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A brief review of Antireflection Coating for Device Application

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Abstract: Anti Reflection Coatings Reflection Coating (ARC) reduce reflection losses and hence are essential for improvement of devices which rely on either transmission or absorption of light passing through it. ARC are thin films whose efficiency depends on two factors- their refractive index and thickness. Mathematical modelling based on Fresnel's law defines definite criteria for refractive index and thickness of ARC films The refractive index should be an optical match between the incident medium and the substrate while the thickness should meet the criterion of quarter-wavelength condition.. Further improvement of ARC is possible by structural modification, where single layer, double layer or multilayer ARC of low and high refractive index are used to curb the reflection losses for a wider range on incident spectrum. The paper discusses in details, the mechanism behind antireflection properties, single layer, double layer or multilayer anti reflection coatings and the different materials used for making anti reflection coatings. Recent work on ARC have also been reviewed.

Keywords: Anti reflection coating, Quarter wavelength condition, Reflection losses, single layer antireflection coating, double layer anti reflection coating, multilayer antireflection coating.

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1. Introduction

When light is incident at the interface between two medium, there are three possible outcomes- reflection at the interface, transmission through the second medium and absorption of light as it passes through the second medium. Devices like solar cells whose performance depends on the amount of light absorbed, or on the amount of light transmitted like sensors and lens, reflection is equivalent to loss of incident energy. Hence, minimization of reflection can increase the efficiency of such devices. This is done by coating the surface of the device by a material which is known as anti reflection coating (ARC).Anti-reflection Coating is basically **a** transparent thin film coated on a surface to suppress the surface reflections.

Reflection losses depend on both the refractive index of Anti reflection (AR) coating and also on the thickness and uniformity of the coating layers. Due to the wide and diversified application of AR coatings, they find application in an extensive range of devices ranging from Solar cells, Anti-glare display front glass, sensors, Laser scanner windows, Anti-reflective coated photography lenses, Holography applications, Automotive cockpit window and many others. [1,2]

Achieving minimal reflection losses is not, however, the only criteria device grade ARC. Besides having minimum reflectance and high transparency good ARC materials should also possesses other properties such as durability, mechanical strength, adhesion to substrate, ant soiling, scratch resistant and cost-effective production process.

2. Basics of ARC

The concept of ARC was first introduced in the 19th century, by Lord Rayleigh, when he noticed that tarnishing of glass surface increases transmission instead of decreasing it. The mathematical model that deals with reflection and transmission of light at the interface of two different media was developed by Augustine Jean Fresnel. According to Fresnel's Laws, at normal incidence, the coefficient of reflection, R or Reflectance (fraction of incident power that is reflected) is given by

$$R = \left(\frac{\eta_1-\eta_2}{\eta_1+\eta_2}\right)^2 \quad \text{-------------------------(1)}$$

where η_1 is the refractive index of the incident medium while η_2 is the refractive index of the medium in which light is transmitted. The greater the difference between refractive indices of the two media, the more will be the reflection while light will pass undeviated if both the media have same refractive index. Reflection may be minimised if a layer of a material having intermediate refractive index is placed between the two media, thereby decreasing the mismatch in refractive indices of the two media.

According to equation 1, the reflectance from a substrate having refractive index η_S at the air substrate medium, R(1) is given by

$$R(1) = \left(\frac{\eta_{air}-\eta_S}{\eta_{air}+\eta_S}\right)^2 \text{--------------------(2)}$$

Let us now consider the figure 1 given below.

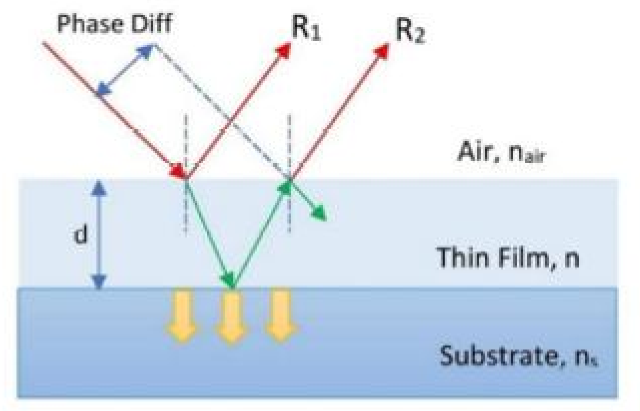

Figure 1: ***Schematics of propagation of light through a single layer***

Suppose, a thin film of refractive index $\eta$and thickness d is introduced between air (refractive index – η_{air} and substrate η_S). In this case there are two interfaces, one between air and thin film and the other between thin film and substrate.Let the reflected ray from the air-thin film interface be R_1 and the reflected ray from thin film- substrate interface be R_2.

For thin film- substrate interface, calculations are done considering the thin film and the substrate as a new substrate. The reflectance in this case, R(2) is given by

$$R(2) = \left(\frac{\eta_{air}-\frac{\eta^2}{\eta_S}}{\eta_{air}+\frac{\eta^2}{\eta_S}}\right)^2 \quad \text{------------------------------(3)}$$

To calculate zero reflectance, the following assumptions are made [3]

I) The reflected waves are considered to be of the same intensity

II) Only one reflected wave per interface is considered

III) Other optical phenomenon such as scattering and absorption are neglected.

If the thin film is designed such that R_1 and R_2 interfere destructively with each other, there will be no reflectance and the total incident ray will be transmitted into the substrate.

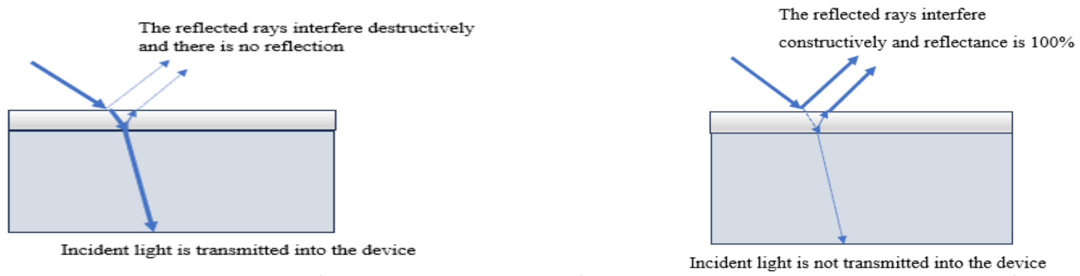

Figure 2: ***Schematics of Destructive Interference and Constructive Interference at the interface between air and thin film***

For destructive interference, the reflected rays should be out of phase by π radians or a phase difference of δ should be (m+1/2)π(where m= integer)

The relation between phase difference δ, refractive index of the film η, the angle of refraction θ, and the film thickness d is given by

$\delta = \frac{2\pi\eta d\ cos\theta}{\lambda}$ -------(4a)

If we substitute the condition of destructive interference, for normal incidence, we get in equation 4, we get

$\eta d = (m + \frac{1}{2})\frac{\lambda}{2}$--------------------(4b)

For the simplest case, m=0,

$d = \frac{\lambda}{4\eta}$--------(5)

This condition, where the optical thickness of the film is equal to the odd multiples of quarter wavelength of the incident light, is known a **quarter wave condition** and must be satisfied for antireflection.[4]

In order to achieve zero reflectance, if r=0, using equation 3, [5]we get

$\eta = \sqrt{\eta_{air} * \eta_s}$ ------------------(6)

3. How Anti-Reflection (AR) Coatings Work

Minimizing Light Reflection

Devices, particularly those made from silicon, tend to reflect a substantial portion of incoming sunlight—especially at specific wavelengths. AR coatings, typically composed of thin layers of materials such as silicon nitride or titanium oxide, are engineered to reduce this reflection.From equation 2, considering refractive index of silicon 3.4 and for the incident light having wavelength = 550 nm, the reflectance for air-silicon surface interface is almost 30%. [6] On the other hand, if a thin glass layer (η_{glass} =1.5), is placed on the silicon surface (as in Fig 1), the reflectance at the air-glass interface decreases to nearly 4%. Therefore, if a material having refractive index between 1 and 1.5 (ideally 1.23) is placed at the air-glass interface, reflection can be further reduced.

Destructive Interference Mechanism

The key to AR coatings lies in their ability to manipulate light waves. By precisely adjusting the thickness and refractive index of the coating, the reflected light waves from the top of the coating and from the surface of the device are made to interfere destructively—essentially cancelling each other out.

Enhancing Light Absorption/ transmission

With reduced reflection, more sunlight penetrates into the devices. The higher rate of absorption or transmission resulting in higher overall efficiency.

4. Challenges of ARC

ARC designed using the above model face certain challenges.

1. The basic requirement of an AR coating is that, in the visible range, total reflection should be below 0.2 – 0.5%. [7] But the quarter wave condition is applicable only for a particular wavelength, hence the ARC film are optimised only for the wavelength of the solar spectrum which has the maximum intensity (550 nm for AMI 1.5), leaving out the rest of the solar spectrum.

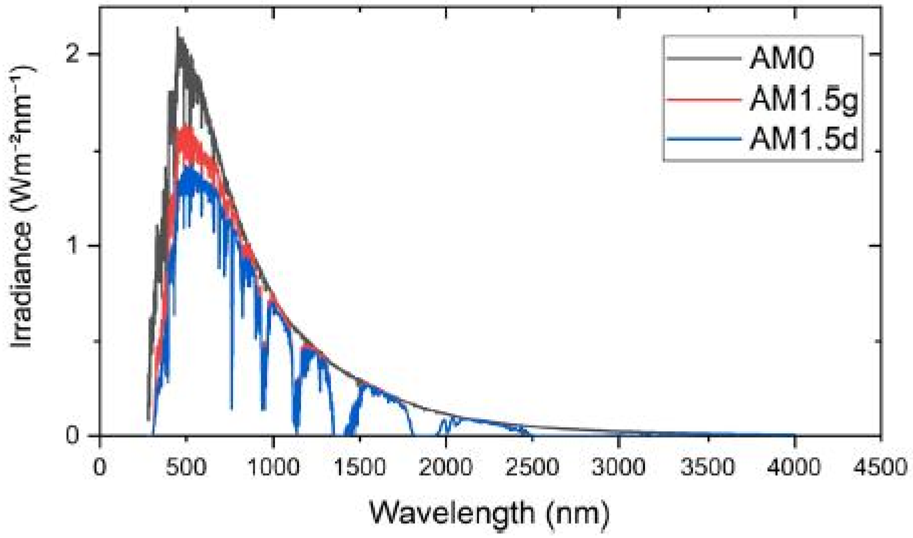

Figure 3*: A plot of solar spectrum at the top of the atmosphere (AM0) and ground level, both global (AM1.5g) and direct (AM1.5d) [8]*

2. Moreover, the above model was designed considering normal incidence only. But, as the angle of incidence increases, so does the reflectance. In fact for angle of incidence equal to 90^o(grazing incidence), the Reflectance changes to 100 %. Therefore, omni directional antireflection is another challenge to be achieved.

The other featuresdesirable in ARC are

3. Uniformity and homogeneity of ARC layers
4. High transparency
5. Environmental stability under conditions of high humidity and temperature
6. Good adhesion to the substrate so that there is no peeling off or cracks

7. The layers should be mechanically strong and durable for longevity of the ARC thin films
8. The production cost of the ARC should also be low.

In order to decrease reflection losses, the ARCs were structurally designed in any one of the following ways.

a) ARC were deposited on the device surface as single layer, double layers or multilayers such that the refractive indices of the different layers provide a better interface matching between the low refractive index of the incident medium and the high refractive index of the substrate/device.
b) The refractive index of the ARC film can be gradually changed by varying the film density, to achieve both high refracted index at one end and low refractive index at the other end.
c) Porous ARC material can be used to obtain low refractive index materials. However, such materials have low mechanical strength

Here we will discuss in detail about single layer ARC, Bilayer ARC and multilayer ARC.

5. Structural Modifications of ARC

Single layer Anti Reflection Coating (SLARC)

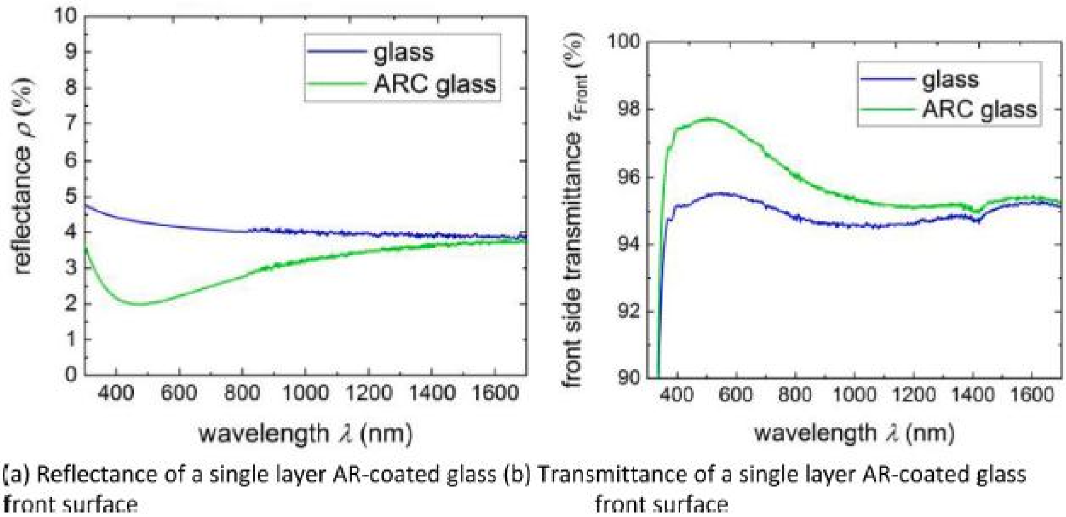

Figure 4: ***Reflectance and transmittance of a typical single layer AR coated glass. [9]***

SLARCs are optimised using quarter wave conditions to minimise reflection. The thin film material is chosen in such a way that $\eta_f > \eta_{air}$ and $\eta_s > \eta_f$; the phase reversal occurs during reflections at both the faces of the film.

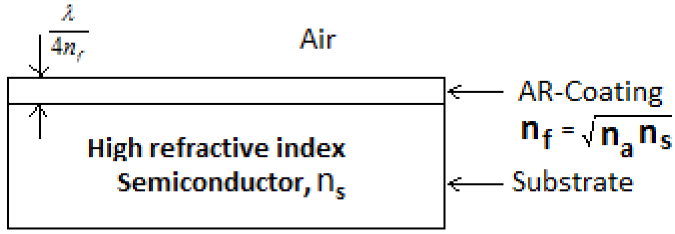

Figure 5: ***Schematics of antireflection coating on the substrate***

Effectiveness of ARC can be demonstrated by the following simple calculation. Without any coating on transparent substrate, the reflection co-efficient or reflectance (usingFresnel's formula) at the air-medium interface depends on the refractive index of air and the refractive index of the substrate, as given in equation 2

If we consider the transparent substrate to be glass, then the percentagereflectance from top surface of a transparent glass slab (η_s =1.5) for normal incidence $\{(1.5 - 1)^2/(1.5+1)^2\}\times 100\% = (0.5/2.5)^2 \times 100\% = 4\%$ and another 4% from bottom surface of glass slab. Therefore, almost 8% incident light will be reflected from glass substrate without AR-coating.

Using SLARC of quarter wave thickness, the two reflected beams are out of phase ($\delta=\pi$) and interfere destructively for the wavelength, λ. The resultant reflectance is can be expressed as a function of the refractive indices of air, the anti reflection coating and the glass substrate, as given in equation 3. Suppose a material having refractive index intermediate between air and glass (η_f = 1.4) is used ARC material. Using equation 3, reflectance from top surface of AR –coating will be

$$R = \left[\left(\frac{1-1.4}{1+1.4}\right) - \left(\frac{1.4-1.5}{1.4+1.5}\right)\right]^2 x\ 100\% = 1.8\%$$

Therefore,coating the glass surface with ARC causes a huge reduction in the reflectance of incident light.

However, SLARCs have certain serious limitations. Anti reflection properties of SLARCs are dependent on the wavelength of the incident wavelength. Though SLARCs can reduce the reflection significantly in the spectral range of 450 – 1100 nm, zero reflectance cannot be achieved. SLARCs also have another problem in the low refractive index regime. ARC with low refractive index is not very common in nature. For example, for glass substrate the refractive index of the ideal ARC should be 1.23. However, among the available materials with low refractive index, MgF_2 has the lowest. [8] This is the closest that we can approach in the low refractive index region, for SLARCs.

In order to overcome these disadvantages of SLARCs, double and then multilayer ARCs were fabricated. [10][11]

Double layer Anti Reflection Coating (DLARC)

Double layer ARC or DLARC are usually made of a bilayer of two different materials with the aim of further reduction of reflectance over a given range of wavelength or to increase the operational range of wavelengths of the ARC. The top layer of DLARC is made of a material that has a lower refractive index (η_1), while the bottom layer has a higher refractive index(η_2) closer to that of the substrate (η_S). [12]

$\eta_{air}<\eta_1<\eta_2<\eta_S$ -------------- (7)

Each layer of the bilayer must satisfy the quarter wave condition in order for the destructive interference to minimise reflection.

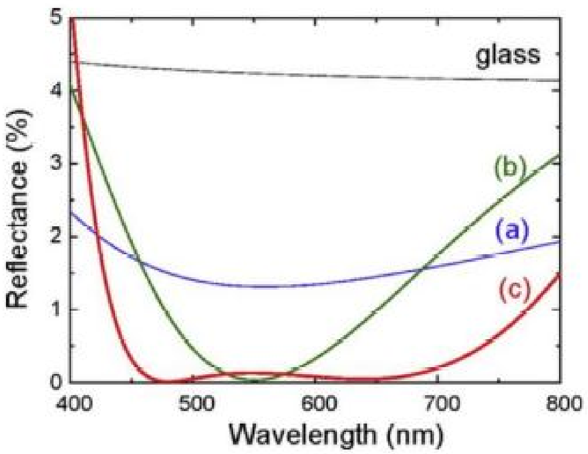

Figure 6: ***Comparison of reflectance of a) SLARC, b) DLARC, c) Three layer ARC [13].***

The criteria for the zero reflectance of a DLARC having equal optical thickness for both the layers is given by [14]

$n_1d_1 = n_2d_2 = \lambda/4$.-------------(8)

From this relation, we can get

$\frac{\eta_1}{\eta_2} = \sqrt{\frac{\eta_{air}}{\eta_S}}$ --------------------------(9)

Or

$\eta_1\eta_2 = \eta_{air}\eta_S$ ---------------------------(10)

These DLARCs are also called V coatings due to their V shaped reflectance curves, indicating zero reflectance of a given (narrow) range of wavelengths. [15,16]

While DLARCs are mainly useful in effective reduction of reflection, in most cases, for a particular range of wavelengths, the concept behind study of multiple layer ARCs was to increase transmittance over a broader range of wavelengths. MLARC are usually made of alternate high and low refractive index transparent materials in different combinations, with the material having the lowest refractive index forming the outermost layer. [17,8]

Multi Layer Anti Reflection Coating (MLARC)

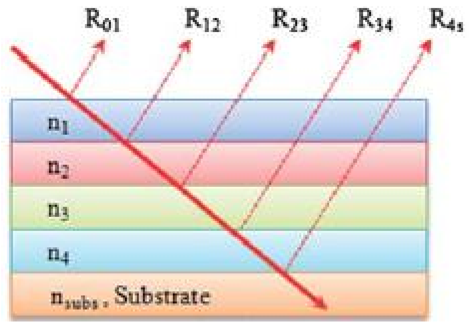

Figure 7: ***Schematics of propagation of light through multilayer films on substrates*** **[16]**

The mathematical analysis for multi-layer ARC is similar to that of SLARC (given above). However, for multi-layer ARC, vector analysis of the summation of individual reflected rays from each layer must be considered. [16]

The reflection vector for the ray reflected from the interface ij of the i^{th} and the j^{th} adjacent layers, having refractive indices η_I and η_j respectively, is given by [3].

$R_{ij} = |R_{ij}| \exp[-2(\delta_i + \delta_j)]$ -----------------------------(11)

Where $|R_{ij}| = [(\eta_i + \eta_j)/(\eta_i + \eta_j)]$

And phase difference $\delta_i = 2\pi\eta_i d_i cos\theta_i / \lambda$

Here $\theta_I$is the angle of refraction in the i^{th} layer, d_i is the thickness of the i^{th} layer

The resultant reflection vector will be the sum of the reflection vectors of the individual reflected rays from each interface. Following Figure 11, the resultant reflection vector is given by [18]

$R_{sum} = R_{01} + R_{12} + R_{23} + R_{34} + R_{4s}$ ---------------------(12)

Where $R_{01} = |R_{01}|$

And

$$R_{12} = |R_{12}| \exp[-2(\delta_1)]$$

$$R_{23} = |R_{23}| \exp[-2(\delta_1 + \delta_2)]$$

$$R_{34} = |R_{34}| \exp[-2(\delta_1 + \delta_2 + \delta_3)]$$

$$R_{4S} = |R_{4S}| \exp[-2(\delta_1 + \delta_2 + \delta_3 + \delta_4)]$$

Hence reflection losses can be minimised by significant decrease of R_{sum}. This can be achieved by adjusting the optical thickness of each layer to the quarter wave condition.

6. Anti reflection coating materials

Anti reflection coating materials can either be silicon based, metal based or polymer based. In this article, only silicon based and metal based ARC materials will be discussed.

A material having refractive index > 2 at 500 nm is usually considered as a material having high refractive index, while a material having refractive index close to that of glass (~ 1.5) is considered have low refractive index for ARC. [8] Materials with low refractive index can reduce the reflection at the air – ARC interface, while materials with high refractive index can reduce reflection at the ARC-device interface. Refractive Indices of some materials used for ARC is given in Table 1.

Table 1: ***Refractive indices of oxides used as Anti reflection Coating Material***

ARC material	Refractive Index	Reference
Porous SiO_2	1.1 – 1.5	[22]
$Si0_2$	1.46	[20]
MgF_2	1.32-1.38	[23]
Al_2O_2	1.65	
HfO_2	1.9	[24]
ZnO	2	[23]
Si_3N_4	2.05	
ZrO_2	2.15	
Ta_2O_5	2.1-2.3	
CeO_2	2.2 – 2.4	
TiO_2	2.35 -2.55	[7]

Conventionally thin films of different dielectric materials like Silica or SiO_2, as TiO_2, ZnO, ZrO_2, Ta_2O_5, HfO_2, Sr_3N_4, MgF_2, Al_2O_3 have been used for ARC fabrication. Some of these materials have low refractive index like SiO_2 and MgF_2 while some like TiO_2 thin films (η = 2.35 -2.55) have several desirable properties like excellent transmittance, uniformity, mechanical resistance, durability and low deposition cost. Zinc oxide (ZnO) has also been another popular choice as ARC material due to it's excellent optical properties and broad bandgap. Nano structuring of these materials can also modify the refractive index. For example, silicon, with high refractive index, is a material that reflects more than 35%

of light incident on it, thus incurring huge reflection loss. However, if the surface of silicon is etched, producing nano structures like nanowires, reflection losses can be drastically reduced. Silicon nanowires with low reflectance of 5.5% in the, range of 250 nm – 800nm [19] have been reported which can used as antireflection coating. Structural modification of silica to porous silica can further enhance the transmittance. Refractive index of porous silica is between 1.1 and 1.5, depending on the porosity and thickness of the film.[20] As in the case of silicon, the reflectivity of metals can be reduced by texturing the metal surface. Toma et al [21] fabricated ARC using nano structured gold thin films which exhibited reflectance (<1%) in the range 450nm-950nm. While ZnO has a refractive index of 2, nano structuring ZnO surface can reduce the refractive index to 1.46.

As discussed in the previous section, double or multilayers of ARC enhanced the antireflection properties over a broader range of the visible spectrum. SiO_2 and $TiO_2$are two of the most commonly used oxides for DLARC and MLARC. Rathanasamy et al fabricated ARC with SiO_2 and TiO_2 and demonstrated increase in transmittance from 83.9% (for ARC uncoated device) to 87.6 % , for SiO_2 SLARC, 90.3% for TiO_2 SLARC, to 95.5% for TiO_2/SiO_2DLARC.[25]

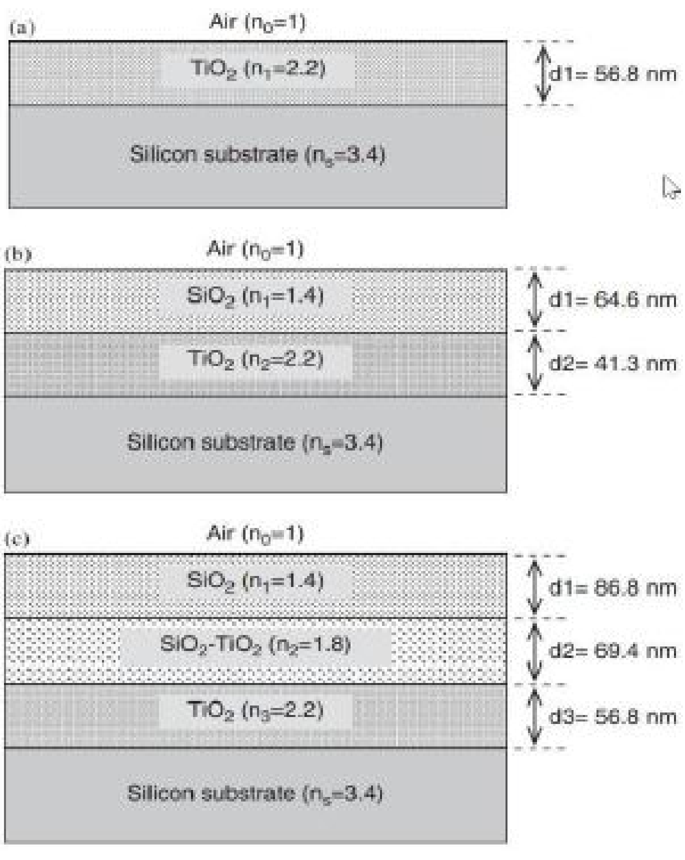

Figure 8: Schematic configuration of anti-reflective coatings on silicon using (a) TiO_2 single layer, (b) SiO_2/TiO_2 double layers and (c) SiO_2/SiO_2-TiO_2/TiO_2 triple layers. [26]

The figure above shows SLARC, DLARC and triple layer ARC as reported by Lien et al[26]. Using DLARC limited reflectance to <4 % while the triple layer ARC reduced reflectance < 2-3% within the 400-1000nm range.[26, 27]

Khuram Ali etal demonstrated that DLARC of SiO_2/ TiO_2 considerably increases the effectiveness and cost effectiveness of the devices. [28, 29]

SiO_2/TiO_2/ZnO triple layer ARC is also very effective for high transmission in the range 400-600 nm range, as reported by Khmissi et al [30]. Naif Almakayeel reported enhanced power conversion efficiency using CeO_2-WO_3 thin film ARC. [29] Low reflectivity (~ 3.3- 3.38 %) was achieved using multilayers of alumina coating and CdSeTe/CdTe thin film ARC [31] Using DLARC of MgF_2/BN on GaAs and Si substrates, a low reflectance (<5%) was achieved.[32]

Bayazit et al studied triple layers of TiO_2/HfO_2/SiO_2 ARC and demonstrated reduction of optical losses.[31].Reflection losses for Single, double and multilayers of ARC fabricated using ZnS, Si_3N_4, and MgF_2 were reported to be 34.66% for single layer, 8.47% for double layer and 5.71% for multilayer.[33]. Siyi Liu et al fabricated triple layer ARC with SiO_2 nano particle/ Al_2O_3/ TiO_2 with SiO_2 nano particles as top layer and achieved low reflectance of 5.506 %[34]

From the literature survey as detailed above, it is evident that by using the appropriate materials and by combining them in double or multilayers, can considerably reduce reflection losses to less than 5% over a considerable range of visible spectrum. The next phase of challenge in this field is to develop multifunctional materials that will combine anti reflecting properties with anti soiling(orself cleaning, anti degradation (UV filtering), good adhesion to the substrate surface, particularly flexible substrates for uniform and homogeneous films, durability and cost effectiveness.[35] Work is already underway, for example multilayers of SiO_2/WO_3/ ZnO combine antireflection properties with self cleaning properties. [33] In terms of durability, porous SLARC have been found to degrade within about five years of outdoor service, while recently developed multilayer ARC had a longer stability and low optical losses. [37, 35] As shown by recent research, specially engineered coating structures are also coming up as viable options for development of multifunctionality in ARC.[35,36]

7. Conclusion

Transparent thin film coating improvement of with good antireflection properties have become critical for improvement of optical devices which rely on absorption and transmission of light such as cameras, lenses, solar cells, and LED lighting [35].ARC have been developed using several techniques like sol gel, spin and dip coating, Chemical and Physical vapor deposition techniques, sputtering and many other processes.[29]. Each of these processes have their own advantages and disadvantages. Sol gel is a cost effective technique producing porous films with tunable refractive index for large area applications.[38]However, the thin films usually contain residual hydroxyl and / or carbon groups, take a long processing time, and often contract during processing. Chemical vapor deposition technique results in uniform high density, high purity films, having good adhesion.to the substrate. However, the process requires high temperature and elaborate equipment thus increasing the processing cost and limiting to temperature sensitive substrates. Using physical vapor deposition on the other hand is a low temperature process that results in dense good quality, low cost films with good adhesion to a wide range of substrate. Among the PVD technique, rf sputtering technique deserves special mention. Rf sputtered films are low temperature deposited, cost effective, homogeneous with significant packing density and excellent interfacial adhesion. Moreover, both conducting and nonconducting substrates can be used.

In this article, we have discussed in details, the theory behind the optical properties of anti-reflection coatings. The refractive index and the thickness of the ARC are major criteria for determining the optical properties of ARC. Single layer, bilayer, multilayer anti-reflection coatings have been discussed. While single layer ARC are simple and easy to fabricate, they show AR property only in a limited wavelength range. Double and multiple layer ARC on the other hand show AR properties over a broader range of visible spectrum. Properties of various metal and nonmetal oxides as anti-reflection coatings along with literature review have also been presented.Materials like SiO_2, MgF_2,ZnO, TiO_2, Al_2O_3, HfO_2 have frequently been used as antireflecting materials.As the refractive index plays an important role in determination of optical properties, layers of materials having different refractive indices havebeen used by different groups for ARC and low reflection losses have been reported. Recent research in ARC aims at development of multifuctionalmaterials that include properties like anti soiling, durability,good adhesion on different types of substrates along with low reflection loss.

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