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EFFECT OF OXYGEN FLOW ON ELECTRICAL AND OPTICAL PROPERTIES OF ITO FILMS SYNTHESIZED BY MAGNETRON SPUTTERING METHOD

Abstract

The tin-doped indium oxide thin films were synthesized by DC magnetron sputtering on the surface of polished silicon samples and glass slides in a mixed argon-oxygen atmosphere. The other deposition parameters: operating pressure, magnetron power and substrate rotation speed were kept constant. Thickness and density of thin films were measured by X-ray Reflectometry. The effects of oxygen flow rate and substrate temperature on the optical and electrical properties were investigated. The electrical properties (resistivity, Hall mobility and charge concentration) of the thin films were measured by the Van der Pauw method using the Hall effect. The minimum value of resistivity 0.52×10^{-3} Ohm·cm, and maximum charge mobility 28 $cm^2V^{-1}s^{-1}$ was achieved at an oxygen proportional gas mixture of 2.6% (0.71 sccm). The transmission spectra of the films were measured in the wavelength range from 300 to 1100 nm. The transmittance of all films exceeds 75% in the visible and near-infrared spectral ranges. It was found that increasing the oxygen flow rate and heating of the substrate up to optimal value 150°C led to an increase in the crystallinity of the films and, consequently, to an increase in the Hall mobility and the transmittance.

Key words: silicon solar cells, crystallization, magnetron sputtering, thin films, indium oxide.

Introduction

Solar energy has become a promising alternative to conventional energy sources due to its abundance and environmental friendliness. However, the widespread use of solar modules as energy sources is limited due to low efficiency and aging effect on efficiency. Scientists around the world are working to increase efficiency and decrease Levelized Cost of Energy. In recent years, record efficiency levels for silicon solar cells have been achieved thanks to Tunnel Oxide Passivated Contact and heterojunction structures. In both technologies, transparent conductive oxides play an important role, which at the same time act as anti-reflective coatings and conductors from the active area to the metal fingers.

Indium tin oxide (ITO) is a transparent conductive oxide with excellent electrical conductivity and optical properties that has attracted considerable attention in various industries including electronics [1], optoelectronics [2], and solar energy [3]. ITO's excellent electrical conductivity and low surface resistivity (~10^{-4} Ohm·cm) promotes efficient collection of electrical charges generated in the solar cell by light absorption. ITO films have high transparency in the visible spectrum (>85%), which allows light to pass through them with minimal absorption and reflection. Despite its high transparency, ITO has optical absorption in various wavelength intervals, especially in the ultraviolet (UV) range. This absorption can lead to reduced transparency and affect the performance of ITO-based devices in certain applications. The refractive index of ITO varies within from 1.8 to 2.0 in the visible spectral range depending on the composition of the film and the method of deposition [1–3]. For example, it is well known that by increasing substrate temperature of ITO films [4, 5, 6], the structure of films can be changed and hence better optoelectronical properties can be obtained in comparison to those of thin films deposited onto unheated substrates. Essential role in the electrical and optical properties of ITO films play such parameters as deposition methods, Sn doping, oxygen partial pressure, crystallinity, substrate features. In our work, the impact of oxygen partial pressure on the electrical and optical properties of ITO thin films was studied. During the study of the influence of temperature on the structure, conductivity, transmittance, and stability of ITO films, it was found that at a substrate temperature of 150°C the films have the highest optical and electrical properties.

Materials and Methods

Thin ITO films were synthesized by DC-magnetron sputtering system using cylindrical target with composition 90 wt.% In_2O_3 and 10 wt.% SnO_2. Polished n-type CZ monocrystalline silicon (100) and optical glass slides were used as a substrate. Glass substrates were cleaned in an HNO_3:H_2O_2: H_2O solution. Surface of the silicon samples was cleaned by the standard Radio Corporation of America method [7]. ITO films were deposited in a MAGNA-TM200-1 installation. Before the sputtering, chamber was pumped down to the base pressure of $5 \cdot 10^{-4}$ Pa. Once the base pressure was achieved, deposition was carried out in an atmosphere of argon and oxygen.

Sputtering was carried out at different oxygen concentration from 1.6 to 3.2% in an argon atmosphere and at a pressure of 0.6 Pa by varying flow rate. Depositions were performed by a power of 250W, deposition time was 120 s. The film thicknesses were determined by X-ray reflectometry (XRR) method using a Complex Ray C6 setup. Hall Effect measurements of ITO thin films were performed using van der Pauw method and Keithley 2400 SourceMeter. The reflection and transmission parameters were measured on the Evolution 300 UV-Vis spectrophotometer in the wavelength range from 300 to 1100 nm.

Results

Figure 1 show the dependence of the electrical properties of ITO films, such as charge carriers' concentration, resistivity, and Hall mobility on the oxygen/argon gas-flow ratio. The oxygen flow in the sputtering process has an essential effect on the crystallinity of the ITO film structure [8]. With increasing oxygen flow, the crystal structure of ITO films are becoming more organized and grain boundaries more distinct. The improved crystal structure and hence reduced scattering at the grain boundaries provide high charge carriers mobility and lower resistivity values. In the substitution process of tin (Sn^{4+}) atoms instead of indium (In^{3+}) atoms, the dopants are activated. Acting as electron donors, the doped Sn^{4+} ions provide the release of free electrons, thereby increasing the conductivity of the films [9–11]. Nevertheless, tin in the amorphous state is activated inefficiently and free carriers are formed mainly due to vacancy-like oxygen defects [12, 13]. Consequently, increasing the oxygen flux up to 2.6% leads to an increase in the carrier concentration. However, as the oxygen (O_2) flux > 2.6% is further increased (Figure 1), there is a decrease in the carrier concentration because the number of oxygen vacancies decreases as the oxygen flow increases. At the same time, oxygen supersaturation of the films leads to deactivation of donors and accumulation of oxygen atoms at the grain boundaries [14], leading to a decrease in the mobility and concentration and therefore an increase in resistivity [15].

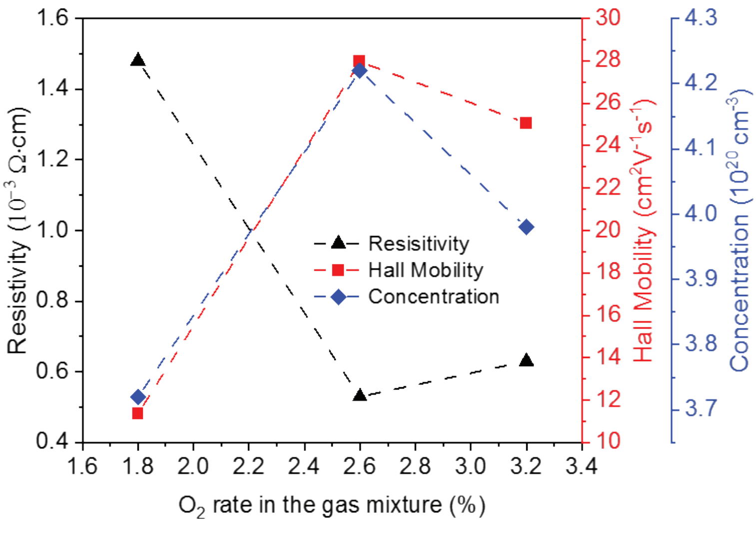

Figure 1 – Dependence of the resistivity, concentration, and Hall mobility of charge carriers of thin ITO films on the O_2/Ar gas-flow ratio

Figure 2 shows the dependence of the electrical properties of ITO films deposited at a magnetron power of 250 W, substrate temperatures of 20 and 150°C, oxygen flow of 0 and 2.6%. The minimum resistivity of 0.52×10^{-3} Ohm×cm is achieved with an increase in oxygen flow up to 2.6%. This low resistance is due to the high values of mobility and concentration of charge carriers 28 $cm^2V^{-1}s^{-1}$ and 4.2×10^{20} cm^{-3}, respectively. As it is described above, the oxygen vacancies presence in the crystal lattice is a fundamental factor affecting the electrical conductivity of ITO films. Therefore, the increase of carrier mobility values were achieved when the substrate temperature was increased up to 150^0C, which is due to the improvement of the crystal structure of ITO films [16]. By increasing the substrate temperature during the sputtering process, the resistivity of the films slightly increases, which is also described in [17].

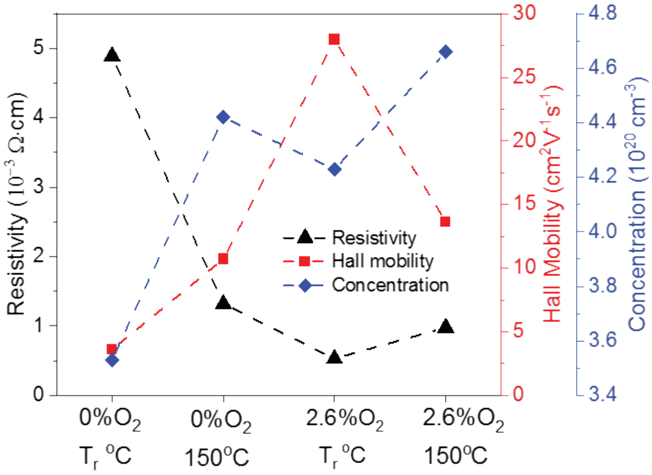

Figure 2 – Dependence of the resistivity, concentration, and Hall mobility of charge carriers of thin ITO films on the substrate temperature

Transmittance is an essential parameter for TCO films. Figure 3 shows the transmittance spectra of ITO thin films deposited at different parameters in wavelengths ranging from 300 to 1100 nm. The transmittance of all films exceeds 75% in the visible and near infrared ranges, and for films grown at O_2 partial concentration of 2,6–3,2% the transmittance in the wavelength range of 400-600 nm is additionally enhanced, being about 82–85% in the range 420-720 nm and more than 90% in near IR spectrum (1000-1100 nm). It can be concluded that the increasing oxygen flow during deposition has a beneficial effect on the optical properties of the films [18,19]. Transmittance results indicate a significant improvement in the visible region of the spectrum. This increase in transmittance changes with increasing oxygen flow rate [20, 21].

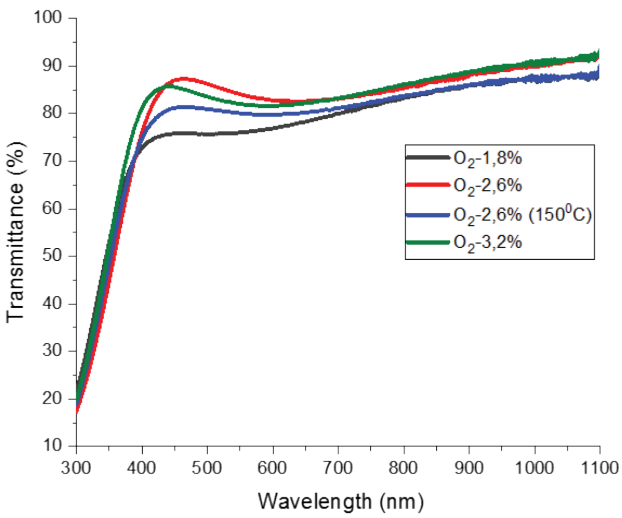

Figure 3 – Transmittance spectra of ITO films on the O_2/Ar gas-flow ratio

In order to investigate the optical properties of ITO in more detail, optical constants were measured using SCOUT software. The obtained results of refractive indices and extinction coefficients are shown in Figure 4.

During the optimal oxygen flow (2.6%) in the sputtering process, we obtain ITO films with low extinction coefficient k (0.04–0.02) within the wavelength range of 450–800 nm (Fig.4), indicating less absorption, which leads to an increase in the transmission coefficient (Fig.3). For an oxygen flow of 2.6% the refractive index (n) has the highest values over the entire wavelength range, reaching maximum value of 3.1 at 300 nm. It can be observed that for films with oxygen consumptions of 2.6%, and 3.2% the refractive indices exceed the values for 1.8% over the entire wavelength. In the visible region 400–800 nm, the extinction coefficients for 1.8% have higher values in comparison with 2.6% and 3.2% approximately retain the natural dependence of the refractive index on oxygen supply. In this case, the optimal oxygen flow (2.6%) leads to the lowest value of the extinction coefficient – 0.023.

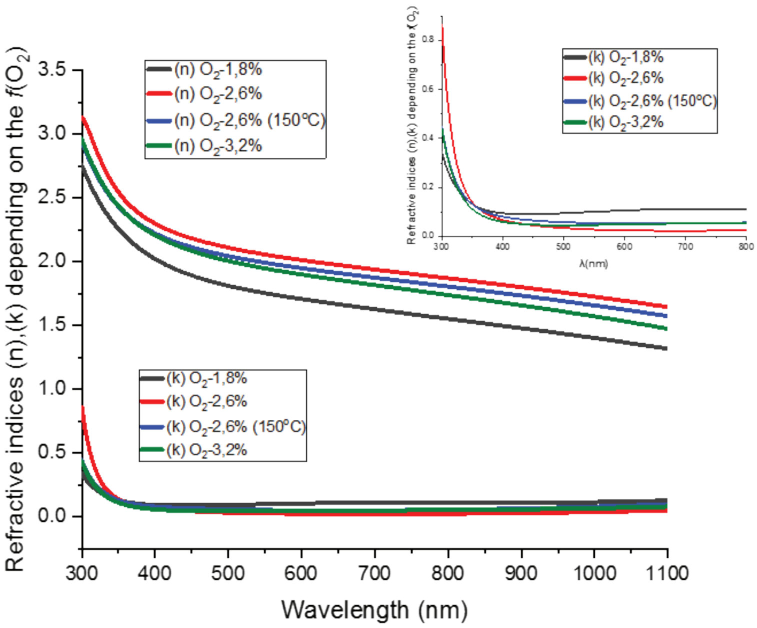

Figure 4 – (a) Refractive index (n) and Extinction coefficient (k) depending on the $f(O_2)$

Knowing the electrical resistivity and optical transmittance of ITO thin films, the quality index ϕ_{TC} proposed by Haacke [22] was calculated using Eq.(1) to compare the performance characteristics of the films.

$$FoM = T^{10} / R_s \qquad \text{Eq,(1)}$$

Where,
FoM – Figure of Merit
T – Transmission
R_s – Sheet resistance

The obtained results are presented in Figure 5. The maximum FoM value was achieved with an oxygen flow of 2.6% and is 222.6×10^{-5}, which can be assessed as a good result in comparison with the previously obtained results. This was achieved by simultaneously increasing the transparency of the films and reducing the electrical resistivity (Figs. 2 and 3). The worst results are achieved with conventional sputtering of an ITO target without the use of oxygen and external heating (Figs. 2 and 5).

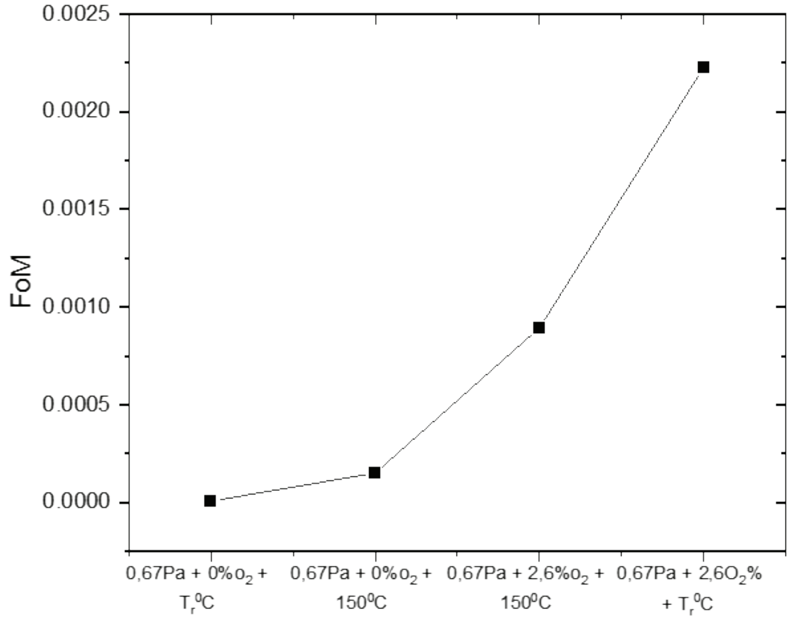

Figure 5 – Calculated Figure of merit of ITO layers as a function of deposition parameters

Conclusion

ITO films were deposited by DC reactive sputtering at a constant magnetron power of 250W. The effect of oxygen consumption during deposition on the optical and electrical properties of the films was studied. The change in electrical properties can be caused by an increase in the content of Sn4+ ions at the position of substitution instead of In3+ ions and, as a consequence, an increase in the number of oxygen vacancies with subsequent deactivation of donors in the process of increasing oxygen consumption. The minimum value of resistivity, 0.52 × 10 Ohm·cm, was achieved at an oxygen flow rate of 2.6% and maximum charge mobility parameters equal to 28 $cm^2V^{-1}s^{-1}$. The transmittance in the wavelength spectral range of 400-650 nm is additionally enhanced, being about 85% in the visible range and more than 90% near IR spectrum (1000-1100 nm) with a minimum extinction coefficient k (0.04-0.02) in the range of 450-1100 nm. The maximum value of FoM was achieved at an oxygen rate of 2.6% and is equal to 222.6×10^{-5}.

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ОТТЕГІ АҒЫНЫНЫҢ МАГНЕТРОНДЫ БҮРКУ АРҚЫЛЫ СИНТЕЗДЕЛГЕН ITO ҚАБЫРШАҚТАРЫНЫҢ ЭЛЕКТРЛІК ЖӘНЕ ОПТИКАЛЫҚ ҚАСИЕТТЕРІНЕ ӘСЕРІ

Аңдатпа

Қалайымен легирленген индий оксидінің жұқа қабықшалары аралас аргон-оттегі атмосферасында жылтыратылған кремний үлгілері мен заттық шынылардың бетіне тұрақты тоқта магнетронды шашырату арқылы синтезделді. Тұндырудың басқа параметрлері: жұмыс қысымы, магнетронның қуаты және субстраттың айналу жылдамдығы тұрақты болып қалды. Жұқа қабыршақтардың қалыңдығы мен тығыздығы рентгендік рефлектометрия әдісімен өлшенді. Оттегі ағынының жылдамдығы мен субстрат температурасының оптикалық және электрлік қасиеттерге әсері зерттелді. Жұқа қабыршақтардың электрлік қасиеттері (меншікті электр кедергісі, холл қозғалғыштығы және заряд концентрациясы) Холл эффектісін қолдана отырып, Ван дер Пау әдісімен өлшенді. Минималды кедергі мәні $0,52\times10^{-3}$ Ом·см және зарядтың максималды қозғалғыштығы оттегі шығыны 2,6% болғанда 28 см2В-1с-1 тең. Қабыршақтардың өткізу спектрлері 300-ден 1100 нм-ге дейінгі толқын ұзындықтарында өлшенді. Барлық қабыршақтардың өткізгіштігі көрінетін және жақын инфрақызыл диапазонда 75%-дан асады. Оттегі ағынының жылдамдығын арттыру мен субстратты 150°С дейін оңтайлы қыздыру қабыршақтардың кристалдылығының жоғарылауына, нәтижесінде холл қозғалғыштығы мен өткізгіштігінің жоғарылауына әкелетіні анықталды.

Тірек сөздер: кремний күн элементтері, кристалдану, магнетронды шашырату, жұқа қабықшалар, индий оксиді.

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ВЛИЯНИЕ ПОТОКА КИСЛОРОДА НА ЭЛЕКТРИЧЕСКИЕ И ОПТИЧЕСКИЕ СВОЙСТВА ПЛЕНОК ITO, СИНТЕЗИРОВАННЫХ МЕТОДОМ МАГНЕТРОННОГО РАСПЫЛЕНИЯ

Аннотация

Тонкие пленки легированного оловом оксида индия были синтезированы методом магнетронного распыления на постоянном токе на поверхность полированных образцов кремния и предметных стекол в смешанной аргон-кислородной атмосфере. Остальные параметры осаждения: рабочее давление, мощность магнетрона и скорость вращения подложки оставались постоянными. Толщину и плотность тонких пленок измеряли методом рентгеновской рефлектометрии. Исследовано влияние скорости потока кислорода и температуры подложки на оптические и электрические свойства. Электрические свойства (удельное сопротивление, холловская подвижность и концентрация заряда) тонких пленок измерялись методом Ван дер Пау с использованием эффекта Холла. Минимальное значение удельного сопротивления $0,52\times10^{-3}$ Ом·см и максимальная подвижность заряда 28 $см^2В^{-1}с^{-1}$ достигнуты при пропорциональной газовой смеси кислорода 2,6%. Спектры пропускания пленок измерялись в диапазоне длин волн от 300 до 1100 нм. Пропускание всех пленок превышает 75% в видимом и ближнем инфракрасном диапазонах. Установлено, что увеличение скорости потока кислорода и нагрев подложки до оптимального значения 150°С приводят к увеличению кристалличности пленок и, как следствие, к увеличению холловской подвижности и коэффициента пропускания.

Ключевые слова: кремниевые солнечные элементы, кристаллизация, магнетронное распыление, тонкие пленки, оксид индия.

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