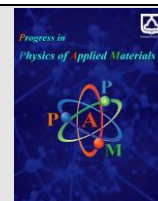




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Simulation and Fabrication of Three-Layer SnO₂/Ag/SnO₂ Nanostructure Coating for Energy Storage

Esmail Shahriari ^{a*}, Morteza Raeisi ^b, Sanaz Alamdari ^c^a Department of Physics, National University of Skills (NUS), Tehran, Iran^b Department of Physics, Faculty of Basic Science, Shahrekord University, Shahrekord, Iran^c Department of Nanotechnology, Faculty of New Sciences and Technologies, Semnan University, Semnan, Iran

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ABSTRACT

In this work, the three-layer SnO₂/Ag/SnO₂ coating on the glass substrate was designed and the optimal thickness was calculated using Film Wizard software. Then the designed samples were fabricated using thermal evaporation technique. Analysis was conducted on optical, electrical, and thermal properties of the samples, including coefficient of merit, surface resistance, and optical transmittance and reflectance. The results showed that the three-layer structures, SnO₂/Ag/SnO₂, have the necessary and favorable conditions for use as heat insulating and energy saving coatings on glass windows. The optimal thickness was determined 20nm for silver and 70 nm for SnO₂. The recorded optical transmission in the visible spectrum and the reflected light in the infrared spectrum are 79.66% and 88.66%, correspondingly. The surface electrical resistance of 11.69 Ω/sq has been obtained for the constructed system, which is a suitable value for this coating. The calculated optical values show that the heat transfer from sunlight is minimized through this coating and this coating can be used as a suitable energy storage.

1. Introduction

In recent years, an important technology has been used for energy wastage by the glass of buildings and cars. One of the projects carried out was to create thin and transparent coatings that improved the thermal performance of glass windows [1]. The coatings are designed according to their type of application to pass visible rays and reflect infrared rays. Optical transparent metal oxide nanocomposites thin films, including SnO₂, ITO, ZnO, BaWO₄, and ZnO, find extensive application in consumer electrical devices and energy storage systems [2–5]. The anti-reflection effect of the oxide/metal/oxide multilayer material structure and the surface plasmon effect of the sandwiched Ag metal layer might enhance the performance of these oxide/metal/oxide multilayer coatings [6, 7]. The presence of an Ag thin layer between the

upper and lower oxide thin layers in the oxide/metal/oxide multilayer structure serves to safeguard the electrical characteristics and stability from the potential deterioration resulting from metal oxidation in a chemical setting. SnO₂ is thermally stable, non-toxic, inexpensive, and plentiful [8]. Furthermore, the SnO₂ film exhibits exceptional transparency within the wavelength spectrum of 350 to 900 nm. Tin oxide coatings have exceptional chemical stability and resistance to corrosion in both acidic and alkaline conditions [9]. It exhibits chemical and thermal stability when subjected to the hydrogen plasma procedures often employed in the manufacturing of solar cells. Within the intermediate stratum, gold, silver, and copper are the most extensively utilized. Gold is considered to be more costly than silver, whereas copper exhibits more sensitivity to oxygen. Therefore, in this study, we studied the SnO₂/Ag/SnO₂ three-layer structures. Previous studies

* Corresponding author.

E-mail address: esmaeil.phy@gmail.com, ashahriari@nus.ac.ir

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have shown several benefits of three-layer transparent conductive films, which have a comparatively smaller thickness, in comparison to single-layer transparent conductive oxide films [10]. Optical and electrical characteristics of three-layer structures are greatly influenced by the thickness and deposition circumstances of silver. For optimal characteristics of high transmittance and low resistance, the silver coating should be thin, homogeneous, and continuous. $\text{SnO}_2/\text{Ag}/\text{SnO}_2$, a kind of three-layer transparent conductive oxide, exhibits excellent optical transparency in the visible spectrum and robust reflection in the infrared spectrum. These characteristics render them well-suited for the application of infrared protective coatings. Within the construction sector, architectural engineers are actively seeking various methods to enhance the insulation of building walls and windows in order to conserve energy. Among these, windows are identified as the primary source of energy wastage throughout both winter and summer seasons. Hence, scholars have lately employed thin layer coatings on building glass as a means to impede the transmission of heat through the windows [11, 12]. By exhibiting increased light reflection in the infrared and ultraviolet regions and enhanced light transmission in the visible range, this coating will be more advantageous for insulation purposes. Rabizadeh and colleagues developed and produced a three-layered structure of $\text{SnO}_2(100\text{nm})/\text{Ag}/\text{SnO}_2(100\text{nm})$ by magnetron sputtering used with glancing angle deposition [13]. With this method they obtained the optimum thickness of Ag as 25 nm. Also, in a comparative fabrication method Shahidi et al. have investigated the optical and electrical properties of $\text{SnO}_2/\text{Ag}/\text{SnO}_2$ by sputtering at zero as well as 75° angles of substrate normal with respect to vapor flow direction from the target [14]. They fixed the thickness of SnO_2 layer on 80 nm and thickness of the silver layer was considered $\sim 15\text{-}25$ nm. However, in this work we fabricated the thin films using by thermal evaporation and we found that the optimum thickness for SnO_2 and Ag as 70 and 20 nm, respectively. In order to save energy, we have designed coatings with a three-layer arrangement of $\text{SnO}_2/\text{Ag}/\text{SnO}_2$ in such a way that they can simultaneously transmit visible rays and reflect near-infrared rays. By using the simulation method, we have determined the optimal thickness of each layer in this system and made the suitable heat reflective layer using the thermal evaporation method. In this simulation, we used Film Wizard software which is the most powerful, versatile, and user-friendly software package ever to be offered to the thin film engineer/scientist. It combines optimization and synthesis design capabilities with powerful analytical tools for interpreting spectrophotometric and ellipsometric data, modeling of film layers, and performing figure of merit to optimum actual thicknesses and indices of deposited layers.

2. Experimental

Semiconductor tin oxide powder with 99.9% purity and Ag powder with 99.9% purity were purchased from Sigma-Aldrich and used as the target material. The multi-layer coating was successively applied without breaking the vacuum by thermal evaporation method. The three-layered structure of $\text{SnO}_2(70\text{nm})/\text{Ag}(x)/\text{SnO}_2(70\text{nm})$ with silver layer thicknesses of 10, 15, 20, and 25 nm was coated on

the glass substrate and the samples were labeled as S1, S2, S3, and S4, respectively. Before stacking the layers, the chamber was evacuated to a pressure of 1×10^{-5} mbar. The chamber pressure during deposition was 5×10^{-6} mbar. The deposition rate and the thickness of each layer were controlled using a quartz crystal thickness monitor. The accumulation rate was chosen as 0.1 nm/s for SnO_2 layers and 0.05 nm/s for Ag layer. The surface resistance of fabricated multi-layer system was also determined by a four-point resistance meter and optical transmittance and reflectance of the samples were determined with a two-beam spectrophotometer model (Shimadzu UV-3100).

3. Results and discussion

The $\text{SnO}_2/\text{Ag}/\text{SnO}_2$ transparent conductive coatings were designed by using the Film wizard code. At this stage, in order to optimize the thickness of each layer, the reflection and transmission spectrum are calculated for each thickness and the surface resistance of the multilayer coating is determined using the following relationship [15]:

$$R_{sh} = \frac{Z_0}{2} \left(\frac{1}{\sqrt{R}} - 1 \right) \quad (1)$$

where R_{sh} is the surface resistance, R is the reflection in the infrared region, and $Z_0=377 \Omega$ is the impedance in free space. The reflection at the wavelength of 1700 nm is used as a criterion for the infrared region, which depends on the density of electrons in this region [16]. On the other hand, figure of merit F_{TC} is an important index in optimizing multilayer coatings that determines the relationship between electrical and optical properties of coatings. This quantity is presented by Huck as follows:

$$F_{TC} = \frac{T^{10}}{R_{sh}} \quad (2)$$

where T shows the maximum transmittance of the coatings. In the dielectric-conductor-dielectric (D/M/D) symmetric structures, it has been shown that if the thickness of the first and the third layers is equal, the transmittance of the structure is maximum [17]. To design this coating, the thickness of the Ag layer is fixed at a default value of 15 nm and the thickness of the SnO_2 layer is changed so much that the coefficient of merit of these coatings reaches its maximum value. The F_{TC} of $\text{SnO}_2/\text{Ag}(15\text{nm})/\text{SnO}_2$ coating for different SnO_2 layers is shown in Fig. 1.

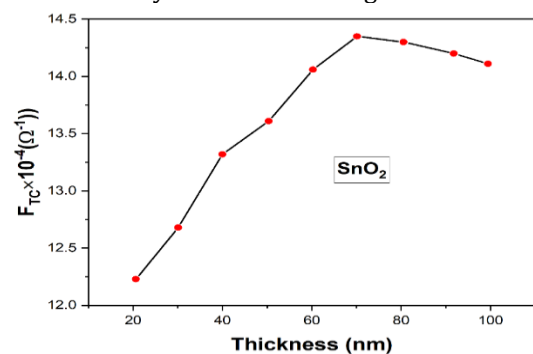


Fig. 1. Changes in the evaluated F_{TC} of SnO_2 layers with respect to changes in the thickness of the silver layer

As can be seen from the coefficient of merit diagram, the highest value of the coefficient of merit is related to the SnO_2 coating with a thickness of 70 nm. In the next step, we keep the thickness of SnO_2 constant and change the thickness of silver so that the coefficient of merit of the

three-layer coating is optimized. First, using the design software, and after determining the optimal thickness, the samples were made using the thermal evaporation method for Ag thicknesses of 10, 15, 20, and 25 nm. The transmission and reflection spectra of the samples were measured in the wavelengths range 300 to 2000 nm and are shown in Figs. 2 and 3, respectively. According to Fig. 2, the transmission spectrum in the silver layer with a thickness of 15 nm in the visible region is slightly higher than that of the silver layer with a thickness of 10 nm, while it is significantly increased in the silver layer with a thickness of 20 nm. But, increasing the silver layer (Ag 25nm) has reduced the transmission spectrum in the visible region. The reduction of the transmission spectrum in thinner layers may be due to the increase of optical scattering in the semi-conducting boundaries, which is caused by the formation of separate islands by silver. On the other hand, the reduction of the spectrum in higher thicknesses of 20 nm is due to the fact that the continuity of the silver layer increases and the interlayer plays the role of mirror scattering. As shown in Fig. 2, increasing the thickness of the 25 nm layer causes the absorption edge shift to longer wavelengths.

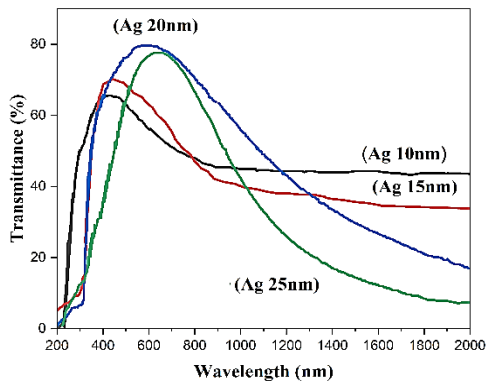


Fig. 2. Optical transmittance of the samples with respect to changes in the thickness of the silver layer

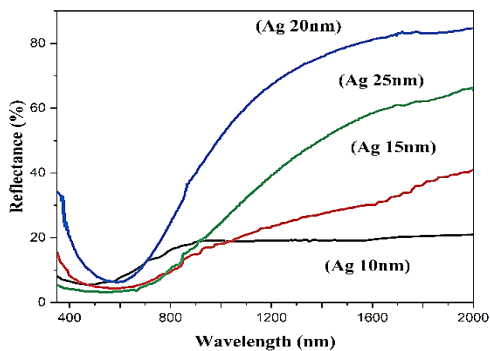


Fig. 3. Reflectance spectrum of the samples with respect to changes in the thickness of the middle silver layer

The transmission and reflection coefficient results are crucial in evaluating the optical performance of materials, especially in applications like coatings, sensors, and photonic devices. In this case, the sample with a 20 nm

thick Ag interlayer shows impressive optical properties, with a transmission coefficient of 79.66% in the visible region and a reflection coefficient of 88.66% in the infrared region (Fig. 3). These results suggest that the Ag interlayer plays a significant role in modulating the optical behavior of the sample across different wavelength regions. The transmission coefficient of 79.66% in the visible spectrum (roughly 400-700 nm) indicates that the 20 nm Ag interlayer is highly transparent in this region. This transparency is desirable for applications where light needs to pass through the material, such as in transparent conductive coatings used in touchscreens, solar cells, or optical filters. Silver, known for its high electrical conductivity, often provides excellent optical transparency when used in thin layers. At this thickness, the Ag layer is likely below its percolation threshold, meaning it allows visible light to pass through while still offering conductive properties. This high transmission can be attributed to the thinness of the layer, where destructive interference of reflected waves occurs, allowing most light to transmit.

Additionally, the increase in silver thickness and the corresponding decrease in sample transparency led to the reduction in transmittance with a 25 nm thickness. Fig. 4 represents the figure of merit for the manufactured $\text{SnO}_2/\text{Ag}/\text{SnO}_2$ layers, where the SnO_2 layer remains fixed and the Ag layers' thicknesses vary from 10 to 25 nm. The diagram clearly demonstrates that the ideal coefficient of merit is associated with silver having a thickness of 20 nm. As a result, this sample is much more suitable for energy storage and heat insulation coating applications.

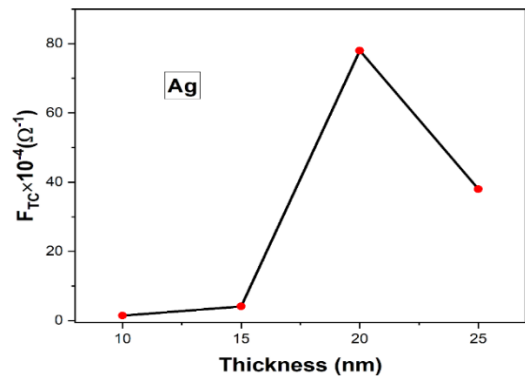


Fig. 4. F_{Tc} variation of $\text{SnO}_2/\text{Ag}/\text{SnO}_2$ thin films with respect to change in Ag layer thickness

Fig. 5 presents the band gap values for silver films with thicknesses of 10, 15, 20, and 25 nm, which are 4.08, 4.05, 3.99, and 3.98 eV, respectively. The data indicate that as the thickness of silver increases, the bandgap of the coatings decreases. The valence band's upward movement and the conduction band's downward shift are responsible for the decrease, which leads to a reduction in the band gap. The dense concentration of charge carriers in the conduction band and the presence of ionized atoms in the silver-plating layer are responsible for the narrowing of the energy band gap.

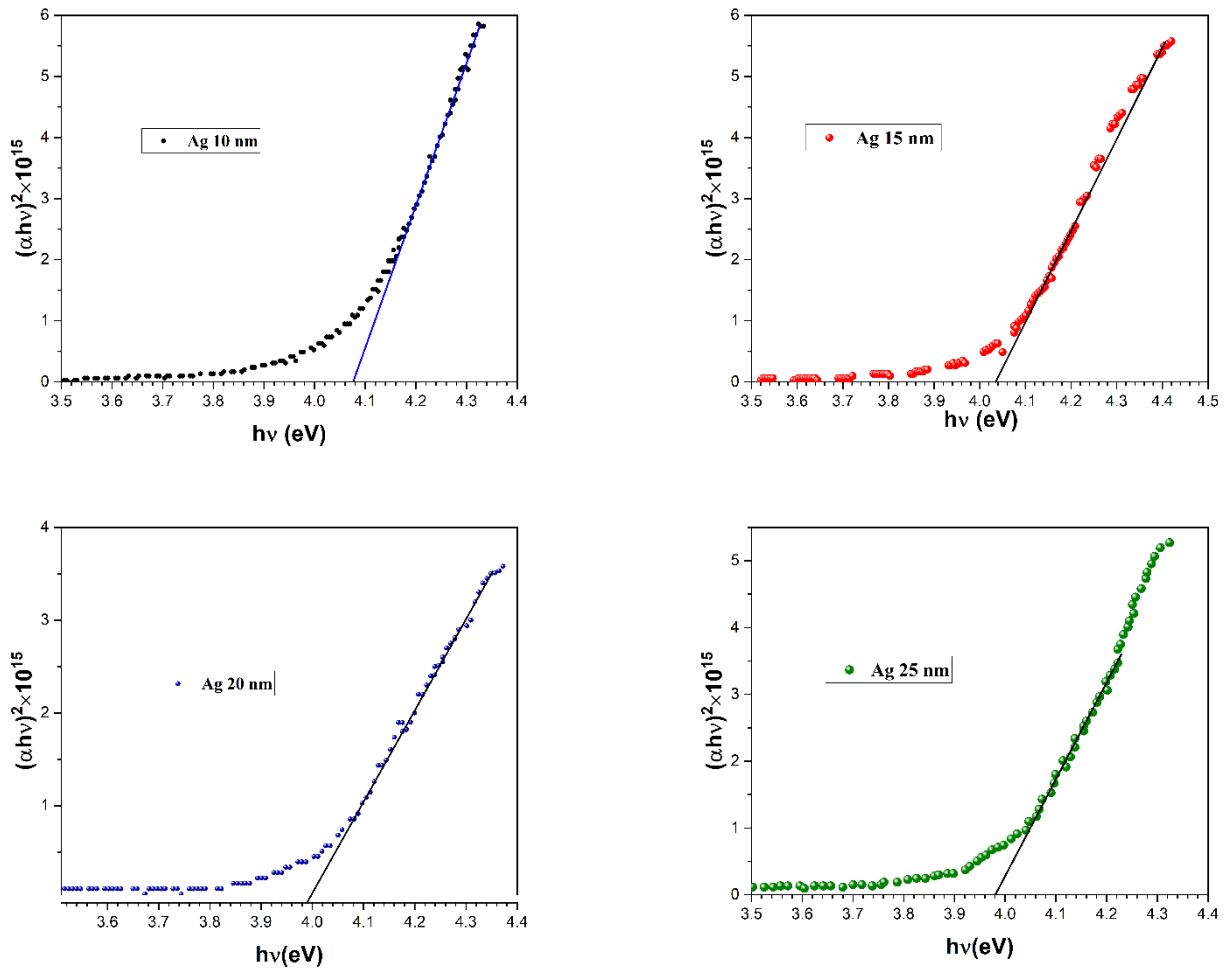


Fig. 5. Band gap of samples with respect to changes in the thickness of the silver layer

When the carrier charge density exceeds the Mott critical threshold, we can use electron-electron and electron-impurity scattering to characterize the band gap in a semiconductor. The removal of electrons from the metal results in their accumulation inside the semiconductor's conduction region, causing silver's surface atoms to acquire a positive charge. This effect led to an attraction between the electrons in the conduction region of the semiconductor and silver atoms. As a result, an electric field is created between the conduction region of the semiconductor and charged metal atoms. This phenomenon has two consequences: a downward shift of the conduction band and an upward movement of the

valence band [19]. In fact, under ideal conditions, the optical and electrical properties of the transparent conductive coating should be optimal, although achieving such a state is not easy. To achieve this goal, optimizing the merit coefficient that expresses the relationship between the optical and electrical properties of the sample is a useful measure. In this work, the merit coefficient of the optimized sample was obtained as $0.0143\Omega^{-1}$.

The emissivity of the sample in the wavelength range of 780–2500 nm can be calculated by using the following equation [20, 21]:

Table 1. The optical and electrical parameters of fabricated samples

Sample	Sheet resistance (Ω/sq)	Emissivity for ($780 < \lambda < 2500$)	Emissivity for ($\lambda > 3\mu\text{m}$)	R_{1700} (%)	Ag thickness (nm)
S1	199.1	-8.72×10^{-2}	2.11	23.64	10
S2	131.6	0.537	1.396	34.67	15
S3	11.69	0.141	0.124	88.66	20
S4	43.59	0.435	0.462	65.96	25

$$\varepsilon = 0.0129R_{sh} - 6.7 \times 10^{-5}R_s^2 \quad (3)$$

where ε is the emissivity of the coatings.

Also, in the range of $\lambda > 3 \mu\text{m}$, sheet resistance can be determined using [20]:

$$\varepsilon = \frac{4R_s}{Z_0} \quad (4)$$

The obtained results of reflectance in 1700 nm (R1700), emissivity and sheet resistance for all the samples, are listed in Table 1. It can be observed that the emissivity in S1 is high, and with increasing the Ag thickness, its value decreases. The lowest value of 0.14 belongs to sample with optimum thickness of Ag. Windows in buildings are primary contributors to energy loss during both the summer and winter seasons. Thus, it is crucial to develop low-emissivity coatings for windows to greatly decrease heat transmission. Optimal material should possess exceptional transparency and little heat absorption. We use the U and G parameters to evaluate the quality of materials selected for energy storage in glass windows. The U-value measures the heat loss/gain through the window assembly, while the G-value measures the amount of solar energy transmitted through the glass. The value of U quantifies the heat transfer that occurs through the glass. Hence, the coatings with a lower U value exhibit superior thermal insulating property. Moreover, the solar thermal gain coefficient refers to the quantity of direct solar radiation that manages to penetrate the glass. Compared to solid glass, a lower G-value results in less heat transmission from direct sunlight. Such samples are appropriate for locations with direct solar exposure [22]. The objective of this study was to determine the U-value and G-value by simulating a window system including a 4 mm thick layer of coated glass and a three-layered SnO₂/Ag/SnO₂ (S3) using Windows software. Table 1 displays the parameters derived from the simulation for each sample. We conducted the simulation using the transmittance and reflection spectra of the samples in the visible and solar areas, along with the emissivity. Sample S3 displays the lowest values of G and U, which are 0.61 and 1.41, respectively. As a result, sample S3 has superior insulating properties against both ambient heat and radiant heat from direct sunlight. Nezhad E and his colleagues conducted research on ZnO/Ag/ZnO materials with varying silver thicknesses, and their findings align well with our results [23].

4. Conclusion

Using the thermal evaporation technique, this work examined the optical, electrical, and thermal characteristics of three-layered SnO₂/Ag/SnO₂ thin films deposited on glass with thicknesses of 10, 15, 20, and 25 nm of silver in the middle layer, and 70 nm for SnO₂ in the upper and lower layers. The data indicate that the silver sample with a thickness of 20 nm had the lowest surface resistance, measuring 11.69 Ω/sq . Research findings indicate that the sheet resistance is strong at low thicknesses of Ag and decreases as the thickness of Ag

increases. Furthermore, it has been shown that the band gap energy of the samples exhibits a decrease as the thickness of Ag increases, namely from 4.08 eV to 3.98 eV. In addition, the analysis of the transmission and reflection coefficients of the samples reveals that the samples with a thickness of 20 nm of Ag exhibit the greatest transmission coefficients in the visible area and reflection coefficients at the wavelength of 1700 nm. Values of emissivity, heat transfer rate via material (U-value), and heat transfer from direct sunlight (G-value) are highly efficient. Sample S3 exhibits the minimum values of G, U, and emissivity, precise at 0.61, 1.41, and 0.14, correspondingly. Consequently, the optimal thickness of Ag for SnO₂/Ag/SnO₂ thermal insulation coatings in smart windows is 20 nanometers.

Conflicts of Interest

The author declares that there is no conflict of interest regarding the publication of this article.

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