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NUMERICAL MODELLING OF COPPER COMPOSITE THIN FILMS FOR SOLAR CONTROL FILTER

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ABSTRACT

Performance of solar control filter depends strongly on the individual layer thicknesses as well as the quality of their interfaces. In this work, a numerical modeling was designed and implemented to optimize the layer thicknesses of $\mathrm{Cu_2O/CuS}$, $\mathrm{Cu_2O/Cu}$ and $\mathrm{CuS/Cu}$ thin films for use as solar control filter using transfer matrix. The result shows that for $\mathrm{Cu_2O/CuS}$, with CuS in contact with the substrate, the optimized layer thickness of 110/30nm and for CuS/Cu and Cu_2O/Cu with Cu in contact with the substrate, the optimized layer thickness are 110/30nm and 100/30nm, respectively. The model also predicted transmittances of 67% for Cu_2O/CuS, 42% for CuS/Cu and 64.5% for Cu_2O/Cu at 550nm. A transmittances of 24.9-36.9% of IR radiation for all the films was realized. These results, when compared with previous works indicate the possibility of improvement in the performance of the filters.

Keywords: Solar Control Filter, Transfer Matrix, Infrared, Numerical Modeling, Transmittance

INTRODUCTION

In regions with warm climates particularly Nigeria, excessive heat is the measure factor in human thermal discomfort. Environmental conditions have impact on all aspect of human life, and since most of our life is spent inside buildings, creating a conducive environment within these buildings is vital to our life and human productivity. The application of selective thin films in glazing allows a more efficient management of heating and cooling loads of a building. The ultraviolet, visible, and infrared (IR) transmittance are three of the most important parameters for the evaluation of glazing lighting and energy performance, which depend significantly on the material of the film, that of the substrate, film thickness, roughness and uniformity (Sultan and Sultana, 2015).

Many different types of innovative optical thin film materials for lighting and solar thermal control have been developed in recent years in order to maximize energy saving by reducing solar heat gain and allowing at the same time good day lighting through windows and facades.

Fabrication and use of copper composite thin films in solar radiation control has been extensively reported by Eya (2010), Correa and Almanza (2004), Nair and Nair (1998), Nair *et-al* (1997). These films can control the reflectance, transmittance and absorptance of specific wavelength of solar radiation when deposited on glass. Correa and Almanza (2004), carried out experimental investigation on cuprous oxide (Cu_2O) and found that it is able to block infrared radiation by about 40 to 50%; while visible radiation is blocked by only 25 to 40% (75 to 60% transmittance). Nair *et al* (1997) reported that copper sulfide (CuS) is opaque to infrared radiation in a great extent. Therefore Correa and Almanza deposited 30 to 50nm CuS on an existing Cu_2O film that resulted to a total thickness of between 100 to 180nm. They finally reported that the infrared transmittance reduced to about 30% and they suggested that if the thickness of CuS film is increased there will be further reduction in the infrared transmittance (lower than 30%), but then, the visible transmittance reduces. Another development, they stated that depositing a metallic copper film on to Cu_2O film also produces another composite film with promising solar control property. These findings and suggestions can be further investigated to establish an optimum thickness for Cu_2O and CuS in Cu_2O/CuS composite, Cu_2O and Cu in Cu_2O/Cu composite and CuS and Cu in CuS/Cu composite that will give efficient solar control filters.

Theoretical modeling is a useful approach to fabricate multilayer thin film system for selective solar radiation applications. It gives insight into the work in a quick and precise manner, reducing the time and resources spent for the experimental work. The modeling of transmittance and reflectance of an optical thin film has been done by various researchers. Sultan and Sultana (2015) use transmittance and reflectance equations based on Fresnel's coefficients to model the optical transmittance of three novel films. Khelladi and Chabane (2013) developed optical model of ZnO on BK_7 glass substrate and ZnO on sapphire substrate using Sellmeire equations. These two methods are promising for single and thin film stack. Pieper *et al* (2006) presented an analysis of thin film optical filters using transmission line model. This approach established a transcription rule between field quantities and electrical quantities in what is referred as the Telegraph equation. Though the method is applicable to multilayered optical stack it appears to be sophisticated since complex calculations must be carried out and actually not necessary for the required level of details. Asdrubali *et al* (2011) investigated a theoretical model used to predict the optical properties of multilayered coated glazing systems. Their approach involves ray tracing method. The method is extremely rigorous, since it allows the predicting of the transmittance and reflectance of double panes combinations, with a little effort. However, for simple, quick and robust simulation Topasna and Topasna (2009) used a Transfer matrix method. This method permits a wider investigation of all different material combinations, with a little effort, it is therefore possible to change many variables such as film thickness, type of coating and their arrangement with respect to the substrate and type of substrate, at the aim of fulfilling requirements in mind.

This work is set to investigate or establish the optimum layer thickness of the copper composite thin films, for the purpose of maximizing the visible light transmittance and minimizing infrared transmittance through modeling transfer matrix of the films.

MATERIALS AND METHODS

A layer film is presented in the diagram below along with the components of the electric and magnetic fields of the incident, reflected, and transmitted waves.

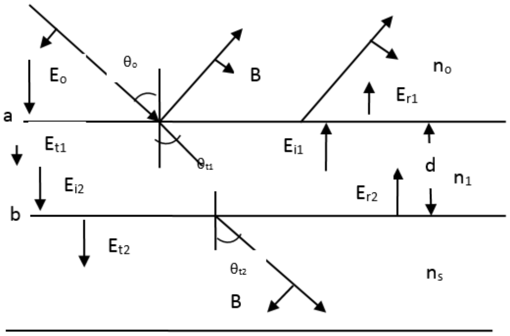

Fig.1: One layer film of thickness d and index of refraction $n_1$on a substrate of index n_s

The propagation of plane electromagnetic wave through optical media is described using classical Maxwell equations. These led to boundary condition which explains the continuity of the waves across the boundaries or interfaces between dielectrics. Assuming a homogenous, isotropic film with index of refraction n_1 deposited on a substrate with index of refraction n_s and placed in a medium with n_o, considering the boundary conditions, the tangential components of the resultant electric and magnetic fields are continuous across the interfaces, and are given by:

$$E_a = E_o + E_{r1} = E_{t1} + E_{i1} \qquad (1a)$$

$$E_b = E_{i2} + E_{r2} = E_{t2} \qquad (1b)$$

and

$$B_a = B_o\cos\theta_o - B_{r1}\cos\theta_o = B_{t1}\cos\theta_{t1} - B_{t1}\cos\theta_{t1} \qquad (2a)$$

$$B_b = B_{i2}\cos\theta_{t1} - B_{r2}\cos\theta_{t1} = B_{i2}\cos\theta_{t2} \qquad (2b)$$

Equations (2a) and (2b) can be written as functions of electric field by using $B = nE\sqrt{\varepsilon_o\mu_o}$:

$$B_a = \gamma_o(E_o - E_{r1}) = \gamma_1(E_{t1} - E_{i1}) \qquad (3a)$$

$$B_b = \gamma_1(E_{i2} - E_{r2}) = \gamma_s E_{t2} \qquad (3b)$$

where $\gamma_o \equiv n_o\sqrt{\varepsilon_o\mu_o}\cos\theta_o$,$\gamma_1 \equiv n_1\sqrt{\varepsilon_o\mu_o}\cos\theta_{t1}$ and $\gamma_s \equiv n_s\sqrt{\varepsilon_o\mu_o}\cos\theta_{t2}$.

$$E_{i2} = E_{t1}e^{-i\delta} \text{ and } E_{i1} = E_{r2}e^{-i\delta} \qquad (4)$$

The phase factors in equation (4) results from the fact that the wave has traveled a distance d inside the film layer; thus it is advanced or delayed by a phase of $\delta = \left(\frac{2\pi}{\lambda}\right) n_1 d\cos\theta_{t1}$. Now equations (1b) and (3b) become
$E_b = E_{t1}e^{-i\delta} + E_{i1}e^{i\delta} = E_{t2}$ and
$B_b = \gamma_1\left(E_{t1}e^{-i\delta} - E_{i1}e^{i\delta}\right) = \gamma_s E_{t2}$.
E_{t1} and E_{i1} can be solved in terms of E_b and B_b

$$E_{t1} = \left(\frac{\gamma_1 E_b + B_b}{2\gamma_1}\right)e^{i\delta} \qquad (5)$$

similarly

$$E_{i1} = \left(\frac{\gamma_1 E_b - B_b}{2\gamma_1}\right)e^{-i\delta} \qquad (6)$$

Substituting the above equations (5) and (6) in the initial field components equation (1a) and (3a), we obtain

$$E_a = E_b\cos\delta + B_b\left(\frac{i\sin\delta}{\gamma_1}\right) \text{ and } B_a = E_b(i\gamma_1\sin\delta) + B_b\cos\delta \qquad (7)$$

or in matrix formalism,

$$\begin{bmatrix} E_a \\ B_a \end{bmatrix} = \begin{bmatrix} \cos\delta & \frac{i\sin\delta}{\gamma_1} \\ i\gamma_1\sin\delta & \cos\delta \end{bmatrix}\begin{bmatrix} E_1 \\ B_1 \end{bmatrix} = M_1\begin{bmatrix} E_1 \\ B_1 \end{bmatrix} \qquad (8)$$

Each layer of the filter has its own transfer matrix and the overall transfer matrix of the system is the product of individual transfer matrices, taken in the order in which the light propagates through the multilayer stack,

$$M = \prod_{i=1}^{N} M_i \qquad (9)$$

Using equations (1) and (3) the transfer matrix (equation (8)) could be rewritten in the form;

$$\begin{pmatrix} E_o + E_{r1} \\ \gamma_o(E_o - E_{r1}) \end{pmatrix} = \begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix}\begin{pmatrix} E_{t2} \\ \gamma_s E_{t2} \end{pmatrix} \qquad (10)$$

where $M_1 = \begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix}$

The coefficient of reflection and transmission are defined as $r \equiv \frac{E_{r1}}{E_o}$ and $t \equiv \frac{E_{t2}}{E_o}$ (Macleod, 2001) and when rewritten in terms of the simplified form of equation (10) are as;

$$r = \frac{\gamma_o m_{11} + \gamma_o\gamma_s m_{12} - m_{21} - \gamma_s m_{22}}{\gamma_o m_{11} + \gamma_o\gamma_s m_{12} + m_{21} + \gamma_s m_{22}} \qquad (11)$$

$$t = \frac{2\gamma_o}{\gamma_o m_{11} + \gamma_o\gamma_s m_{12} + m_{21} + \gamma_s m_{22}} \qquad (12)$$

The reflectance R and transmittance T are given by $R = |r|^2, and\ T = |t|^2$, respectively (Macleod, 2001). To design a copper composite based solar control filter, a computer program based on the above mathematical formalism could be implemented using Mathcad. To run the simulations the following parameters must be provided (as shown in equations (8, 11 and 12)); the thickness (d) of all the films and, all the corresponding optical constants for all different materials deposited.

In the present study, the script for the calculation of the bilayer transmittance was written using the math palette in Mathcad version 14 program (Mathcad, 1999). Mathcad accepts and processes equations as they are typed. By defining variables and functions, equations can be linked together with intermediate results in further calculations. Variables in Mathcad field are defined from top to bottom and left to right. On a script once a variable is defined computation can be performed anywhere below and to the right of the definition.

The processes involved in writing the script (for modeling) included the following;

(i) The first step involved deriving the necessary equations. This was implemented in section (2) of this paper.
(ii) The second step was entering the parameters and constants. This involved defining the refractive indices of the films, substrate and air medium, also the film thicknesses (nm), wavelength range, permittivity and

permeability of free space, other constants and phase differences.

(iii) Creation of transfer matrices (Equation 8). The 'vector and matrix' palette was used. The matrices require the parameters and constants listed in (ii) above as inputs.

(iv) Defining the transmission coefficient (Equation 12) and percentage transmittance equation ($T = 100|t|^2$). This was achieved using Mathcad 'arithmetic and Greek' symbol palettes.

(v) Generating output value, each time the coating thicknesses are changed the program checks the script in the wavelength range not exceeding the upper range (1000nm) and prompted at that value. The Mathcad processes the equations and gives a table of percentage transmittance versus the wavelength (nm) in accordance with the wavelength range.

For graphical display, the default 'graph' palette was used. The axes were defined (T(%) Versus wavelength (nm)). Clicking the cursor in any empty space, it takes the script inputs processes all the equations and returns the graph output.

Computing the transmittance over the visible range, it is found that at 550nm, the middle of visible region, for example, the transmittance is 67.7% for Cu_2O/CuS, 65% for Cu_2O/Cu and 42.20% for CuS/Cu, Fig. (ii) and (iii).

Seventy-two (72) structures of Cu_2O/Cu, Cu_2O/CuS and CuS/Cu were investigated, by inspection those structures that show maximum transmittance in the visible as well as minimum transmittance in the infrared regions were selected and presented in table 1 as the summary of the optimized filters. It could be observed that the numerical values for the thickness are whole numbers; this is because a slight change in the thickness doesn't have significant effect on the optical response of the films. For effective thickness above (t_1+t_2=130nm) the films generally indicate that the optical response is very similar to that of bulk material indicating low visibility and high reflection in IR (Table 1). Eya (2010), Correa and Almanza (2004) and Chang and Paravae (2013) observed the same.

Table (2) compares the transmittances of Cu_2O/CuS and Cu_2O/Cu composites on glass substrate with the results in Eya (2010) and those of Correa and Almanza (2004). The calculated transmittance in the visible region are found to be 64.5% and 67% for Cu_2O/Cu and Cu_2O/CuS, respectively, which show slight difference with the values of 40% for Cu_2O/CuS (Correa and Almanza, 2004) and 35% (Eya, 2010). In the infrared region the transmittance is found to be 24.9% and 34.9% for Cu_2O/Cu and Cu_2O/CuS, respectively, which reveals differences with the values of 55% for Cu_2O/Cu and <30% for Cu_2O/CuS (Correa and Almanza, 2004), and less than 10% (Eya, 2010). These differences in values are due to the differences in the thickness of the films and the techniques employed.

Generally, the films have transmittance that increased with wavelength in UV and part of visible regions but decreased exponentially with wavelength in the remaining visible and NIR regions. The composites showed average transmittance of greater than 50% throughout UV-Vis-NIR regions.

Table 1: Summary of the optimized thickness copper composite thin films for solar control filter

Copper composite films	Film in contact with substrate	Optimized Layer Thickness(nm) (t_1)	(t_2)	Transmittance in visible region (T_{550}) (%)	Transmittance in IR region (T_{710}) (%)
Cu_2O/Cu	Cu	100	30	64.5	24.9
		100	40	64.5	25.1
		100	50	64.9	25.1
Cu_2O/Cu	Cu_2O	80	30	62.0	25.4
		90	40	64.1	33.0
		100	30	64.9	32.0
Cu_2O/CuS	CuS	100	30	66.1	35.2
		100	40	67.7	34.9
		110	30	67.1	34.1
Cu_2O/CuS	Cu_2O	90	40	62.8	36.5
		100	30	64.5	33.9
		110	30	63.6	32.1
CuS/Cu	Cu	90	50	45.0	36.9
		110	40	45.3	36.6
		110	30	45.8	35.7
CuS/Cu	CuS	100	30	42.4	35.5
		100	50	42.0	35.1
		110	40	42.2	36.0

Table 2: Comparison of this work with Correa and Almanza, and Eya works

Authors	Copper composites	Visible Transmittance (%)	Infrared Transmittance (%)	Effective Thickness (nm)	Method employed
Correa and Almanza (2004)	Cu_2O	60	50	~100	Sputtering
	Cu_2O/Cu	50	55	100	
	Cu_2O/CuS	40	<30	180	
Eya (2010)	Cu_2O/CuS	35	<10	540	CBD
This work	Cu_2O/Cu	64.5	24.9	130	Theoretical
	Cu_2O/CuS	67.7	34.9	140	
	CuS/Cu	42.4	35.5	140	

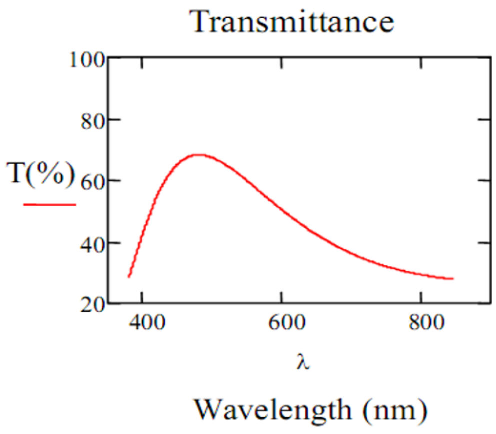

Fig. 2: Modeled Transmittance of Cu_2O/CuS films on glass substrate

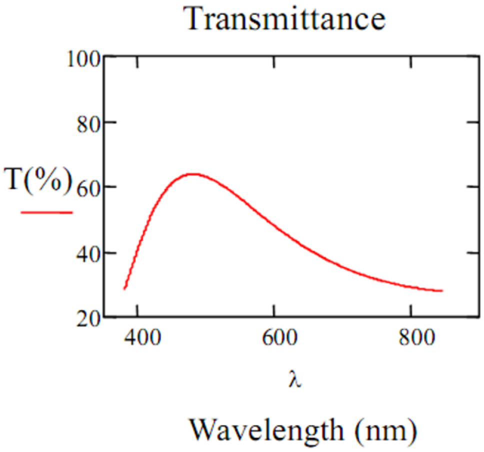

Fig. 3: Modeled Transmittance of Cu_2O/Cu films on glass substrate

Conclusion

In oder to save time and material resources numerical modeling is reccommended for the fabrication of thin film optical filter. The characteristics matrix of thin film and Fresnel's equations has been abstracted to optimise bilayer thickness of Cu_2O/CuS, CuS/Cu and Cu_2O/Cu thin films. The optimised layers for the films were 110/30, 110/30 and 100/30nm respectively for visible transmition of 42-67% at 550nm wavelength and IR transmittance of 25-37% at 710nm. These results, when compared with previous works, indicate the possibility of improvement in the performance of the filters.

REFERENCES

Asdrubali Francesco, Georgio Baldinelli and Smuele Schiavoni, (2011). *Experimental and Ray Tracing Evaluation of the Transmittance of Glazing Systems with Selective Coatings at Various Angles of Incidence*. Proceedings of Building Simulation 12th Conference of International Building Performance simulation Association Sydney 14-16 November, 2011.

Chang Chih-hung and Paravae Vas-Umnuay. (2013). *Growth Kinetics of Copper Sulfide Thin Films by Chemical Bath Deposition*. ECS journal of solid state science and Technology vol.2 (4) (120-129).

Correa, G. and Almanza, R. (2004). *Copper Based Thin Films to Improve Glazing for Energy-savings in Buildings.* Solar Energy, 76 (1-3); 111-115.

Eya, D.D.O. (2010). *Chemical Bath Deposition and Characterization of* Cu_2O-Cu_xS *Thin Films*. Turk Journal of Physics 34 (51-57)

Khelladi N Bouchenak and Chabane N.E. (2013). *Simulation Study of Optical Transmission Properties of ZnO Thin Film Deposited on Different Substrates.* American Journal of Optics and Photonics vol.1 (1) (1-5)

Macleod, H. A. (2001). *Thin-film Optical Filters* (3rd ed.). Arizona: Institute of Physics, London. Pp 22-23.Mathcad users guide mathcad 2000 Professional (1999). Math soft,Inc. Massachusetts USA

Nair, T.S. and Nair, P K. (1998). *Chemical Bath Deposition of* Cu_xS *Thin Films and their Prospective Large Area Application.* Second. Sci. Technol.4. 191-19

Nair,T.S.,Nair,P.K.,Huang,L.,Hailin,H.,Meyers,E.A and Zingaro,R.A. (1997). *Formation of p-type* Cu_3BiS_3*absorber Films by Annealing Chemically Deposited* Bi_2S_2-CuS *films*. J. Matter. Res., 12, 651-656.

Pieper R, Shirvalkar M, and Salvatierra J. (2006). *A Transmission Line Model for Analysis of Thin Film Optical Filters.* Proceedings of the 30th Southasian symposium on System Theory TennessseeTechnological University Cookeville,TN U.S.A March 5-7 2006

Sultan, Md, and Sultana, N. (2015). *Analysis of Reflectance and Transmittance Characteristics of Optical Thin Film for Various Film Materials, Thicknesses and Substrates.* Journal of Electrical & Electronics 4.3:1.

Topasna Daniel. M. and Topasna Gregory. A. (2009). *Numerical Modeling of Thin Film Optical Filters* Proc. SPIE. (http//spie.org/x102159.xml)