Research Article

Synthesis of Neutral SiO₂/TiO₂ Hydrosol and Its Application as Antireflective Self-Cleaning Thin Film

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A neutral SiO₂/TiO₂ composite hydrosol was prepared by a coprecipitation-peptization method using titanium tetrachloride and silicon dioxide hydrosol as precursors. It is not only an antireflective self-cleaning coating material but also an environmental-benign material. Even heated at 700°C for 5 minutes in the tempering process, the as-prepared SiO₂/TiO₂ thin film still demonstrated antireflection and photocatalytic self-cleaning effect. The SiO₂/TiO₂ thin film increased near 2% of transmittance; however, the TiO₂ thin film decreased 5% of transmittance at least. In addition to antireflection, the SiO₂/TiO₂ thin film decomposed the surface coated oleic acid under ultraviolet light and showed superhydrophilicity under dark for two days. The SiO₂/TiO₂ thin film also showed good photocatalytic degradation of methylene blue. With these antireflection, persistent superhydrophilicity, and photocatalytic self-cleaning effects, this prepared neutral SiO₂/TiO₂ hydrosol would be a good coating material for tempered glass and other building materials.

1. Introduction

A solar cell or building glass with antireflective self-cleaning coating layer on the front glass would be a tendency for solar energy or building materials [1–3]. The semiconductor-based photocatalyst has been extensively studied since the discovery of electrochemical photolysis of water on TiO₂ electrodes [4]. Among various photocatalysts, TiO₂ has been widely applied to building materials because of its environmental-benign property and self-cleaning effect [1, 2]. The self-cleaning effect is caused by the photoinduced superhydrophilicity and photocatalysis on the photocatalytic layer [5]. However, the self-cleaning effect is inhibited without ultraviolet light or sunlight, and the large refractive index of anatase TiO₂, $n \sim 2.52$ would cause high reflection and reduce the transmission of visible light. In addition, the anatase TiO₂ thin film may easily transfer to rutile phase

and reduce photocatalytic activity after tempering process, heating at 700°C for 5 minutes [6, 7]. Therefore, pure TiO₂ is not a good coating material for solar cell or tempered glass.

In general, silicon dioxide is often employed as an additive to TiO₂ because of its chemical inertness, transparency to UV radiation, high thermal stability, large specific surface area, and low refractive index [8]. The SiO₂-modified TiO₂ material enhances the photocatalytic activity and increases the thermal stability [7, 9]. Zhang et al. [10, 11] used electrostatic attraction method to deposit single-layered TiO₂ particles on SiO₂ particles and formed an antireflective photocatalytic layer. In the preparation process, an electrostatic layer foamed by cationic or anionic polymer should be coated before SiO₂ or TiO₂ deposition. Liu et al. [12] and Jiang et al. [13] also used solvent-based SiO₂ and TiO₂ colloid solutions to prepare the TiO₂/SiO₂ bilayer in which TiO₂ was the upper layer and demonstrated antireflection

and higher photocatalytic activity. In contrast, Lee et al. [14] prepared a $\text{SiO}_2/\text{TiO}_2$ thin film consisting of sequential pairs of positively charged TiO_2 layer and negatively charged SiO_2 layer via layer-by-layer method without organic binder. An antireflective, antifogging, and photocatalytic thin film glass could be prepared by increasing the number of $\text{SiO}_2/\text{TiO}_2$ bilayers. Similarly, Permpoon et al. [15] found that the upper SiO_2 layer of $\text{SiO}_2/\text{TiO}_2$ bilayer showed natural hydrophilicity and stable Si-OH surface bond so that $\text{SiO}_2/\text{TiO}_2$ bilayer exhibited the optimal persistent superhydrophilicity under dark. Although literature data demonstrated excellent results, it takes many steps and energy to prepare $\text{SiO}_2/\text{TiO}_2$ or $\text{TiO}_2/\text{SiO}_2$ bilayers.

Some researchers have prepared SiO_2 - TiO_2 mixed sol via sol-gel methods, and an antireflective self-cleaning thin film could be manufactured directly [16–19]. However, the solvent-based SiO_2 - TiO_2 mixed sol is not benign to environment. Zhang et al. [20] dispersed the prepared $\text{SiO}_2/\text{TiO}_2$ powder in water by ultrasonic treatment and formed the SiO_2 -modified TiO_2 hydrosol, but their results did not show any antireflection and self-cleaning effects. However, some sediment may appear in the prepared SiO_2 -modified TiO_2 hydrosol during storage. In this study, a modified coprecipitation-peptization method was used to directly synthesize a neutral $\text{SiO}_2/\text{TiO}_2$ hydrosol using titanium tetrachloride and SiO_2 hydrosol as raw materials. The characteristics of the synthesized hydrosol were investigated, and the optical and self-cleaning effects of as-prepared thin film were evaluated. The optimal one-step-prepared $\text{SiO}_2/\text{TiO}_2$ composite thin film showed persistent superhydrophilicity, antireflection, and photocatalytic self-cleaning effect.

2. Experimental

2.1. Preparation of Neutral $\text{SiO}_2/\text{TiO}_2$ Hydrosol and Thin Film. The neutral $\text{SiO}_2/\text{TiO}_2$ hydrosol was synthesized by a modified coprecipitation-peptization method using TiCl_4 solution and SiO_2 hydrosol as precursors. Titanium tetrachloride (99%, Showa chemical) was added into deionized water at 4°C to form 1 M transparent TiCl_4 solution. In order to ensure complete hydrolysis, the NH_4OH (4 M) solution was added into the 1 M TiCl_4 solution and adjusted the pH value to 7.0 [7, 21]. A white precipitate, $\text{Ti}(\text{OH})_4$, was then obtained after aging for 2 hours. The precipitate was filtered and washed with deionized water for several times to remove ammonium and chloride ions. A proper amount of deionized water was then added to the filtered cake, and an aqueous H_2O_2 solution (30 wt%) was added to oxidize and peptize the $\text{Ti}(\text{OH})_4$ precipitate to form titanium peroxide solution. The weight ratio of $\text{H}_2\text{O}_2/\text{TiO}_2$ was controlled to be 2.0, and the peptization process should be kept for two hours with vigorous stirring [22]. The titanium peroxide solution was heated at 90°C for 4 hours, and various amount of crystalline SiO_2 hydrosol (30%, Ludox SM-30, Aldrich) was then added into the solution. The neutral transparent $\text{SiO}_2/\text{TiO}_2$ hydrosols with various $\text{SiO}_2/\text{TiO}_2$ weight ratios were obtained by heating the mixture at 90°C for 4 hours. The weight ratios of $\text{SiO}_2/\text{TiO}_2$ in the prepared $\text{SiO}_2/\text{TiO}_2$

hydrosols were 0.0, 1.5, 3.0, and 5.0, which were labeled as TiO_2 , ST_1.5, ST_3, and ST_5, respectively. And the solid concentration of TiO_2 was fixed at 1 wt% for all prepared hydrosols. For comparison, a pure SiO_2 (3 wt%) hydrosol was also prepared by directly diluting the SM-30 with deionized water.

In order to simulate the tempering process, a quartz glass, 10 cm × 6 cm × 0.15 cm, was used as the substrate, and the heating condition was controlled at 700°C for 5 minutes with increasing rate of 10°C/min. The quartz glass was immersed in 1 M NaOH solution for 30 minutes to remove oily pollutants and rinsed with deionized water for several times. After drying with air spray, a dip-coating method was used to deposit TiO_2 , SiO_2 , or $\text{SiO}_2/\text{TiO}_2$ thin film on the quartz glass with corresponding hydrosol. The quartz glass was pulled up at the rate of 10 cm/min; then, it was dried at 60°C for 5 minutes and heated at 700°C for 5 minutes in air.

2.2. Characterization of $\text{SiO}_2/\text{TiO}_2$ Hydrosol and Thin Film.

The zeta potential of various hydrosols was analyzed by Malvern Zetasizer Nano-ZS. The specific surface area of TiO_2 or $\text{SiO}_2/\text{TiO}_2$ composite material which had been heated at 700°C for 5 minutes was measured by Micromeritics ASAP 2020 using the Brunauer-Emmett-Teller (BET) method. The morphology of TiO_2 , SiO_2 , and $\text{SiO}_2/\text{TiO}_2$ particles was characterized by transmission electron microscopy (TEM: Phillips Tecnai G2 F20, 100 kV). The crystal structure of TiO_2 and $\text{SiO}_2/\text{TiO}_2$ was determined by X-ray diffractometer (XRD: Bruker AXS/D2, Cu K α radiation, 30 kV, 10 mA). The chemical bonds of $\text{SiO}_2/\text{TiO}_2$ composite materials were analyzed by Fourier transform infrared spectroscopy (FTIR: HORIBA FT-730). The topography and roughness of $\text{SiO}_2/\text{TiO}_2$ thin film were measured by atomic force microscopy (AFM: Force Precision Instrument SThM). The thickness and transmittance of coating layer on quartz glass were measured by reflectometry (Micropack NanoCalc-2000) and UV-VIS spectrometer (JASCO V-530), respectively.

2.3. Evaluation of Photocatalytic Self-Cleaning Effect

2.3.1. Contact Angle Measurement. The measurement of water contact angle is a standard method for evaluating the self-cleaning performance of photocatalytic material [23]. This method deposits oleic acid on photocatalytic plate as oily pollutants. While the oleic acid is decomposed by the photocatalytic plate, the surface would be more hydrophilic and the contact angle decreases. The water contact angle was measured by FTA 125, First Ten Angstrom, and the volume of water droplet was 1 μL .

First, the test piece should be irradiated under ultraviolet light (FL20SBLB, Sankyo Denki Co.) for 24 hours, and the intensity was adjusted at 2 mW/cm² measured by UV photometer (UV-340A, Lutron). Second, the test piece was dipped in a 0.5 vol% oleic acid solution (75093, Sigma-Aldrich Co., diluted with *n*-heptane), pulled up at a rate of 60 cm/min, and then dried at 70°C for 15 minutes. Next, the water contact angle of test piece was measured before

UV irradiation and periodically under UV irradiation (1 mW/cm^2) until less than 5° . Then, the test piece was put in dark, and the persistent hydrophilicity could be evaluated by the periodic measurement of water contact angle.

2.3.2. Decomposition of Methylene Blue. The decomposition of methylene blue in an aqueous medium is also a standard method for measuring the self-cleaning activity of photocatalytic surface [24, 25]. A modified reactor was made of acrylic glass with higher ultraviolet transmittance, and the inner size was 7 cm (length) $\times$ 1 cm (width) $\times$ 6 cm (height). It held the test liquid (35 mL, $10 \mu\text{mol/L}$ of methylene blue) and a test piece. Two UV lamps irradiated the test piece from both sides. The UV lamps and photometer were the same as contact angle measurement, but the intensity was 2 mW/cm^2 measured on both sides of the surface of the test piece. The concentration of methylene blue in test liquid was measured by a UV-VIS spectrometer (V-530, JASCO) at 664 nm every 20 minutes for 3 hours.

3. Results and Discussion

3.1. Characteristics of $\text{SiO}_2/\text{TiO}_2$ Hydrosols and Thin Films. The properties of prepared $\text{SiO}_2/\text{TiO}_2$ hydrosols and as-prepared thin films are summarized in Table 1. The pH value of TiO_2 and ST serial hydrosols was around 7 to 8 which could be considered as a neutral hydrosol. Because of their neutral property, they could be applied to various substrates without the problem of corrosion. Meanwhile, each absolute value of zeta potential was higher than 40 mV and showed good stability even in neutral condition. In our prior study [7], ST_1.5 and ST_3 hydrosols have been stored at room temperature for more than two years without any sediment. However, the hydrosol using ultrasonic treatment for suspension only could be stable for 2 weeks [26]. This stability for storage may not only be caused by the preparation method but also by the addition of SiO_2 . Because the zero point of charge was 5.5 for TiO_2 and 2.1 for SiO_2 , the surface charge of both TiO_2 and SiO_2 was negative while the pH value was around 7 to 8. The negatively charged TiO_2 and SiO_2 particles tended to separate each other and promote the stability of suspension. The TEM micrograph in Figure 1(a) indicates that the shape of TiO_2 in TiO_2 hydrosol was arrowhead-like with long axis of 30–40 nm and short axis of 6–10 nm, and Figure 1(b) shows the spherical SiO_2 particles with the size of 10 nm. However, the TiO_2 and SiO_2 hydrosols showed some aggregation. Figures 1(c) and 1(d) indicate very slender rodlike shape TiO_2 crystals (short axis of 2–4 nm) embedded deeply in spherical SiO_2 particles and well dispersed in the ST_1.5 and ST_3 hydrosols. Therefore, the addition of SiO_2 in the preparation process could decrease the particle size of TiO_2 and increase the stability of neutral $\text{SiO}_2/\text{TiO}_2$ composite hydrosols.

A high temperature calcination was usually used to promote the immobility of TiO_2 on a substrate but it would decrease the surface area and the photocatalytic activity of TiO_2 [27]. The surface area shown in Table 1 demonstrated that the surface area of $\text{SiO}_2/\text{TiO}_2$ composite material was

increased with SiO_2 content. For example, the surface area of ST_3 was almost three times that of TiO_2 . Figure 2 shows the XRD patterns of TiO_2 and $\text{SiO}_2/\text{TiO}_2$ composite materials after heating at 700°C for 5 minutes. All the four XRD patterns showed anatase TiO_2 structure, and the peak ($2\theta \sim 22^\circ$) as signed for SiO_2 was only observed in $\text{SiO}_2/\text{TiO}_2$ composite materials. Using the Scherrer's equation (i.e., $d = 0.94\lambda/\beta \cos \theta$) to calculate the crystallite size of TiO_2 based on the XRD patterns in Figure 2, it was 40.5 nm, 8.5 nm, 8.3 nm, and 7.8 nm for TiO_2 , ST_1.5, ST_3, and ST_5, respectively. On the other hand, the aggregation of TiO_2 at high temperature could be retarded by the added SiO_2 particles which suppressed the growth of crystallite size. The higher thermal stability of $\text{SiO}_2/\text{TiO}_2$ composite materials may be caused by the Ti–O–Si bond formed in the $\text{SiO}_2/\text{TiO}_2$ hydrosols, as shown in Figure 3.

In Figure 3, the FTIR spectra of TiO_2 , SiO_2 and three $\text{SiO}_2/\text{TiO}_2$ hydrosols heated at 100°C for one hour were depicted. The interaction between TiO_2 and SiO_2 in the prepared $\text{SiO}_2/\text{TiO}_2$ hydrosols was clearly shown in the Ti–O–Si bond ($\sim 970 \text{ cm}^{-1}$) [7–9]. The spectra of $\text{SiO}_2/\text{TiO}_2$ hydrosols showed that only part of SiO_2 reacted with TiO_2 to form Ti–O–Si structure. The Ti–O–Si bond improved the thermal stability of TiO_2 and suppressed the phase transformation from anatase to rutile. The band at 1100 cm^{-1} was assigned as the asymmetric Si–O–Si stretching vibration, observed for SiO_2 and all $\text{SiO}_2/\text{TiO}_2$ hydrosols. And the band at 1400 cm^{-1} was attributed to Ti–O–Ti vibration, observed for TiO_2 , ST_1.5 and ST_3 hydrosols. The spectra showed that the intensity of Ti–O–Ti band was weaker while the weight ratio of $\text{SiO}_2/\text{TiO}_2$ was higher, and it could not be measured at ST_5. The band at 1630 cm^{-1} was assigned to the bending vibration of the O–H bond of chemisorbed water. And the broad band around 3400 cm^{-1} was due to the stretching mode of the O–H bond of free water, both mainly caused by the hydrophilicity of SiO_2 . This natural wettability attributed to the upper O–H bonds on the ST serial composite thin films would prefer to adsorb moisture than oily pollutant and keep persistent superhydrophilicity.

Figure 4 depicts the transmittance spectra of SiO_2 , TiO_2 , and $\text{SiO}_2/\text{TiO}_2$ composite thin films coated on quartz glass and heated at 700°C for 5 minutes; the heating condition was similar to the tempering process. The spectrum of TiO_2 thin film showed the lowest transmittance because of its highest refractive index, 2.52. In contrast, the spectrum of SiO_2 thin film exhibited the highest transmittance and antireflection effect because of its lowest refractive index, 1.46, whereas SiO_2 thin film has no photocatalytic activity. While adding SiO_2 into the preparation of neutral TiO_2 hydrosol, the prepared neutral $\text{SiO}_2/\text{TiO}_2$ hydrosol had lower refractive index as presented in Table 1. And the as-prepared $\text{SiO}_2/\text{TiO}_2$ thin film exhibited photocatalytic activity and higher transmittance. The ST_1.5 thin film increased by 1.6% of transmittance than TiO_2 thin film at 600 nm but still lower than the substrate. When the $\text{SiO}_2/\text{TiO}_2$ weight ratio was raised to 3, the transmittance was increased by 0–1.6% as compared to that of quartz glass in the range of 440 to 800 nm. Further raised $\text{SiO}_2/\text{TiO}_2$ ratio to 5, the transmittance was raised by 2.7%, whereas some fluctuation

TABLE 1: Properties of SiO₂/TiO₂ hydrosols and as-prepared thin films.

Nomenclature	SiO ₂ /TiO ₂ hydrosol				SiO ₂ /TiO ₂ thin film				
	SiO ₂ /TiO ₂ conc. (wt%)	pH	Zeta potential (mV)	Surface area ^a S _{BET} (m ² /g)	Refractive index ^b	Transmittance at 600 nm ^c (%)	Thickness (nm)	Roughness ^d Ra (nm)	Degradation of MB (%)
TiO ₂	0.0/1.0	7.2	-49.4	55	2.52	90.5	65	4.25	54.0
ST_1.5	1.5/1.0	7.7	-42.0	122	1.81	92.1	84	6.20	64.9
ST_3	3.0/1.0	7.8	-42.2	147	1.67	94.8	165	4.04	70.2
ST_5	5.0/1.0	7.9	-41.0	177	1.60	94.4	216	2.36	90.2
SiO ₂	3.0/0.0	9.0	-45.3	—	1.46	97.8	129	0.86	9.7

^aBefore the measurement of surface area, the material should be heated at 700°C for 5 minutes first.

^bThe refractive index of ST serial bulk composite material was calculated by the molar composition of TiO₂ and SiO₂ [18].

^cThe transmittance of quartz glass at 600 nm was 93.4%.

^dThe average roughness in 5 μm × 5 μm area.

FIGURE 1: TEM micrographs: (a) TiO_2 , (b) SiO_2 , (c) ST_1.5, and (d) ST_3 hydrosol.

happened in the spectrum of ST_5 and even lower than the glass substrate during 663 to 843 nm. The thickness of ST_5 thin film was too thick, may be a reason for the fluctuation.

Previous studies [28, 29] have reported that the refractive index of an oxide thin film could be reduced by the presence of porosity, which can be explained by the equation $n_p^2 = (n_d^2 - 1)(1 - P) + 1$, where n_p and n_d are the refractive indices of the oxide on porous and nonporous states and P is the porosity percentage. Thus, the refractive index of a porous thin film could become much lower than its nonporous form. The topography of the uncoated quartz glass and coated thin films was analyzed by AFM and showed in Figure 5. The AFM 3D images showed the surface of uncoated quartz glass was much smoother than other thin-film-coated glasses. The SiO_2 thin film had a uniform porous structure; thus it exhibited the highest transmittance and antireflection effect. Although the porous structure on the ST_3 and ST_5 thin film was not so uniform, the high porosity of thin film reduced the value of n_p and thus enhanced the transmittance and led to higher antireflection effect than the substrate.

3.2. Photocatalytic Self-Cleaning Effect of $\text{SiO}_2/\text{TiO}_2$ Thin Film. The water contact angle was continuously measured

under dark after the contact angle was lower than 5° . If the photocatalytic thin film could keep the water contact angle lower than 5° for long time, it means that the thin film has better persistent superhydrophilicity. As reported in previous studies [18, 19], $\text{SiO}_2/\text{TiO}_2$ composite materials had the property of natural wettability and tended to attract moisture more than oily pollutants; thus, the surface could keep clean even in dark. Figure 6 shows the results of water contact angle measurement on TiO_2 , ST_1.5, ST_3, and ST_5 thin films. Although the initial contact angle of TiO_2 thin film was as high as 58° , TiO_2 thin film still presented the highest decomposition rate of oleic acid, and it only took 6 hours to reach 0° . The natural wettability of ST_1.5, ST_3, and ST_5 thin film suppressed the adsorption of oleic acid on the surface and exhibited lower initial contact angle of 39° , 36° , and 27° , respectively. It indicated that the natural wettability may be correlated to the $\text{SiO}_2/\text{TiO}_2$ ratio. This was consistent with the previous studies [18, 19]. However, it was very difficult to decompose the oleic acid adsorbed on SiO_2 ; thus, ST_1.5, ST_3 and ST_5 thin film took 8, 24, and 48 hours to decompose oleic acid thoroughly. When the superhydrophilic samples were stored in dark, TiO_2 thin film only kept the contact angle at 0° for 4 hours; then it increased to 30° at the 24th hour and further to 47° at the

FIGURE 2: XRD patterns of TiO_2 and ST serial composite materials heated at 700°C for 5 minutes: (a) TiO_2 , (b) ST_1.5, (c) ST_3, and (d) ST_5.

FIGURE 3: FTIR spectra of TiO_2 , SiO_2 , and ST serial composite materials heated at 100°C for one hour: (a) TiO_2 , (b) ST_1.5, (c) ST_3, (d) ST_5, and (e) SiO_2 .

48th hour. In contrast, ST_1.5, ST_3 and ST_5 all kept the contact angle at 0° for 48 hours and exhibited good persistent superhydrophilicity. This persistent superhydrophilicity is very important to the self-cleaning effect for building material or solar cell, especially in cloudy day or at night.

The wet decomposition of methylene blue is a typical method to evaluate the activity of photocatalytic thin film. Figure 7 shows the results which performed by various thin films prepared by SiO_2 , TiO_2 , and various $\text{SiO}_2/\text{TiO}_2$ hydrosols. While the SiO_2 thin film demonstrated the highest transmittance, the degradation of methylene blue was almost zero, only adsorption. In contrast, the TiO_2 thin film exhibited photocatalytic decomposition of methylene blue

FIGURE 4: Transmittance spectra of TiO_2 , SiO_2 , and ST serial composite thin films coated on quartz glass and heated at 700°C for 5 minutes.

but the transmittance is the lowest. The $\text{SiO}_2/\text{TiO}_2$ composite thin films showed higher degradation rates of methylene blue and higher transmittance. The degradation of methylene blue was enhanced with increasing the SiO_2 ratio which was attributed to the higher surface area. Furthermore, the photoinduced negative-charged surface of n-type semiconductor TiO_2 attracted the cationic dye of methylene blue and then adsorbed on the surface of SiO_2 . As presented in Table 1, ST_5 thin film had the highest surface area and thickness; thus, it performed the highest degradation ratio of methylene blue. However, the FTIR spectrum of ST_5 thin film showed the Ti-O-Ti bond was too weak to be detected. The corresponding contact angle measurement of ST_5 also reveals its lower photocatalytic activity. Thus, it could be inferred that the degradation of methylene blue over ST_5 may mainly be caused by adsorption instead of photocatalysis effect. As described in the introduction, a good antireflective self-cleaning thin film should have higher transmittance than substrate, fair photocatalytic activity and persistent superhydrophilicity. As a result, ST_3 thin film should be the optimal choice.

4. Conclusions

In this study, a neutral $\text{SiO}_2/\text{TiO}_2$ composite hydrosol was synthesized by a modified coprecipitation-peptization method using TiCl_4 and SiO_2 hydrosol as precursors. The prepared $\text{SiO}_2/\text{TiO}_2$ composite hydrosol is an environmental benign material and has good stability for keeping in ambient for more than two years. The additive SiO_2 decreased the refractive index, suppressed the aggregation of TiO_2 , and formed Ti-O-Si bond with TiO_2 . The lower refractive index of $\text{SiO}_2/\text{TiO}_2$ thin film could increase the transmittance of visible light, and the Ti-O-Si bond could retard the transformation of TiO_2 from anatase to rutile. In addition, SiO_2 particles separated TiO_2 particles and suppressed the growth of TiO_2 ; thus, $\text{SiO}_2/\text{TiO}_2$ composite material had larger surface area after treating with high temperature. Simultaneously, the natural wettability of $\text{SiO}_2/\text{TiO}_2$ thin film contributed to the persistent superhydrophilicity. While the $\text{SiO}_2/\text{TiO}_2$ weight ratio was 3, the prepared $\text{SiO}_2/\text{TiO}_2$ thin

FIGURE 5: AFM 3D images: (a) uncoated quartz glass, (b) SiO_2 , (c) ST_3 , and (d) ST_5 thin film coated on quartz glass.

FIGURE 6: Water contact angle of TiO_2 , $\text{ST}_1.5$, ST_3 , and ST_5 thin film coated with oleic acid, first 48 hours irradiated with UV, next 60 hours in dark.

film exhibited antireflection, persistent superhydrophilicity, and self-cleaning effect even treated at 700°C for 5 minutes for simulating the tempering process. The results showed

FIGURE 7: Photocatalytic decomposition of methylene blue over SiO_2 , TiO_2 , and ST serial composite thin films.

the neutral $\text{SiO}_2/\text{TiO}_2$ composite hydrosol could be a good antireflective self-cleaning coating material for solar cell or building materials.

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