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Enhancement of transmittance of indium tin oxide coated glass plates

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The transmittance of a glass plate with a transparent conducting coating of a material like indium tin oxide (ITO) can be enhanced by depositing a thin film of a material like silicon dioxide (whose thickness need not be tightly controlled) on top of the transparent conducting coating, avoiding the necessity of designing and depositing a complex multilayer antireflection coating on the opposite surface of the glass plate. The theory behind this performance of the silicon dioxide film and some actual experimental results have been reported.

Keywords: Indium tin oxide, Transmittance, Silicon dioxide, Thin film, Antireflection coating

1 Introduction

Transparent conducting coatings of materials like indium tin oxide (ITO) are commonly deposited on glass and plastic substrates for a variety of applications: as light-transmitting electrodes in device applications like solar cells, electroluminescent and liquid crystal displays and light emitting diodes for their good visible transmittance and low electrical resistivity[1,2,3,4]; for architectural applications like thermal insulation of windows and prevention of radiative cooling because of their high infrared reflectance[5]; and for electromagnetic interference (EMI) shielding applications[6]. Over the past few years, other materials such as In_2O_3:Mo (IMO), In_2O_3:Ti (ITiO) and other high mobility transparent conducting oxide (TCO) materials[7]; reduced-indium materials such as ZnO-In_2O_3, In_2O_3-SnO_2 and Zn-In-Sn-O multicomponent oxides and indium-free materials such as Al- and Ga-doped ZnO (AZO and GZO)[8], organic materials such as intrinsically conducting polymers like PEDOT[9] and carbon nanotubes[10], have also been proposed as alternatives to ITO. However, the most widely used transparent conducting coating material continues to be ITO.

In the visible region, an ITO-coated glass plate will typically transmit about 82-89% of the incident light intensity. For various applications, it may be desirable to increase the transmittance of the ITO-coated glass plate. The overall transmittance of an ITO-coated glass plate can be enhanced by air annealing, as described by Trejo-Cruz *et al.*[11], but annealing shifts the transmission window of the ITO film to longer wavelengths, without significantly enhancing the transmittance in the visible region. Often, the increased transmittance of an ITO-coated glass plate is accomplished by depositing a broadband antireflection (BBAR) coating (comprising 3 or more films of two or more materials and of tightly controlled thicknesses) on the uncoated back surface of the glass plate, but this can only increase the transmittance by 4% at most (if the BBAR coating is ideal and reduces the reflectance at the glass-air interface from the uncoated value of about 4% (Ref. 1) to zero). In the present study, using the concept of a two-layer antireflection coating comprising two films of high and low indices (as compared to the substrate index) and appropriate thicknesses, it has been shown theoretically that a thin film of low index material like silicon dioxide, deposited on top of the ITO film, can increase the overall transmittance of the glass plate by about 2-10% over the visible region of the spectrum. This film is much easier to deposit than the BBAR coating and its thickness need not be controlled anywhere nearly as tightly as the thicknesses of the individual films in the BBAR coating need to be controlled. Experimental results for the transmittance of an ITO-coated glass plate, and the same plate with an overcoat of a thin film of silicon dioxide of a range of thicknesses, have been presented to demonstrate the practical viability of the theoretical calculations.

It may be noted that the deposition of a thin silica layer on the transparent conducting layer need not interfere with the conducting applications of the latter layer (as required in solar cells or display panels) because the required electrical contacts to the conducting layer can be made, or the necessary areas masked off, before the deposition of the silica layer on top of the conducting layer.

2 Theory

A two-layer antireflection coating (ARC) deposited on a particular substrate can reduce the reflectance from the bare substrate to zero at a certain wavelength, unlike a single layer antireflection coating which can minimize the reflectance but can reduce it to zero only for a certain value of the film refractive index[12]. The structure of a two-layer ARC on a semi-infinite substrate is shown in Fig. 1, where n_0 and n_s are the refractive indices of the ambient medium and the substrate, respectively, and n_1, d_1 and n_2, d_2 are the refractive indices and thicknesses of the two films 1 and 2 deposited on the substrate, respectively. Here, R and T denote the reflectance (into the ambient medium) and transmittance (into the semi-infinite substrate), respectively. It can be shown[12] that for the reflectance to be reduced to zero at wavelength λ the following conditions must be satisfied:

$$\tan^2 \delta_1 = \frac{(n_s - n_0)\,(n_2^2 - n_0\, n_s)\, n_1^2}{(n_1^2\, n_s - n_0\, n_2^2)\,(n_0\, n_s - n_1^2)}$$

$$\tan^2 \delta_2 = \frac{(n_s - n_0)\,(n_0\, n_s - n_1^2)\, n_2^2}{(n_1^2\, n_s - n_0\, n_2^2)\,(n_2^2 - n_0\, n_s)} \qquad \ldots (1)$$

where $n_2 > n_s > n_1$ and $\delta_1 = 2\,\pi\, n_1\, d_1 / \lambda$, $\delta_2 = 2\,\pi\, n_2\, d_2 / \lambda$. If n_0, n_s, n_1 and n_2 are known, Eq. (1) can be solved to obtain the values of d_1 and d_2. There will be two sets of solutions for d_1 and d_2.

It can be seen that the coating structure in Fig. 1 is similar to that of the current requirement, a high-index ITO film coated on glass, and a low-index film deposited on top of the ITO film, which should lower the reflectance to zero at a certain wavelength and to low values around this wavelength, implying that the transmittance of the ITO-coated glass plate can be enhanced by depositing a low-index film on top of the ITO film. Therefore, if we choose $n_0 = 1$ (air), $n_s = 1.52$ (glass), $n_1 = 1.45$ (SiO_2) and $n_2 = 1.90$ (ITO), then by solving Eq. (1) two solutions for d_1 and d_2 can be found. These solutions for several different wavelengths are presented in Table 1.

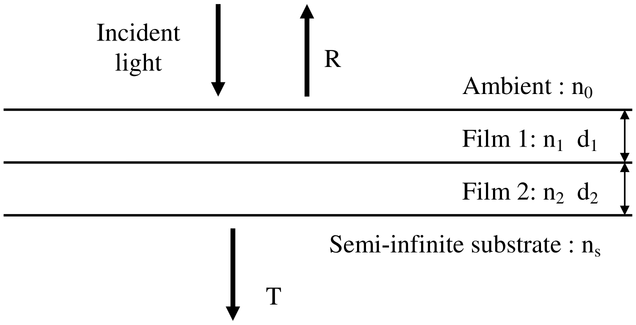

Fig. 1 — Schematic diagram showing the light reflection from and transmission through a semi-infinite substrate coated with two films, and the various parameters used to describe the system

It is found that $d_1 < d_2$ for solution I, while $d_1 > d_2$ for solution II. The thicknesses in solution II are generally chosen for the deposition of a two-layer ARC because the high-index film which is harder to deposit has lower thickness, the overall thickness of the coating is less than in solution I (at all wavelengths), and the low reflectance zone around the wavelength λ is broader than for solution I (Ref. 12). From published data of manufacturers[13] and our own long experience in the deposition of ITO coatings on glass, the ITO coatings required in most practical applications have thicknesses in the range 40-150 nm. Since the thicknesses d_2 in solution I are in this range, this class of solutions should be of more practical use for the applications we are considering here.

We now have to calculate the transmittance of a glass plate with an ITO film coated on it, and compare it with the transmittance of a glass plate with a SiO_2 film coated on top of the ITO film. For this calculation, the back surface reflection from the back surface of the substrate has to be taken into account. Moreover, in the visible region the ITO film will be somewhat absorbing, although the SiO_2 film and glass substrate will have very low absorption. We follow the formulations of MacLeod[13] and Dobrowolski[14] to calculate the overall net transmittance T_{total} of the structure shown in Fig. 2, where the substrate now has a large but finite thickness. For the sake of generality,

Table 1 — Solutions for thicknesses of the two layers (with specified refractive indices) of a two-layer ARC on glass, to yield zero reflectance at different wavelengths

Wavelength λ (nm)	Solution I		Solution II	
	d_1 (nm)	d_2 (nm)	d_1 (nm)	d_2 (nm)
350	48	67	72	25
400	55	77	83	28
450	62	87	93	32
500	69	96	103	35
550	76	106	114	39
600	83	115	124	42
650	90	125	134	46
700	97	135	145	49

absorption losses in the two films and substrate have been taken into account by assigning finite values of the extinction coefficient k for them. The refractive indices, extinction coefficients and thicknesses of film 1, film 2 and substrate are (n_1, k_1, d_1), (n_2, k_2, d_2) and (n_s, k_s, D), respectively, while the ambient medium has refractive index n_0. In Fig. 2, T_1 and T_2 are the transmitted intensities across the coating-substrate interface and the substrate-ambient interface, while R_1 and R_2 are the reflected intensities for light traveling from substrate to coating and from substrate to ambient medium, at these same interfaces. The reflected intensity into the ambient medium is R. Then the net transmittance T_{total} is given by[14]:

$$T_{\text{total}} = \frac{T_1\, T_2\, \tau}{1 - R_1\, R_2\, \tau^2} \qquad \ldots (2)$$

where $\tau = \exp(-\alpha_s D)$, $\alpha_s = 4\pi\, k_s / \lambda$.

The characteristic matrices of the films 1 and 2 are:

$$M_1 = \begin{pmatrix} \cos\tilde{\delta}_1 & \dfrac{i}{\tilde{n}_1}\sin\tilde{\delta}_1 \\ i\,\tilde{n}_1 \sin\tilde{\delta}_1 & \cos\tilde{\delta}_1 \end{pmatrix},$$

$$M_2 = \begin{pmatrix} \cos\tilde{\delta}_2 & \dfrac{i}{\tilde{n}_2}\sin\tilde{\delta}_2 \\ i\,\tilde{n}_2 \sin\tilde{\delta}_2 & \cos\tilde{\delta}_2 \end{pmatrix} \qquad \ldots (3)$$

where $\tilde{\delta}_1 = \dfrac{2\,\pi\,\tilde{n}_1\, d_1}{\lambda}$, $\tilde{n}_1 = n_1 - i\, k_1$, and similarly for film 2.

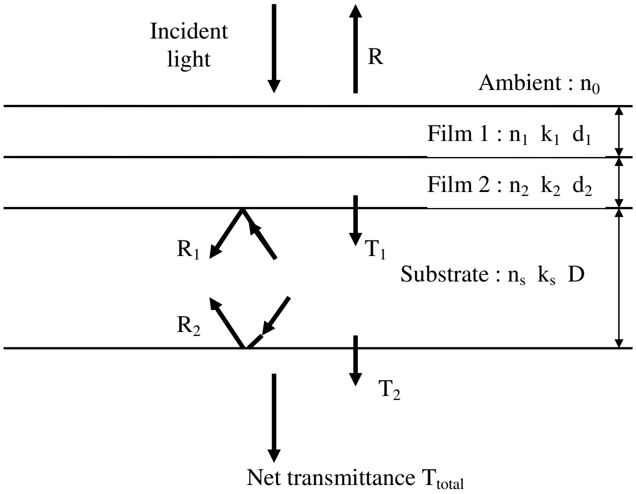

Fig. 2 — Schematic diagram showing the light reflection from and transmission through a substrate of finite thickness, coated with two films, and the various parameters used to describe the system

Using these matrices, intermediate steps in the calculation yield:

$$R = |r|^2 \,,\; r = \frac{n_0\, E_0 - H_0}{n_0\, E_0 + H_0} \,,\; \begin{pmatrix} E_0 \\ H_0 \end{pmatrix} = M_1\, M_2 \begin{pmatrix} 1 \\ \tilde{n}_s \end{pmatrix}$$

$$T = \frac{n_s}{n_0} |t|^2 \,,\; t = \frac{2\, n_0}{n_0\, E_0 + H_0}$$

$$R' = |r'|^2 \,,\; r' = \frac{\tilde{n}_s\, E_s - H_s}{\tilde{n}_s\, E_s + H_s} \,,\; \begin{pmatrix} E_s \\ H_s \end{pmatrix} = M_2\, M_1 \begin{pmatrix} 1 \\ n_0 \end{pmatrix}$$

$$T' = \frac{n_0}{n_s} |t'|^2 \,,\; t' = \frac{2\, n_s}{\tilde{n}_s\, E_s + H_s} \qquad \ldots (4)$$

where E_0, H_0 and E_s, H_s are the amplitudes of the electric and magnetic fields of the light wave in the ambient medium and substrate, respectively.

The reflected and transmitted intensities R_1, R_2, T_1 and T_2 in Fig. 2 are then given by:

$$T_1 = T = T' \qquad R_1 = R'$$

$$R_2 = \left| \frac{\tilde{n}_s - n_0}{\tilde{n}_s + n_0} \right|^2 \qquad T_2 = 1 - R_2 \qquad \ldots (5)$$

Using Eqs (2)-(5), the net transmittance of the coated substrate can be calculated, given the optical constants and thicknesses of the films and substrate.

Figure 3 shows this calculated transmittance at different wavelengths from 350 to 700 nm. The optical constants and thicknesses of the SiO_2 and ITO films, as also the optical constants of the glass substrate, were measured over this wavelength range. The dispersions in the optical constants of the SiO_2

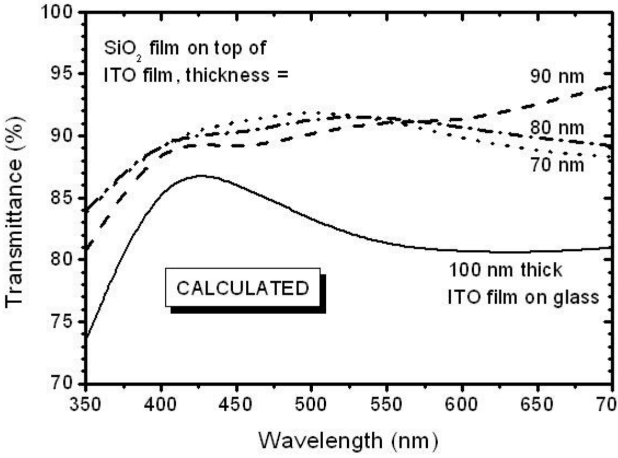

Fig. 3 — Theoretically calculated transmittance of light versus wavelength through a glass plate coated with a 100 nm thick film of ITO and a SiO_2 film of thickness 70, 80 and 90 nm

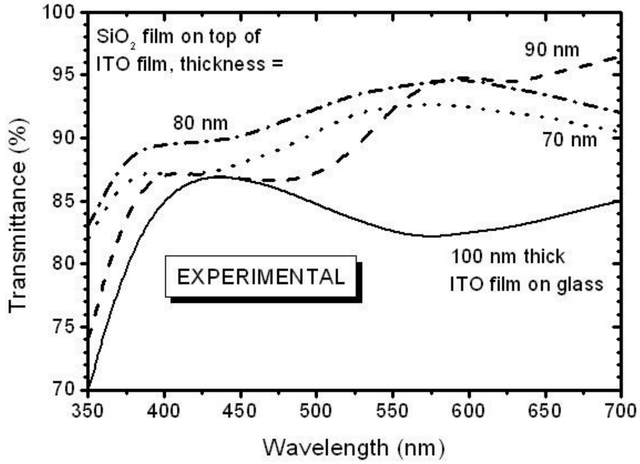

Fig. 4 — Measured transmittance of light versus wavelength through a glass plate coated with a 100 nm thick film of ITO and a SiO$_2$ film of thickness 70, 80 and 90 nm

film and the glass substrate were found to be very small in this wavelength range and can be ignored. The dispersion of n and k for the ITO film were found to be appreciable, and were taken into consideration for the calculations of transmittance. The thickness of the ITO film (film 2) was chosen to be 100 nm. The parameters chosen for the calculations are:

$n_0 = 1$, $n_1 = 1.45$, $k_1 = 0.0001$, $d_2 = 100$ nm, $n_s = 1.52$, $k_s = 0.00001$, $D = 1$ mm
(n_2, k_2) vary from (2.22, 0.016) at 350 nm to (1.70, 0.040) at 700 nm.

The bold curve in Fig. 3 shows the transmittance of a glass plate with a 100 nm thick layer of ITO coated on it. The dotted, dash-dotted and dashed curves show the transmittance of the ITO coated plate with additional films of SiO$_2$, having thicknesses $d_1 = 70$, 80 and 90 nm, coated on it.

3 Experimental Details

In order to experimentally verify the conclusions of these theoretical calculations, ITO films of thickness about 100 nm were deposited on plates of ophthalmic glass by reactive electron-beam evaporation in a Leybold L-560 vacuum coating plant. The residual oxygen pressure was maintained at about 6×10^{-4} mbar, the substrate temperature at 250°C and the film deposition rate was about 0.5 Å/s. Thereafter, thin films of SiO$_2$ of different thicknesses were deposited on the ITO film by e-beam evaporation, at a base pressure of less than 10^{-5} mbar, and at a deposition rate of about 5Å/s. The optical constants and thicknesses of the ophthalmic glass substrate and the ITO and SiO$_2$ films were determined by white light spectrophotometry (Filmetrics, model F-20) and spectroscopic ellipsometry (Woollam, model V-VASE), and the film thicknesses were confirmed by measurements of step height with an Ambios XP-200 Stylus Profiler. The transmittance spectra of the film-coated substrates were measured using a Shimadzu UV 3101 PC spectrophotometer over the spectral range 350 to 700 nm. Typical spectra are shown in Fig. 4, for a 100 nm thick ITO film-coated substrate and for substrates coated with this ITO film and with an overcoat of SiO$_2$ films of thicknesses 70, 80 and 90 nm.

4 Conclusions

From the results of the theoretical calculations in Fig. 3, it can be clearly seen that the deposition of the SiO$_2$ layer increases the overall transmittance by about 2-10% over the visible region. Moreover, the variation in transmittance at different wavelengths in the visible region is somewhat less for the ITO-coated plates with an additional film of SiO$_2$, than for the bare ITO coated plates. Finally, the thickness of the over coated SiO$_2$ film is not very critical; as any thickness between 70 and 90 nm yields about the same average transmittance over the visible region.

The experimental curves in Fig. 4 are seen to be in general agreement with the theoretically calculated curves in Fig. 3. The differences may be due to the very significant inhomogeneities (especially refractive index grading with depth) in experimentally deposited ITO films, which have been ignored in the calculations on which Fig. 3 is based, although the average dispersions in n and k for the ITO film have been taken into consideration for calculations. The main point to be noted is that the conclusions drawn from the theoretically calculated transmittance curves in Fig. 3, with respect to the increased values of the transmittance of ITO coated glass plates with an overcoat of SiO$_2$ films, are validated by the experimental transmittance curves in Fig. 4.

Thus, a SiO$_2$ thin film overcoat (thickness not too critical) on an ITO coated glass plate can significantly increase the transmittance of the plate in the visible region, which can be advantageous for device applications involving transparent conducting coatings.

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