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Non lithographic block copolymer directed self-assembled and plasma treated self-cleaning transparent coating for photovoltaic modules and other solar energy devices

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ABSTRACT

Through a combination of sol-gel based self-assembly and plasma based approach we have developed highly transparent, self-ordered, superhydrophilic and photoactive TiO_2 thin film coatings. TiO_2 sol used for such coatings comprises a block copolymer which functions as a structure directing agent. This structure directing agent aid to formation of regular pores in the TiO_2 thin film, thereby, remarkably reducing the refractive index values (~ 1.31) and enhancing the transparency (4% antireflection gain) of the coatings. Further, such porous TiO_2 coatings show an excellent ability to photo-decompose organic pollutants, due to the photocatalytic ability of such metal oxide semiconductor. Enhancement in the photocatalytic activity has been obtained by porous surface created using a block copolymer and shifting the band gap energy by incorporating nitrogen so as to utilize part of the visible region of the solar spectrum for photocatalysis. An optimum condition is achieved by varying the RF self-bias potential and time of plasma treatment. Nitrogen plasma treatment, in addition to enhancing the photocatalytic activity of TiO_2 is also found to enhance the mechanical stability and hydrophilicity, without hampering the optical transmission of coatings. Such coatings are also found to exhibit superhydrophilicity with water contact angle (WCA) < 5° under optimized condition. Thus, the coatings developed, qualify as a suitable candidate to be applied on solar PV panel and other energy devices. Treatment with nitrogen plasma extends the photocatalytic activity towards visible region of the spectrum and also ensures the mechanical stability of the otherwise porous network.

1. Introduction

With the ever rising technological advancement to leverage solar energy in the modern era, maintaining optimum performance of such energy devices has become an important issue. The major solar energy applications are in the form of photovoltaic (PV) module to generate electric power and concentrated solar thermal power (CSP). The dust fouling has an extremely detrimental effect on PV panels and it varies depending on area and its environmental conditions [1–3]. It has been reported that in dry and lower rainfall areas like Saudi Arabia that loss may range as high as 26–40% and the transmission of glass cover on PV panels reduced to 70% because of dust in summer [4,5]. Under USA SunShot CSP program, Oak Ridge National Laboratory has developed optically transparent superhydrophobic materials and coatings based on nanostructured silica surfaces that can address the soiling and maintenance issues of CSP systems by maintaining optimized adhesion, optical transmission/reflection, water and dirt repellent properties [6]. Superhydrophobic surfaces comprises of micro-/nanostructures and a low-surface energy self assembled monolayer (SAM). In order to achieve self-cleaning, the surface must have low adhesion and high water contact angle. These surfaces mimic the self-cleaning mechanism of the lotus leaf [7]. It has, thus, been observed over a period of time that superhydrophobic coatings gradually begin to fail and the performance starts to reduce, exhibiting poor self-cleaning performance than uncoated glass [8,9]. On the other hand, more durable superhydrophilic surfaces can be achieved through surface structuring, depositing nanoparticle films, or photo-induced hydrophilicity (PIH) of semiconductor films [10–12]. As the liquid film on the surface spreads

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rapidly, it can dislodge contaminants and remove them from the surface. Self-cleaning superhydrophilic coatings have been found to outperform superhydrophobic coatings at both the lab and industrial environment [9,13]. Most of the superhydrophilic coating is found to consist of TiO_2, a photocatalyst, capable of breaking down organic contaminant when exposed to light [14,15]. Some commercial hydrophilic self-cleaning coatings have also emerged in the recent past. These are mainly chemical based coatings that are coated on ceramic materials using spray technique. A well-known Japanese manufacturer, Sketch Co. has come up with nano-coating materials that provide coated substrates with a variety of functions such as anti-static, superhydrophilic, with an appreciable degree of hardness, weather resistance property and high optical transparency. Besides, there are some commercially available self-cleaning glass coatings such as Pilkington Activ (TiO_2 based hydrophilic coating) [9]. All such self-cleaning coating glass surfaces, other than possessing desired mechanical property, should also not be compromised in its high optical transparency. Other successful coating recipes, often using sol-gel process, are being used by solar panel manufacturers like Yingli Solar (CleanARC). Sprayable formulations like TitanShield (TSG80-01HD), Nanomagic (Magic Solar Coat^{SiO2}, etc. are also available. Similarly, Sentech (China) offer commercially Self-cleaning features in their monocrystalline Solar panels. Sandia National Laboratories, USA have developed a Sol-gel based coating recipe which has been licensed to Samsung for their display applications.

Our aim is to come out with a chemistry based (Sol-gel & Plasma Chemistry) highly transparent self-cleaning coating recipe that can be applied on the top side of a low iron tempered glass used in fabricating Si solar PV panels [16]. Their application again is not limited to Solar Glass cover and cost of ownership of the basic equipment will be comparatively low. Solar glass covers consists of low iron toughened glass to sustain extreme weather conditions like hail storm, dust storm, high temperature and humidity [17]. To achieve large-scale self-cleaning coatings on such energy systems several techniques have been developed and implemented that uses chemistry-based approaches such as spray coating, dip coating and aerosol deposition [1,18–20]. Other wet chemical methods which have been widely studied include layer-by-layer assembly, other variants of sol-gel process and nanoparticle coatings using suitable binder [21–24]. Sol-gel coatings were studied and applied since long in various fields such as AR coatings, optical filters, biomedical instruments, textile and ceramic industries [25].

Again one of the very promising techniques for applying self-cleaning coating is a gaseous plasma based approach which includes pretreatment of substrate under oxygen, argon and other gases in plasma state, post deposition plasma treatment, plasma based etching of substrates using different fluorinated hydrocarbons and plasma assisted aerosol vapor deposition process [26,27]. By integrating both the sol-gel and plasma-based techniques it has been possible to develop coatings which exhibit unique optical and surface properties. The basic aim is to prepare a self-cleaning coating which can either be superhydrophilic or superhydrophobic in nature with appreciable optical transmission that will either maintain or enhance the transparency of glass cover. In addition to enhancing antireflection behavior using such combined processing we could make use of photocatalytic activity of TiO_2 to degrade organic contaminants even in the presence of visible light by incorporating N atoms through plasma treatment, thereby, reducing the energy band gap of TiO_2 thin film. It may be noted that, unlike other approaches using a bilayer of SiO_2 and TiO_2, in the present study, we report a single layer structured titania film, which has been found to satisfactorily meet the desirable functional requirements of a Solar PV panel glass cover [20,28].

In order to create TiO_2 thin films with high surface area and unique optical and photocatalytic properties we used block copolymer assisted evaporation induced self-assembly (EISA) method. This method has been used since a long time to produce highly ordered colloidal arrays [29]. The method adopted by us makes use of high molecular weight triblock copolymer Pluronic F127 in combination with metal oxide based sol-gel chemistry. In this approach the block copolymer acts as a structure directing agent to impart required porosity needed to achieve enhanced optical transmission. In order to achieve the required anti-reflection behavior in such coatings there must be destructive interference of light reflecting off the two (or more) film surfaces which can be typically achieved by proper tuning of thickness and refractive index of constituent layers. Two conditions have to be fulfilled for a single layer ARC: (1) For a given wavelength (λ) and angle of incidence, the required optical thickness of the ARC must be $\lambda/4$, and (2) the amplitude matching of the two reflected beams requires an effective refractive index of the ARC (η_{AR}), which is the square-root of that of the optical substrate [30]. Most common transparent substrate material such as glass has a refractive index of around 1.5, thus requiring η_{AR}~1.22. Low iron toughened glass which has also refractive index close to common glass substrate is used to cover solar panels.

A further decrease in the refractive index can only be achieved by the introduction of porosity on the sub-wavelength length scale. Most of the research on sol-gel based approaches emphasize on porous silica-based material, offering tunable refractive index and thickness with an appreciable adhesion to glass surface. In numerous occasion it has been found that the pores have a tendency to adsorb pollutants under outdoor environment which significantly affect the optical properties of such coatings, thereby, resulting in poor antireflection behavior. Moreover, such coatings are also mechanically fragile. Absorption of organic contaminants from the ambient atmosphere results in the deterioration of photovoltaic module efficiency and affects the optimum performance and its long-term usability, requiring more frequent mechanical cleaning. Thus, for long term sustainability of PV modules photoactive metal oxide semiconductor i.e. TiO_2 is preferred as a coating to degrade the adsorbed hydrocarbons [31]. However it needs to be suitably tailored to achieve optimum AR behavior for instance by paring with silica as a mixed composition or can be used as a bottom layer coating [10,32,33]. Another elegant approach to overcome this problem is using block copolymer as a structure directing agent to prepare porous TiO_2 coatings. In addition to having desirable anti-reflecting and photoactive properties such coatings need also to show either superhydrophilic or superhydrophobic nature with WCA $< 5^\circ$ or $> 150^\circ$, as the case may be.

Faustini et al. reported double layered anti-reflecting coating using block copolymer assisted patterned TiO_2 with refractive index as low as 1.756 at 700 nm [34]. Later on Guldin et al. reported self-cleaning and low cost antireflecting coating by self-assembly of block copolymer in combination with sol-gel chemistry to obtain TiO_2 nanocrystals with regular porosity [31]. However, in most of the cases the block copolymer being used is very expensive restricting their usage to the surfaces with small area. Thus, in order to explore the possibility of block copolymer as a structure directing agent we explored various less expensive block copolymers and choose Pluronic F127 for our experiments. This triblock copolymer is not only cost efficient but also compatible with the solvent used to synthesize TiO_2 coatings by sol-gel chemistry. It is noted that *Miao et al.* have demonstrated application of porous TiO_2 coating synthesized using Pluronic F127 block copolymer in the form of double layered SiO_2-TiO_2 coating on the solar glass cover [35]. Although, with such bilayer systems they have reported an average of 3.4% gain in transmission, the self cleaning activity is noticeable only on prolong exposure to UV light irradiation. By taking motivation from this work we tried to develop single layer structured TiO_2 thin films with regular porosity and further treated it under nitrogen plasma to shift the band gap of otherwise wide band gap metal oxide semiconductor to make use of some of the visible part of the solar spectrum as well. However, due to porosity of these functionalized templated mesoporous TiO_2 thin films, optical and structural characterization is a major challenge. In order to overcome this problem Ortel et al. has proposed an approach to determine the porosity of thin films by means of electron probe microanalysis (EPMA) either by

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