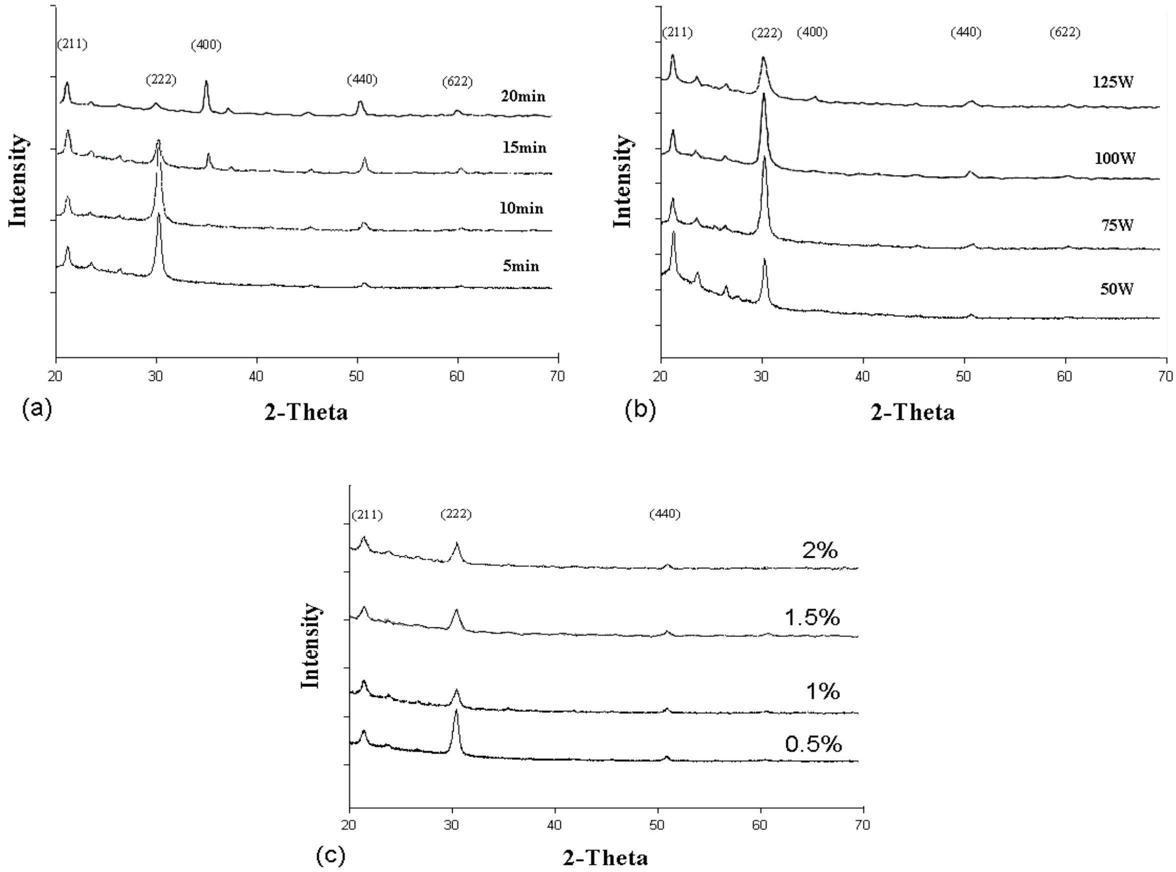

Figure 2. XRD Spectra of ITO Films Varied with (a) Deposition Time, (b) RF Power, and (c) Oxygen Admixture

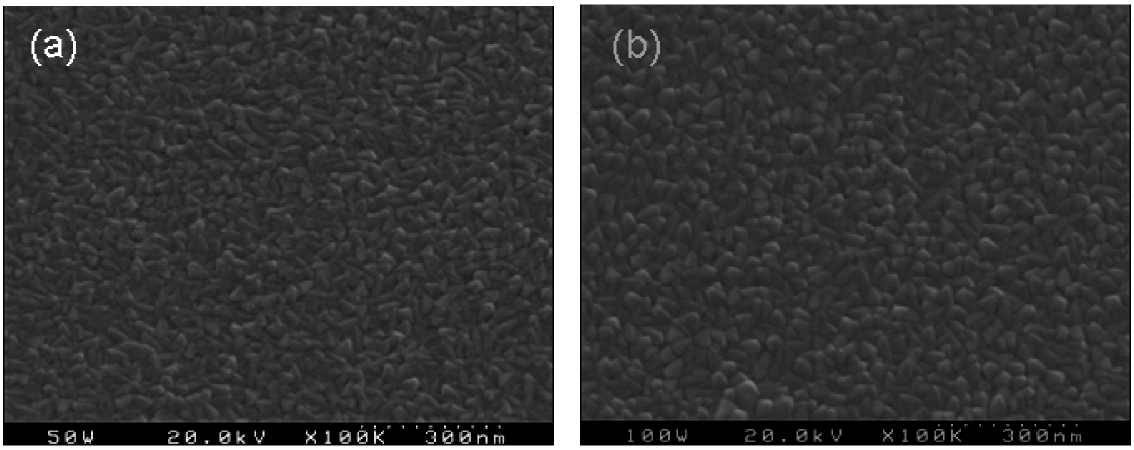

Figure 3. SEM Images of Films Deposited at (a) 50 Watts and (b) 100 Watts

lead to films with larger grain size. As reported by Thilakan *et al*., the (400) crystal orientation is believed to promote larger grains as films grown exactly oriented at normal to the film, thus having better grain uniformity [10]. This structure is preferred to obtain films with lower resistivity because of less grain boundaries associated with electron scattering. Films having (222) orientations tend to have more random growth behavior in every direction, resulting in smaller grains.

The AFM observation result, shown in Figure 4, confirmed the surface morphology revealed by SEM as surface roughness of the films increased for longer deposition time and higher RF power. As deposition time increased, the lowest root mean square (RMS) value increased from 1.627 nm at 5 minutes to 14.899 nm at 20 minutes while at a constant thickness, as RF power increased, the RMS value increased slightly from 4.295 nm at 50W to 5.053 nm at 125W RF power. The surface roughness value increased for higher film thickness and higher RF power was applied. Apart from dominant (400) crystal orientation, this may be due to higher bombardment energy during film deposition to the substrate associated with higher RF power. Higher surface roughness can be beneficial for photovoltaic cell applications that require a light trapping scheme such as in silicon thin film solar cells.

Electrical properties. The minimum films resistivity obtained without substrate heating was 3.1 x 10^{-3} Ω•cm, while films deposited at 200 ^{o}C substrate temperature had minimum 5.36 x 10^{-4} Ω•cm. As given in the Figure 5(a), increasing deposition time and RF power reduced the resistivity of the films. The reason behind this is mainly due to the change in structure and crystallinity of the films. Increasing deposition time and RF power produced films with larger grains and, therefore, fewer grain boundaries, which can hinder the electron movement and increase the films resistivity. Crystallinity might also play important part in determining the resistivity of the films, for example, the presence of (400) crystal orientation yields larger grains, which reduced the resistivity. It was also revealed that at 50W RF power deposition the resistivity decreased dramatically when the deposition time increased from 5 to 10 minutes and decreased linearly further afterwards. Film deposited at 50 W and 5 minutes deposition time had a low thickness and poor crystallinity, which may contribute to increase electron scatterings at the film's surface and reduce electrons' mobility in the film.

ITO is an *n*-type semiconductor where the conductivity electrons arise from oxygen vacancies or from Sn^{4+}ions on In^{3+} sites. Apparently the resistivity of the films was increased by addition of oxygen as shown in Figure 5(b). Oxygen admixture reduces the conductivity by

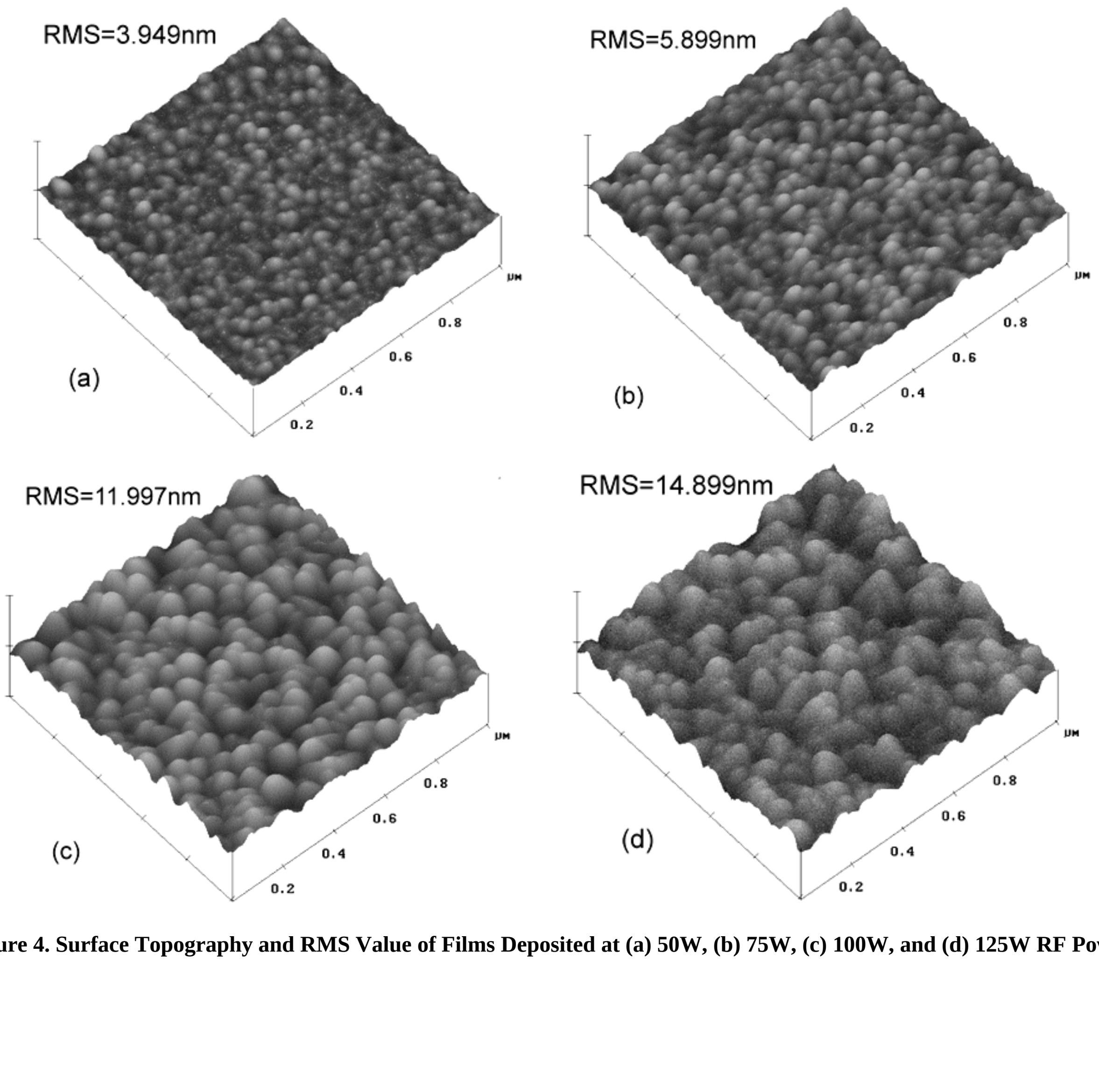

Figure 4. Surface Topography and RMS Value of Films Deposited at (a) 50W, (b) 75W, (c) 100W, and (d) 125W RF Power

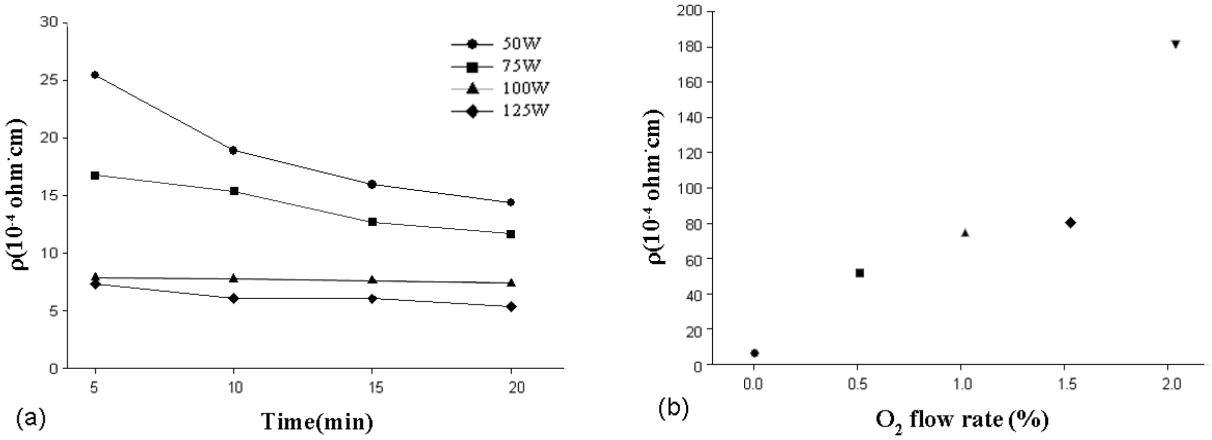

Figure 5. Resistivity of ITO Films Varied with (a) Deposition Time and RF Power, and (b) Oxygen Admixture

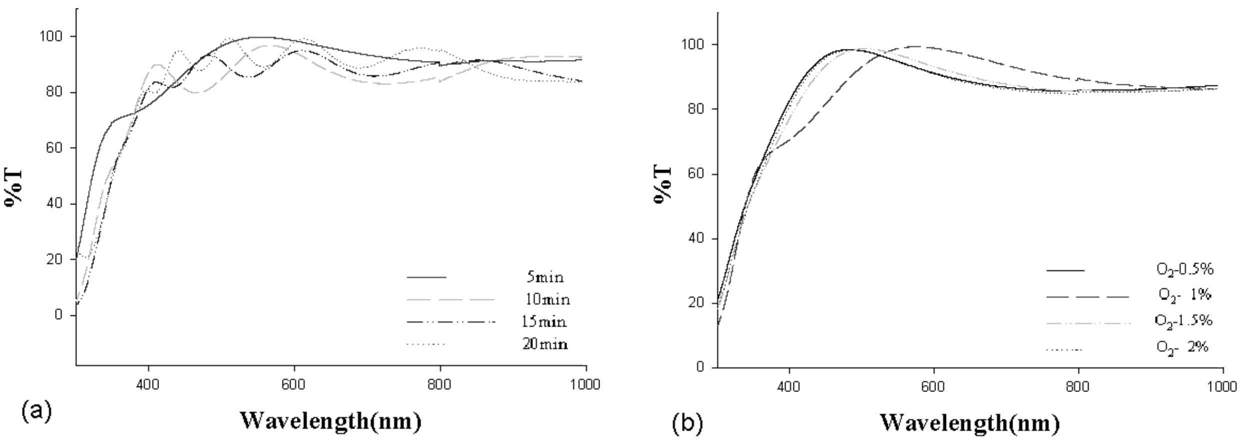

Figure 6. Transmittance Spectra of ITO Films at (a) Various Deposition Time, and (b) Oxygen Admixture

oxygen incorporation into the lattice and filing the oxygen vacancies or by deactivating the Sn-donor by forming $Sn^{+}O^{-}$ complexes, thus limiting carrier mobility and increases films' conductivity [11].

Optical properties. The transmittance of the films was characterized by UV-Vis-NIR. All films showed high transmittance over 85% in average, as shown in Figure 6(a). The transmittance was mainly affected by the degree of light scattering in the film, which depend on thickness and structure of the films. Films with lower thickness have higher transmittance as less scattering happened. As film thickness increased, the transmittance curves shifted towards higher wavelength (to the right) and reduced the overall transmittance. When the films thickness was kept constant at 150nm, the transmittance showed an average value of 90% or more. Figure 6(b) shows optical transmittance of the films deposited with oxygen admixture. There are no significant differences in the transmittance properties by adding small oxygen. All of the films had over 90% transmittance commonly required for display devices applications.

4. Conclusions

ITO thin films were deposited at several deposition rates using RF magnetron sputtering using pure argon or argon with oxygen admixture generated plasma. As thickness and deposition rate increased, preferred crystal orientation changed from (222) to (400) and (440). At 150 nm thickness, crystal orientation changed from (222) to (440) as deposition rate increases. Grain size and RMS value also increased according to the film thickness. To obtain films with better crystallinity, it is necessary to apply substrate heating during deposition. With optimized growth condition, the lowest film's resistivity was 5.36 x $10^{-4}\Omega\bullet$cm. Apart from improved crystallinity to the (440) orientation, the addition of small oxygen admixture did not improve the film properties in general, as the addition of oxygen reduces the oxygen vacancy so that the carrier mobility decreases and, consequently, resistivity increases. The presence of (400) crystal orientation is preferable to have a lower films resistivity. All of the films showed more than 85% optical transmittance, which increased to more than 90% at 150 nm film thickness.

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