Design and Evaluation of Homogeneously Mixed Dielectric Antireflection Coatings with ZEMAX

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Abstract
Mixed dielectric films of ZnS and MgF₂ have been modeled in IR (3-5μm) band to reduce a reflectance from ZnS substrate which is around 14%. Reflectance value are enhanced starting from a mixed quarter single layer, double and triple layer as innermost and intermediate layers (quarter-quarter-quarter) and (quarter-half –quarter) ARC's. The designed layers are optimized with Zemax-EE operand to reach the target by varying their thickness and refractive indices simultaneously. The analysis has shown that the proposed mixing multilayer construction are very effective in enhancing the transmittance for ZnS.

Keywords: Mixed dielectric films, ARC's coatings, Zemax software, Lorentz-Lorenz theory, Drude dispersion theory.

1. Introduction
Even today, optical filters and coating are widely used in all optical and infrared optical instruments. Antireflection coatings(ARC’s) still exceed all the other types of filters and coatings. The need for ARC’s depends on applications; which required reducing of surface reflections and increasing in transmission considerably [1-4]. Further, ARC’s can be used to protect substrate in solar cell applications [1]. As it is a known fact that the incident radiation upon the optical material surface was separated into reflected ”R”, transmitted ”T”, absorbed ”A” and scattering fractions all determined by the refractive indices, angle of incidence and mode of polarization of incident radiation. Antireflection coatings can be range from a single layer having virtually zero reflectance at just one wavelength to a multilayer system of many layers having virtually zero reflection over a wide spectral range [3,5,6]. Various types of AR coatings have been theoretically and experimentally investigated [1-10]. Usually thin film optical coatings especially in the two atmospheric windows at the 3.2-4.8μm and 8-12μm band associated with several difficulties: in addition to the limited choice among the regular adhesion difficulties are magnified as a result of increasing thickness of the layers. For example, in changing the incident radiation wavelength from λ₀ = 4μm to λ₀ = 10μm , the thickness of each layer in a coating stack should be more than doubled. The limitation can be overcome by using homogeneously mixed materials . Jacobsson[ 11,12] has shown theoretically that the refractive index of homogeneously mixed films can be predicted by assuming that the material obeys the Lorentz-Lorenz or the Drude dispersion
Various combinations have been experimentally investigated for visible region [13,14] and the infrared [15]. Work has also been done on the homogeneously mixed films [15,16]. The structure and homogeneity of the ZnS-MgF$_2$ mixed coatings have been investigated by Yadava et al. [17]. They also studied the refractive index of the mixture for different ratios and different thickness. Several methods [3,18] are used to produce the homogeneously mixed films of various components. However, most of the work on homogeneously mixed films has been limited to the optical constants of films. This paper gives the optimum results obtained adopting Zemax for homogeneously mixed ZnS-MgF$_2$ films of various optical thickness and refractive index simultaneously varied to be function in the spectral region (3-5 μm).

2. Theory of antirefection coatings

Antirefection coatings are classified due to the number of layers and substrate selection. Single layer ARC’s is the simplest types of antirefection coatings [3]. This layer depends on the cancellation of the two reflected beams at the upper and lower of two surfaces. To achieve ARC properties a necessary refractive index condition must be satisfied. Assuming $n_0$, $n_1$ and $n_s$ the refractive indices of air, film and that of substrate, then to achieve ARC properties, the ratios of the refractive indices at each boundary should be equal, that is:

$$\frac{n_0}{n_1} = \frac{n_1}{n_s} \quad \text{with layer thickness} \quad n_1 d_1 = \lambda_0 / 4$$

$d_1$ is the geometrical thickness of the film, $\lambda_0$ the reference (design) wavelength. Reflectance profile shows only one minimum in the reflectance configuration. For broadband losses reduction, more layers are required. All design contractions have been optimized in present work adopting ZEMAX software [19] by simultaneously varied in refractive index and thickness of the individual layer in addition to their sequences.

2.1 Multilayer transfer matrix calculations

Transfer (characteristic) matrix calculations determine the spectral reflectance and transmittance profile for multilayer structures on substrate. Consider normally incident radiation, absorption free multilayer design, and optically homogenous films. Due to characteristic matrix theory, the electric $E_{m-1}$ and magnetic $H_{m-1}$ field vectors at the incident boundary of a film are related to the electric $E_m$ and magnetic $H_m$ field vectors at the boundary of the adjacent film by the product of successive matrices per layer [3]. The boundary conditions between each layer requires that the tangential components of $E$ and $H$ vectors are continuous across each boundary to the equations of wave propagation. This can be expressed in matrix form as [3]:

$$\begin{bmatrix} E_{m-1} \\ H_{m-1} \end{bmatrix} = \begin{bmatrix} \cos \delta & (i \sin \delta) / \eta_s \\ i \eta_s \sin \delta & \cos \delta \end{bmatrix} \begin{bmatrix} E_m \\ H_m \end{bmatrix}$$ (1)

The characteristic matrix (2x2) including only film properties with phase thickness specified by equation:

$$\delta = \frac{2\pi}{\lambda} nd \cos \theta$$ (2)

And $\eta_1 = \frac{n_1}{n_t}$, is the ’tilted optical admittance of the 1st layer. $n$ and $d$ are refractive index and geometrical thickness of deposited layer. $\theta$ is the angle of refraction specified with Snell’s law as
\[ \cos \theta = \sqrt{1 - \left(\frac{n_0 \sin \theta_0}{n}\right)^2} \] (3)

\( \theta_0 \) is the angle of incidence, \( n_0 \) and \( n \) are refractive indices of incident and/or material and substrate, respectively. For a single layer the matrix expression (eq.1) will be:

\[
M_1 = \begin{bmatrix} A & iB \\ iC & D \end{bmatrix}
\]

Where, \( \cos \delta = A \), \( \frac{i \sin \delta}{\eta} = B \), \( i \eta \sin \delta = C \).

For successive double layer:

\[
M = \begin{bmatrix} A_2 & iB_2 \\ iC_2 & D_2 \end{bmatrix} \begin{bmatrix} A_1 & iB_1 \\ iC_1 & D_1 \end{bmatrix}
\] (4)

After multiplication of above matrices we have:

\[
M = \begin{bmatrix} AA & BB \\ CC & DD \end{bmatrix}
\] (6)

Therefore, for an assembly \( q \) multilayer:

\[
\begin{bmatrix} E_q \\ H_q \end{bmatrix} = \prod_{m=1}^{q} M_m \begin{bmatrix} E_0 \\ H_0 \end{bmatrix}
\] (7)

With the aid of equation (7), the reflection losses and transmittance can be calculated for the assembly of multilayer as follows:

\[
T = \frac{4n_s n_0}{(n_0 AA + n_s DD)^2 + (n_0 n_s BB + CC)^2} \] (8)

\[
R = \frac{(n_0 AA - n_s DD)^2 + (n_0 n_s BB - CC)^2}{(n_0 AA + n_s DD)^2 + (n_0 n_s BB + CC)^2} \] (9)

\( n_s \) represent substrate index of refraction. Based on the matrix theory, Zemax software was adopted to design and simulate the optical performance of multilayer coatings[20].

2.2 Modeling with Zemax

Zemax program was used as technique to analysis/simulation and design/optimization of optical system and coatings. An exhaustive searching for optimum solution has been done using ZEMAX adopting least squares form [19] merit function:

\[
MF = \left( \frac{\sum W_i (V_i - T_i)^2}{\sum W_i} \right)^{1/2}
\] (10)
Where $W$, $V$ and $T$ represent the "weight of the operand", "current" and "target" values (the absolute value) and "i" indicates the operand number.

3. Results and discussion

3.1 Design construction

A certain characteristics of thin film materials are required with high transparency, homogeneity, hardness, good adhesion, low stress, high packing density and ability to survive in different deposition and environmental conditions [21-23]. In the present work, ZnS were selected as substrate and high index ($n_H = 2.25$) intermediate layer to enhanced ZnS transmittance altogether with low index MgF$_2$ ($n_L = 1.35$)[1,21]. The proposed design construction are single quarter layer and triple ($\lambda_0/4$-$\lambda_0/4$-$\lambda_0/4$) ARC’s as follows:

- air/L/ZnS and air/ LHL /ZnS

modeled at design wavelength $\lambda_0 = 4\mu m$. Figure 1 illustrate reflectance profile which will be choise as a good "initial design" in the next step.

![Fig.1 Reflectance of air/ L/ ZnS and air/ LHL /ZnS vs.wavelength compared with bare ZnS substrate, $n_H = 2.20$, $n_L = 1.35$ and $\lambda_0 = 4\mu m$.](image)

ZEMAX software package based on the characteristic matrix theory as thin film analysis, has ability to develop layers and change their positions during the optimal design procedure. The layer thickness with real refractive index were changed from operand “CODA” that constrain the coating multiplier. Controlling the values of target and weight, the program arrive nearly unity ZnS transmittance as shown in figure 2. Results depicted that wide band ARC’s over the spectral region (3-5 $\mu m$) was achieved. The final optimum design are sumerized for three proposed models as follows:

**Model (1):**

| ZnS  | 2.252 | 1.927 | 1.350 | $\lambda_0$ | 0.2871 $\lambda_0$ | 0.2423 $\lambda_0$ | 1.000 |
|------|-------|-------|-------|-------------|-----------------|-----------------|-------|

**Model (2):**

| ZnS  | 2.252 | 1.953 | 1.940 | 1.350 | 0.0730 $\lambda_0$ | 0.1546 $\lambda_0$ | 0.2402 $\lambda_0$ | 1.000 |
|------|-------|-------|-------|-------|-----------------|-----------------|-----------------|-------|

**Model (3):**

| ZnS  | 2.252 | 1.869 | 1.410 | 1.493 | 2.030 | 1.350 | 0.1773 $\lambda_0$ | 0.1951 $\lambda_0$ | 0.2879 $\lambda_0$ | 0.5034 $\lambda_0$ | 0.2326 $\lambda_0$ | 1.000 |
Fig. 2 illustrate that the spectral reflectance approach the target due to:

model (1): \((R_{\text{min}}=0.157\% \text{ & } R_{\text{max}}=0.47\%)\),

model (2): \((R_{\text{min}}=0.10\% \text{ & } R_{\text{max}}=0.42\%)\) and

model (3): \((R_{\text{min}}=0.0014\% \text{ & } R_{\text{max}}=0.21\%).\)
Fig. 2. Optimum reflectance design of mixed dielectric ARC’s for:

(a) Model (1):

|            | Zns | 1.927 | 1.350 | air     |
|------------|-----|-------|-------|---------|
| Thickness  | 2.252| 0.2871λ₀ | 0.2423λ₀ | 1.00 |

(b) Model (2):

|            | Zns | 1.953 | 1.940 | 1.350 | air     |
|------------|-----|-------|-------|-------|---------|
| Thickness  | 2.252| 0.0730λ₀ | 0.1546λ₀ | 0.2402λ₀ | 1.00 |

(c) Model (3):

|            | Zns | 1.869 | 1.410 | 1.493 | 2.030 | 1.350 | air     |
|------------|-----|-------|-------|-------|-------|-------|---------|
| Thickness  | 2.252| 0.1773λ₀ | 0.1951λ₀ | 0.2879λ₀ | 0.5034λ₀ | 0.2326λ₀ | 1.00 |

With $n_H(ZnS) = 2.20$, $n_L(MgF_2) = 1.35$ and $λ₀ = 4μm$.

3.2 Computation of refractive index of the mixture

For our investigation, we have used Lorentz-Lorenz formula to find the refractive index of the mixture, which is suitable in the case of single source evaporation. Assuming the volume of the mixture to be equal to the sum of the volumes of the components, the molar refractivity of the mixture is given by the sum of the contributions due to each components. The refractive index in this curve is given by [14],

$$n = \left[ \frac{n_1^2}{\rho_1} \left( \frac{1}{C_1} - 1 \right) + \frac{n_2^2}{\rho_2} \frac{a_2}{a_1} \right]^{1/2}$$

Where, $a_1 = \frac{1}{n_1^2 + 1}$ and $a_2 = \frac{1}{n_2^2 + 1}$, $n_1$ and $n_2$ are the refractive indices of the two materials. $\rho_1$ and $\rho_2$ are their densities ($\rho_{MgF_2} = 3.0g/cm^3$, $\rho_{ZnS} = 4.1g/cm^3$) and $C_2$ is the concentration in parts by weight of components 2. The construction of ZnS is about 0.41 parts by weight to get a mixture of $n = 1.63$ and parameters summarized in Table 1.
Table (1) Homogeneous mixed dielectric ARC’s optimum conditions.

| Layer No. | Refractive index | Geometrical thickness in terms of \( \lambda_0 \) | Concentration (parts by weight) |
|-----------|------------------|---------------------------------|---------------------------------|
|           | Model 1 | Model 2 | Model 3 | Model#1 | Model # 2 | Model#3 |
| 1st layer | 1.35 (unmixed) | 1.35 (unmixed) | 1.35 (unmixed) | 0.242 3 | 0.24 02 | 0.232 6 | (unmixed) | (unmixed) | (unmixed) |
| 2nd layer | 1.927 | 1.94 | 2.03 | 0.287 1 | 0.51 46 | 0.503 4 | \( \text{ZnS(69\%)} + \text{MgF}_2(31\%) \) | \( \text{ZnS(71\%)} + \text{MgF}_2(29\%) \) | \( \text{ZnS(82\%)} + \text{MgF}_2(18\%) \) |
| 3rd layer | - | 1.953 | 1.493 | - | 0.07 3 | 0.287 9 | - | \( \text{ZnS(73\%)} + \text{MgF}_2(27\%) \) | \( \text{ZnS(17\%)} + \text{MgF}_2(83\%) \) |
| 4th layer | - | - | 1.410 | - | - | 0.195 1 | - | - | \( \text{ZnS(9\%)} + \text{MgF}_2(81\%) \) |
| 5th layer | - | - | 1.860 | - | - | 0.177 3 | - | - | \( \text{ZnS(62\%)} + \text{MgF}_2(38\%) \) |

4. Conclusion

From result, it can be conclude that the homogeneously mixed ZnS-MgF\(_2\) films can be used for obtain various shape of AR coating in the (3-5 \( \mu \)m) region. Because many random design are computed, it is capable now to find designs which are less sensitive to manufacturing errors. The optimization with Zemax is very fast and reach the target easily. Also, we hope that such coatings can be useful in various multilayer optical interference coating for normal and oblique incidence of radiation in future. Further material doping can provide better durability in environmental tests in addition to more flexibility in the synthesis of indices.

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References

[1] Gholampour, M., A. Miri, S. I. Karanian, and A. Mohammadi. "Design and fabrication of multilayers infrared antireflection nanostructure on ZnS substrate." Acta Phys. Pol. A 136 (2019): 527-30.
[2] Mansoor, M., Abubakar, S. Zaman, Liaqat Ali and Shaheed Khan. "Design of Three-Layer Antireflection Coating for High Reflection Index Lead Chalcogenide." Materials Research-ibero-american Journal of Materials 22 (2019).
[3] Macleod, H. Angus. Thin-film optical filters.5th edition , CRC press, 2017.
[4] Asghar, M. H., M. B. Khan, and S. Naseem. "Modeling high performance multilayer antireflection coatings for visible and infrared (3-5\( \mu \)m) substrates." Semiconductor Physics Quantum Electronics & Optoelectronics 6 (2003):508-13.
[5] Rancourt, J. D. Optical thin films: user handbook. SPIE Press, 1996.
[6] Thelen, A. J. "Design of optical interference coatings 1992." In Lens and Optical Systems Design, vol. 1780, p. 17802G. International Society for Optics and Photonics, 1993.
[7] Liddell H.M, 1981. “Computer-aided techniques for the design of multilayer filters” (Bristol: Adam Hilger).
[8] Dobrowolski, J. A., and F. Ho. "High performance step-down AR coatings for high refractive-index IR materials." Applied optics 21, no. 2 (1982): 288-292.
[9] Larouche, S., and Ludvik M. "OpenFilters: an open source software for the design and optimization of optical coatings." In Optical Interference Coatings, p. WB6. Optical Society of America, 2007.
[10] Rashid, H. G., Alaa Nazar, and Muhseen SE Al-Amshani. "Design and fabrication of nano-layered optical edge filter." Journal of Materials Science and Engineering A: Structural Materials: Properties, Microstructure and Processing 2, no. 2 (2012): 203-209.
[11] Jacobsson, Roland, and John Olof Mårtensson. "Evaporated inhomogeneous thin films." Applied optics 5, no. 1 (1966): 29-34.
[12] R. Jacobsson, in Optical properties of dielectric films, N.N.Axelrod, Ed. (Electrochemical Society, New York, 1968), p. 169.
[13] Hradaaynath, R., K. N. Chopra, and O. P. Grover. "Homogeneously mixed dielectric films as double-layer antireflection coatings." Applied optics 18, no. 3 (1979): 328-330.
[14] Chopra, K. N., O. P. Grover, and R. Hradaaynath. "Antireflection coatings effective at two wavelengths simultaneously in the visible region using homogeneously mixed dielectrics." Applied optics 18, no. 11 (1979): 1750-1752.
[15] Lubrzy, I., E. Ceren, Z. Taubenfeld, and H. Zipin. "Efficient and durable AR coatings for Ge in the 8–11.5-µm band using synthesized refractive indices by evaporation of homogeneous mixtures." Applied optics 22, no. 12 (1983): 1828-1831.
[16] Chopra, K. L., Shiv K. Sharma, and V. N. Yadava. "Equivalent refractive index of multilayer films of different materials." Thin Solid Films 20, no. 2 (1974): 209-215.
[17] Yadava, V. N., S. K. Sharma, and K. L. Chopra. "Variable refractive index optical coatings." Thin Solid Films 17, no. 2 (1973): 243-252
[18] Chopra, KL (1969)."Thin Film Phenomena." McGraw-Hill, New York
[19] Radiant ZEMAX, Zemax13," Optical Design Program User's Guide,” ZEMAX Development Corporation (2014). https://my.zemax.com/en-US/
[20] Hadeel T. H, Hussein T. H. and Hayfa G. R." Optimum Design of high reflection Mirror with ZEMAX". J. Phys.: Conf. Ser. 1530, 012136 (2020):1-7.
[21] https://refractiveindex.info/
[22] Selhofer, Hubert, Elmar Ritter, and Robert Linsbod. "Properties of titanium dioxide films prepared by reactive electron-beam evaporation from various starting materials." Applied optics 41, no. 4 (2002): 756-762.
[23] G J Hawkins, Spectral Characteristics of Infrared Optical Materials and Filters, PhD Thesis, University of Reading, Department of Cybernetics (1998).
[24] Chandrashekhar Meshram, Rabha W. Ibrahim, Ahmed J. Obaid, Sarita Gajbhiye Meshram, Akshaykumar Meshram, Alaa Mohamed Abd El-Latif, Fractional chaotic maps based short signature scheme under human-centered IoT environments, Journal of Advanced Research, 2020, ISSN 2090-1232, https://doi.org/10.1016/j.jare.2020.08.015.