

Fabrication and characterization of spectrally selective glazing dielectric multilayer structures

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We report the fabrication of three- and five-layered based TiO₂/SiO₂ dielectric structures as the back-end reflector application in thin film silicon solar cells. These dielectric structures are prepared by the combined sol-gel and spin-coating techniques. X-ray diffraction (XRD) analysis of both the three- and five-layered based structures confirmed the anatase phase of TiO₂ with its dominant peak at $2\theta = 25^\circ$. Field-emission scanning electron microscopy (FESEM) study demonstrated the formation of three and five alternate layers of TiO₂ and SiO₂ films. Comparatively, five-layered based reflector yielded the maximum (100 %) reflectance in the near-infrared (NIR) wavelength region as evidenced by the UV-Vis spectroscopy investigation.

Keywords: sol-gel method, thin films, bragg reflectors, dielectric materials.

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1. Introduction

A Bragg reflector is an optical component which is nothing but the multilayer structure of two alternating materials of distinct refractive index. This optical device has got various applications as in listed in Fig. 1. These includes as the back-end reflector in solar cells, clads in optical waveguiding, as reflector in light-emitting devices, light splitter in photonic devices and their smart applications in new-generation fabrics, heat-resistance smart windows, and windshielding in automobiles [1–3].

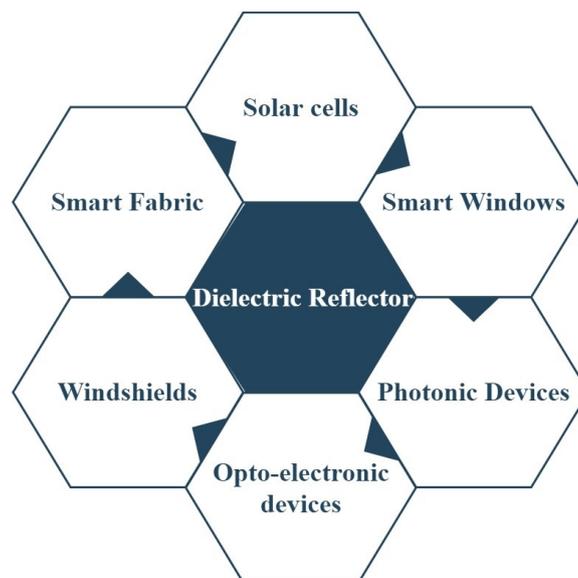


FIG. 1. Applications of dielectric reflectors

In thin film solar cells, the longer wavelength light is unabsorbed through the thin absorbing region and transmitted to the bottom-end [4]. Therefore, harvesting of light is very important in such thin film solar cells so that unabsorbed light can be reutilized. In this view, such reflectors are promising for the enhancement of light absorption in the

thin absorbing layer and hence, conversion efficiency. Other than dielectric materials such as zinc oxide, zirconium oxide, etc., the TiO_2 and SiO_2 are the best opted dielectric materials due to their high refractive index contrast and non-hazardous during the processing by the sol-gel method. These materials have also been demanded for their potential applications such as anti-reflection coating, optical waveguides, photonic crystals etc. [5]. Sol-gel spin coating approach needs optimization of the process to attain the good step coverage, uniformity and crack free films [6]. The properties of $\text{TiO}_2/\text{SiO}_2$ layered based multilayer structure can be tuned for the specific applications, i.e., as the UV, visible and IR filters including lasers. Several investigations have been reported on the fabrication of reflective and anti-reflective coatings [7–14]. Calvo et al. introduced a synthetic route in fabricating Bragg mirrors with TiO_2 thin-films by adopting the sol-gel spin coating [15]. He prepared the visible reflector with six layers of TiO_2 - TiO_2 with periodic variation of refractive index and controlled porosity. The reflector showed reflectance below 100% in the visible spectrum and further, suggested the preparation of Bragg reflectors by tuning the precursor solution. Hinczewski et al. presented the preparation of optical filters based on SiO_2 and TiO_2 multi-layers by adopting the sol-gel spin coating technique [16]. The prepared structure was composed of nine alternate layers of $\text{TiO}_2/\text{SiO}_2$ which endorsed 90 % reflectivity in near-UV region with 90%. The experimental results were also in good agreement with the theoretical investigations. Nagayoshi et al. prepared and studied the dielectric structure fabricated by using TiO_2 nanoparticles mixed in SiO_2 solution [17]. The spin coated films evidenced good reflectance (90 %) in the near-infrared region. These studies were explored by varying the number of dielectric layers, optical thickness, and the choice of coating techniques i.e. spin or dip coating. Sol-gel spin coating is an inexpensive and simple process based technique. Under the atmospheric conditions one can maintain the good homogeneity of the coating. To attain this, annealing temperature and the molar ratio of the precursors are the essential factors.

This paper mainly deals with the fabrication of three and five layered based dielectric structures of TiO_2 and SiO_2 film which serves as distributed Bragg reflector (DBR). The periodic arrangement of three- and five-consecutive layers of TiO_2 and SiO_2 films showed as much as 74 % and 100 % reflectance in the NIR region respectively. Section 2 presents the experimental process for fabricating the dielectric reflectors. The investigations are discussed in the Section 3 and Section 4 concludes the work.

2. Experimental process

Titanium Isopropoxide (TTIP) and Tetra Ethyl Ortho Silicate (TEOS) precursors procured by Sigma-Aldrich were used as the Ti and Si sources. De-ionized and ethanol procured by Changshu Hongsheng Fine Chemicals were used as the solvent while hydrochloric acid supplied by Fischer Scientific as the chelating agent. All the reagents were of analytical grade and used without any further purification. The preparation of dielectric reflectors is presented in the typical flow chart as shown in Fig. 2.

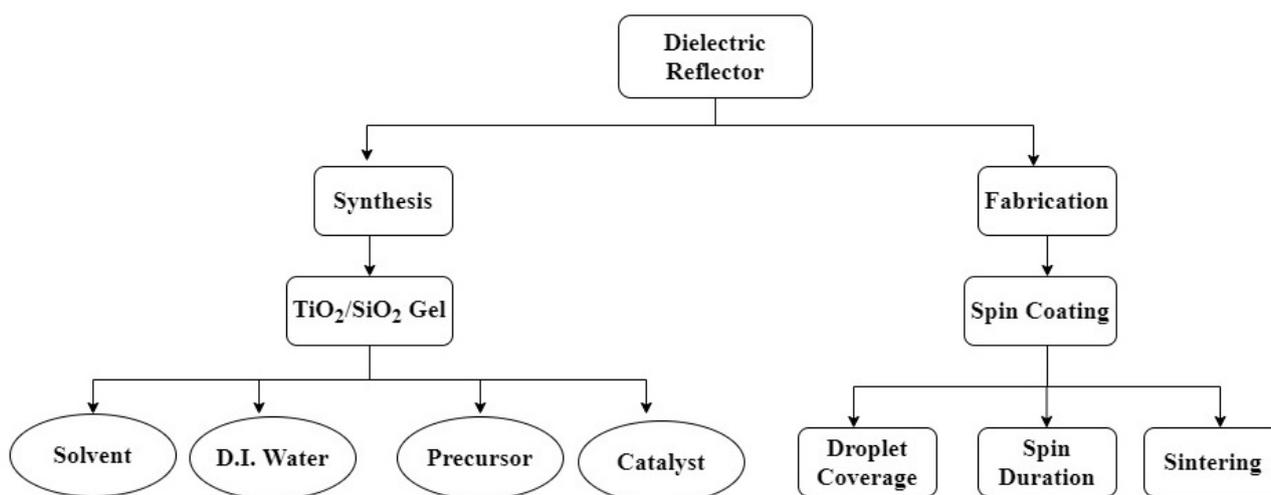


FIG. 2. Flow chart of synthesis and fabrication of dielectric reflector

To prepare the gels of TiO_2 and SiO_2 , at first 20 ml ethanol was added in 1 ml DI water and kept for 5 min stirring. Afterwards, 1 ml precursor TTIP/TEOS drop-wise was added under vigorous stirring while maintaining 2 min interval in order to prepare the $\text{TiO}_2/\text{SiO}_2$ solution. Finally 0.2 ml HCl was added to the above solutions in order to promote the rate of reaction and kept for continuous stirring for two hr. Later, both the solutions were aged for 24 hours to

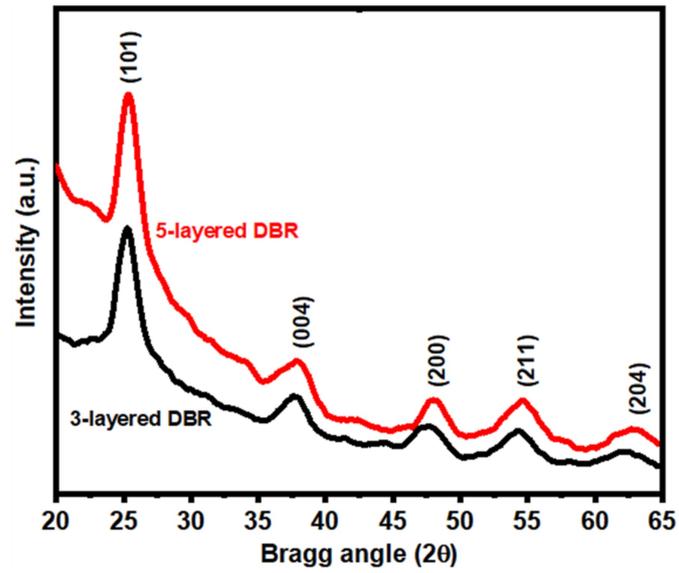


FIG. 3. XRD pattern of three and five-layered based DBR

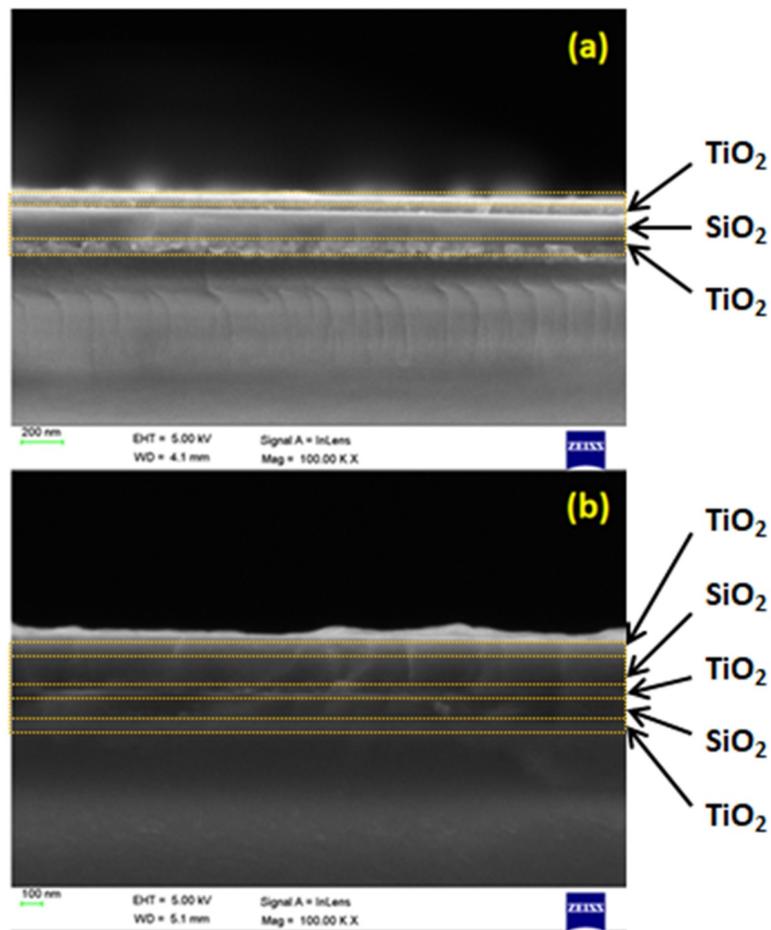


FIG. 4. (a) Cross-section FESEM images of three-layered based DBRs. (b) Cross-section FESEM images of five-layered based DBRs

get the enough viscous and transparent gels. For the preparing the multilayer structures of TiO_2 and SiO_2 films, the glass substrates were thoroughly cleaned with the soap solution and sonicated in ethanol for 30 min. Further, after rinsing in water and drying, alternate layers were deposited by the spin coating process at 3000 RPM speed for 30 sec. After deposition of each film, these were dried in the hot air oven at temperature 100°C for 30 min, in order to remove the volatile solvents. The as-prepared TiO_2 and SiO_2 films were sintered at temperature 500°C and 300°C for 1 hr to eliminate the organic compounds and to form the crystalline structure. Further, the fabricated multilayer structures were examined by using XRD, FESEM and UV-Vis spectroscopy.

3. Results and discussion

XRD study was carried out to investigate the crystallization phase presence in the spin coated three and five-layered based structures. Fig. 3 depicts the XRD pattern which endorses the anatase phase of TiO_2 .

The diffraction peaks located at $2\theta = 25^\circ, 37^\circ, 48^\circ, 55^\circ$ and 62° are indexed corresponding to the planes (101), (004), (200), (211), and (204) respectively of anatase- TiO_2 [17]. Our XRD results coincide with the JCPDS File No. 21-1272. One can notice the broadening of a Bragg peak located at $2\theta = 25^\circ$ ascribed to the plane (101) of TiO_2 , which usually indicates the existence of amorphous phase of SiO_2 . In addition, it is observed that the sequential annealing of each film did not affect the crystallinity of the multilayer structures.

Figure 4(a,b) shows the cross-section FESEM images of three and five-layered based $\text{TiO}_2/\text{SiO}_2$ reflectors. The brighter and lighter layers shown in cross-section FESEM images indicate the deposition of TiO_2 and SiO_2 film respectively. The estimated thicknesses of each layer from bottom to top were 71 nm, 127 nm and 53 nm, and 56 nm, 97 nm, 59 nm, 135 nm and 56 nm corresponding to three- and five-layered based $\text{TiO}_2/\text{SiO}_2$ reflectors. Here, we can observe the periodic structure of titania and silica films with their high and low refractive indices.

Figure 5 depicts the reflectance spectra of three- and five-layered based reflectors. The three-layered reflector showed as much as 70 % reflectance in the wavelength range from 500–1100 nm with its center wavelength 745 nm.

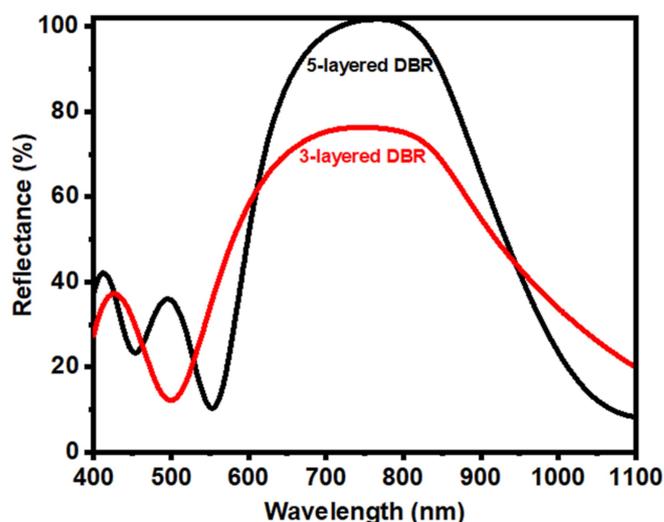


FIG. 5. Reflectance spectra of three- and five-layered based DBRs

Further, the increased layers of TiO_2 and SiO_2 films i.e. five-layered structure demonstrated the 100 % reflectance with its center wavelength 764 nm. Remarkably, both the reflectors dominated their reflectance bands in the near-infrared region. Finally, we could attain maximum 100 % reflectance with the use of only five layers of TiO_2 and SiO_2 films by depositing the films using spin coating process.

4. Conclusion

We have prepared the dielectric reflectors of $\text{TiO}_2/\text{SiO}_2$ films on glass substrates and studied the structural and optical properties. XRD results endorsed the presence of anatase- TiO_2 phase in both the samples of three- and five-layered structures of TiO_2 and SiO_2 films. Cross-section FESEM studies revealed the formation of alternate layers of TiO_2 and SiO_2 . Finally, both the reflectors characterized to study the light behavior. As compared to three-layered reflector, the five-layered one showed the maximum reflectance i.e., 100 % in the broad wavelength range with its

center wavelength 764 nm. This investigation is useful to tune the reflectivity in the desired wavelength range by adopting a simple and cost-effective fabrication technique.

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