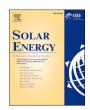
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Surface wettability effects on self-cleaning efficacy: Outdoor experimental study

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ABSTRACT

Self-cleaning coatings of photovoltaic (PV) panel and building glass have received a great deal of attention over the last two decades, using both hydrophobic and hydrophilic treatments. Here, we present a comparison study of the outdoor self-cleaning performances of superhydrophilic, hydrophobic and superhydrophobic glass coatings, over 200 days, using different tilt angles. It was found that the average transmission of all the samples linearly decreased over time after rainfall. We have defined relative self-cleaning efficacy (RSCE), using the average transmission difference of coated and uncoated samples, at a given tilt angle. We have found that the RSCE depends on both surface wettability and tilt angle. The superhydrophobic coating was found to have the best RSCE, with the superhydrophilic coating next, although both superhydrophobic and superhydrophilic coating have similar RSCE when cleaned by rainfall. Surprisingly, the RSCE is higher at a lower tilt angle for all the samples, despite a higher transmission at a higher tilt angle, suggesting that the RSCE is higher at lower tilt angles due to weaker interaction between dust particles and the surfaces.

1. Introduction

Dust accumulation and surface contamination are of great concern in situations involving window glass [1], photovoltage (PV) panels [2-6], etc., although they can be cleaned by laborious water washing, electrostatic [7,8] and mechanical [9,10] processes. However, such processes add costs. To solve this problem with minimal effort and lower costs, a self-cleaning surface concept is here proposed, using a coating surface that can remove dirt, contaminants, and other unwanted substances without the need for external cleaning methods [11,12,13,14]. This can be achieved by leveraging the effects of water or airflow. The self-cleaning surface originated by inspiration from the lotus leaf effect, which uses a superhydrophobic surface, based on its micro- and nanohierarchical morphologies and low surface energy [12,13,14]. The self-cleaning efficacy is generally associated with superhydrophobicity, including the water contact and roll-off (or slide) angles [11,14,15]. Superhydrophobicity is defined as a static water contact angle (SWCA) larger than 150° and a roll-off angle (RA/SA) less than 10°, while a surface with SWCA of less than 10° (close to 0°) is referred to as

superhydrophilic. [16,17]. This gives rise to two types of self-cleaning surfaces or coatings: hydrophilic self-cleaning and hydrophobic selfcleaning. Although most hydrophobic self-cleaning surfaces refer to superhydrophobic surfaces, while hydrophilic self-cleaning surfaces refer to superhydrophilic surfaces, there are also some surfaces constructed with hydrophobic (SCWCA, greater than 90 degrees, less than 150 degrees) surfaces. In the case of a hydrophilic coating, the water spreads spontaneously over the surface, carrying away the dirt and other impurities, whereas in the case of a hydrophobic surface, the water droplets slide and roll over the surface, thereby cleaning it. In addition, hydrophilic coatings using suitable metal oxides (e.g., TiO2) have an additional property of chemically breaking down the complex dirt deposits by a sunlight-assisted cleaning mechanism. Therefore, selfcleaning performance can be attributed to the following mechanisms: [18] (i) rainfall driven, (ii) gravity driven, (iii) wind driven, and (iv) sunlight driven(photocatalytic). In most cases, self-cleaning performance results from a combination of these effects. Currently, the performance of self-cleaning products remains uncertain since there are no specific parameters or standardized methods to evaluate them [19]

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although light transmission and water contact angles have been used to estimate the self-cleaning performance [2].

Researchers have used either the SWCA and SA or RA for hydrophobic surfaces, or the SWCA only for hydrophilic surfaces, to assess self-cleaning performance because it is directly related to rainfall cleaning. In addition, the researchers also measured the deposition of dust particles on superhydrophobic [20,21,22,23] and superhydrophilic [23,24,25,26] surfaces in laboratory environments. Especial recently Zu-Sheng Huang et al,[23] reported that the anti-soiling (dust particle deposition) effects on the surface wettability, of superhydrophilic, hydrophilic, hydrophobic and superhydrophobic coatings on glass from laboratory tests. Their results indicated that there are obvious effects of the surface wettability of the coatings on the antidust from laboratory dust particle deposition. Current published research work on selfcleaning surfaces primarily emphasizes the contribution of waterfall or rainfall effects either in outdoor or indoor. However, self-cleaning has also been linked to wind, gravity and organic contamination [27,28]. There has been little work on the combination of effects on self-cleaning. Here we investigate the self-cleaning contributions of this combination of factors, in addition to rainfall effects, by measuring the outdoor light transmission through the different samples and tilt angles.

2. Experimental

2.1. Samples preparation

Four samples, listed in Table 1, were used for testing. The glass substrate was obtained from a local market (soda-lime-silica glass) and was 2 mm thick and 30x30 cm square. The slide glass with the coatings was also used for the UV-Vis spectra measured due to the limited sample size of the UV-Vis spectrometer. It was first cleaned by ethanol sonication, to remove all oil and dust, then dried at 60 $^{\circ}$ C in oven for 1 hr. Sample A is uncoated glass; sample B is hydrophobically coated with a commercially available rainproof agent, TM101D (base catalyzed dimethoxydimethylsilane in ethanol) supplied by Solmont Technologies, which is applied by following processing: Place a few drops of the liquid on a clean glass, use a non-woven cloth or paper towel to spread it evenly over the entire surface, wait 1 min, then use a wet towel to wipe off excess liquid, rinse the surface with distilled water and dry at room temperature for 3 h or in an oven at 60° C for 1 h. Sample C is superhydrophilic coating, which is prepared with a commercially available agent, OS3100 (a TiO₂-SiO₂-PAA nanocomposite), provided by Solmont Technology; a detailed information about the nanocomposite synthesis and deposition can be found elsewhere [29]. Sample D is a commercially available superhydrophobic coating, TM101F (hydrophobic SiO2 with fluorocarbon resin binder); its preparation, durability and performance detailed information can be found elsewhere [18].

The experiment used tilt angles of 20, 45, and 80 degrees based on the following criteria. A tilt angle of 20 degrees is optimal for rooftop PV panels application, 45 degrees for most ground solar panel setups (30–45 degrees), and 80 degrees is best for curtain wall glass and building-integrated photovoltaics (BIPV) applications. The obtained data will be useful for various application targets.

2.2. Characterization

Transmittance through the glass was measured using a UV–Vis-NIR spectrophotometer (TU - 1901, Beijing PUXI General Instrument Co., LTD) and a LS116 light transmittance meter (Shenzhen Linshang Technology Co., LTD). It should be noted that the transmission data by LS116 light transmittance meter was average light transmittance from 400 to 700 nm. Surface morphology and energy dispersive spectroscopy (EDS) measurements were performed using a Hitachi S-4800 Scanning Electron Microscope with a cold field emission gun. SWCA measurements were carried out on a SL200B Static and Dynamic Optical Contact Angle Goniometer (Shanghai SOLON Information Technology Co., Ltd.) using an experimental setup that has already been described elsewhere [18,29,30,31].

Outdoor testing.

The outdoor experimental was conducted on the roof of the building located in Wuxi, Jiangsu, China. Wuxi City is located at 119°33′-120°38′ E longitude, near Shanghai, China. The local average solar radiation is 8800-20000 kJ/m²/day depending on the month of the year. Its air quality, PM2.5 and PM1.0 can be from 10 to 100, in recently 5 years from local unpublished database. Outdoor Measurements were conducted from Sept.2020 to April 2021. In order to distinguish the effect of rainfall, we chose only three rainfalls in our experiment and moved the samples indoors during the other rainfalls, which made the experiments more comparable. A schematic outdoor experimental setup is illustrated in Fig. 1. The backside of the glass was protected by saran wrap for each testing without considering the contamination effect from backside. The testing site was selected from top, middle to bottom, from left, middle to right, therefore each sample will be measured with at least 9 sites and then we estimated with average data. Both coated and uncoated glass samples were positioned 40 cm above the ground (rooftop) on a metal bracket to avoid dust from splashing onto the plates during rainfall.

3. Results and discussions

Fig. 2 shows SEM photomicrographs of all sample morphologies, indicating that both uncoated (A) and hydrophobic (B) samples are flat and have no specific surface features. Their initial SWCAs are about 33°

 Table 1

 list of the samples with different coatings and their wettability.

Sample label	A	В	С	D
description	uncoated	Hydrophobic coating	Superhydroph ilic coating	Superhydroph obic coating
Photographs of the droplet	Nation Address Assets		1	0
Static water contact angle (degree)	33	90	3	152
Slide angle	n.a.	40	n.a.	<5

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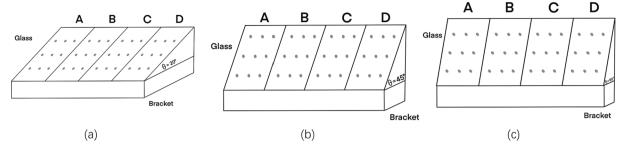


Fig. 1. A schematic outdoor experimental setup of different sample with different tilt angle θ . (a) $\theta = 20^{\circ}$, (b) $\theta = 45^{\circ}$, and (c) $\theta = 80^{\circ}$. Black dots are measured point in each sample.

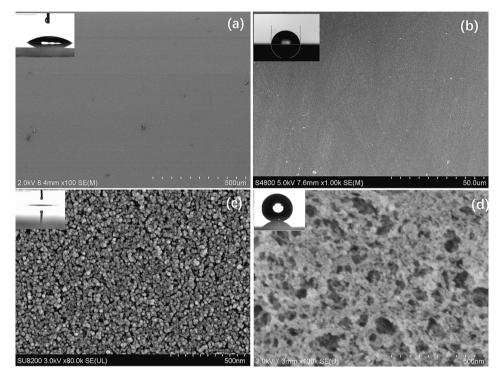


Fig. 2. Surface morphology of different samples. (a) uncoated glass A, (b) hydrophobic coating B, (c) superhydrophilic coating C, and (d) superhydrophobic coating D. Insert is water droplet on the surface.

and 90° , respectively (Table 1). Fig. 2c indicates that the surface of sample C, the superhydrophilic coating, is composed of 20–30 nm nanoparticles, with only a limited number of pores in the coating surface. Sample D, the superhydrophobic coating, is composed of 20–30 nm size nanoparticles with many pores (Fig. 2d), reaching 100 nm.

Fig. 3a displays the UV-Vis spectra of the samples. It is noted that all coated samples increased transmission, compared with the uncoated glass. This means that the coated samples have antireflection performance in the visible region. The highest transmission is for sample C (superhydrophilic coating), next is sample D (superhydrophobic coating) and there is little increase of transmission for the sample B (hydrophobic coating). The antireflective performance of the samples C and D can be attributed to the refractive index of the coatings being less than 1.45 (glass). The two samples C and D are either SiO2-TiO2-PAA nanocomposite [29] or SiO2 nanoparticles modified by methyl and fluorocarbon resin nanocomposite [18]. The refractive index of the two coatings (sample C and D) can be 1.2-1.3 due to the porosity and adding of the polymers(e.g., PAA for sample C and fluorocarbon resin for sample D)of the coatings. However, for sample B, the hydrophobic coating, due to a single molecule layer bonded on the glass surface, its thickness is less than 2 nm. For a quarter-wavelength antireflective coating of a transparent material with refractive index n and light

incident on the coating at a free-space wavelength λ , the thickness d which gives minimum reflection is estimated by

$$d = \frac{\lambda}{4n}$$

In our case, there is little antireflection for sample B because the thickness is too small (single molecule layer). While samples C and D, with about 100 nm, meet the above equation, which shows an increased transmittance.

The average transmission both for the uncoated and coated samples, between 400 and 700 nm, measured by LS116, as a function of outdoor exposure time, for different tilt angles can be found in Fig. 3b – d. We recorded the average transmission, before and after rain, three times over our observation period of over 200 days, as labeled by arrows in Fig. 3b – d. The figures also reveals that the average transmission of the samples varies with the coating and the tilt angle. The continued decrease of the average transmission for all samples before rainfall indicates the deposition of dust particles. The rainfall action plays an essential role in surface cleaning. The data presented in Fig. 3b – d indicates that there can be an average transmission difference of 4 % (observed after the first rainfall in Fig. 3b) to 35 % (observed after the

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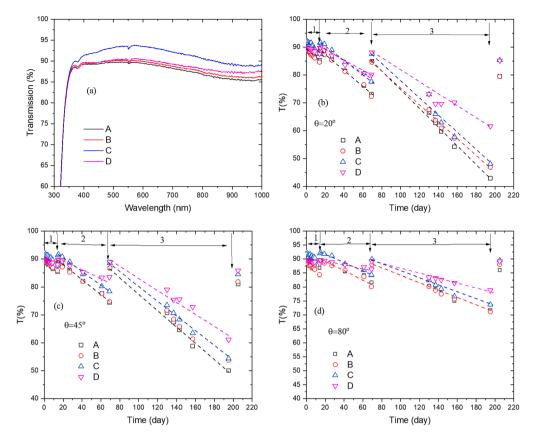


Fig. 3. UV–Vis spectra of the samples (a), a comparison of average outdoor transmission (visible region, 400–700 nm) of the samples with different treatments as a function of time, for different tilt angles (θ).(b,c,d).

third instance of rainfall in Fig. 3b), suggesting that rain cleaning has a greater impact on the transmission difference. As previously mentioned, self-cleaning surfaces can remove dust particles by using water droplets on hydrophobic/hydrophilic surfaces. However, self-cleaning mechanisms differ for superhydrophilic and superhydrophobic surfaces during rainfall or waterfall. Superhydrophilic surfaces achieve self-cleaning by promoting even spreading of water droplets due to their strong interaction with water. On hydrophobic surfaces, half-spherical droplets remove dust and dirt by sliding over the surface. A study showed that rolling droplets clean a surface more effectively than sliding droplets [32]. To improve self-cleaning performance, a hydrophobic surface should possess a high static water contact angle (SWCA), and a lower SA/RA can be helpful. A hydrophilic surface can achieve an efficient self-cleaning mechanism using a smaller SCWA surface. A careful check the change of the rainfall/waterfall effect on the coating in Fig. 3 and Fig. 4, one noted that the cleaning efficacy of both superhydrophobic

and superhydrophilic coating is better than hydrophobic and uncoated glass.

The tilt angle is also critically important to the light transmission in both coated and uncoated samples: the larger of the title angle, the greater the light transmission. The variation of the light transmission for all samples linearly decreases with time after rainfall, in the three regions of the figure. Because the light transmission of the glass, with time, is directly related to the surface dust deposition, the differences of the light transmission in different samples with time can be attributed to the surface self-cleaning effects, when compared with the uncoated surface, at a given tilt angle. The linear decreases in Fig. 3b – d, can be express as

$$T = T_0 - Kt \tag{1}$$

where T is the average transmission, T_0 is initial transmission, and K is a proportionality coefficient. At a given tilt angle, K_A , K_B and K_C (for samples A, B, and C) are almost the same, but K_D is smaller. The tilt angle

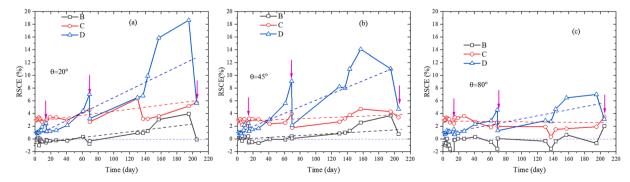


Fig. 4. Relative self-cleaning efficacy (RSCE) as a function of time for different coatings. The tilt angle is (a) $\theta = 20^{\circ}$, (b) $\theta = 45^{\circ}$, and (c) $\theta = 80^{\circ}$. The dashed line is the corresponding average.

effect on K can be found in Fig. S1, revealing that the larger the tilt angle, the smaller the value of K for a given sample.

To better evaluate the self-cleaning efficacy, we define the *relative* self-cleaning efficacy (RSCE) as follows.

$$RSCE = T_t - T_r \tag{2}$$

where, T_t is the average transmission of the coated sample, and T_r is the average transmission of the uncoated sample, for a given title angle. The variation of RSCE with time, for different tilt angles, is shown in Fig. 4. Thus, RSCE depends on both surface wettability and title angle. The hydrophobic coating has a very small self-cleaning efficacy (black dash line is average, which is near zero in Fig. 4), the superhydrophilic coating is next (average is labeled by red dash line) and the superhydrophobic coating (average is labeled blue dash line) has the best selfcleaning performance. The RSCE combines gravity, wind, and rainfall, and is the generalized self-cleaning capacity of the surface. It is obviously seen that both superhydrophobic and superhydrophilic coating have significant self-cleaning properties, even at lower tilt angle, e.g. 20°. At the same time, it is observed that the RSCE increases over time and varies according to surface wettability, as demonstrated in Fig. 4. For a tilt angle of 20°, the RSCE increases from 1 % to 18 % for the superhydrophobic surface, from 2.5 % to 6 % for the superhydrophilic coating, and from 0 % to 4 % for the hydrophobic coating. There are minor alterations for steep tilt angles, such as the 80° scenario demonstrated in Fig. 4c. Obviously, at an 80° tilt angle, the RSCE exhibits the smallest amount of 1-6 %, 3 %, and 0 for the superhydrophobic, superhydrophilic and hydrophobic coatings, respectively.

Combining Eqs. (1) and (2), we can have

$$RSCE = T_t - T_r = (T_{t0} - K_t t) - (T_{r0} - K_r t) = ((T_{t0} - T_{r0}) - (K_t - K_r)t$$
 (3)

or

$$RSCE = \Delta T_0 - \Delta Kt \tag{4}$$

where

$$\Delta T_0 = T_{t0} - T_{r0}, \text{ and } \Delta K = K_t - K_r$$
(5)

It reveals that the RSCE decrease with the time is linearly based on eq. (4). This is essentially consistent with the experimental data in Fig. 4.

Fig. 5 plots the dependence of the average transmission difference (ΔT) between the 20° tilt angles for different samples. For the same surface, the average transmission difference can be written as

$$\Delta T(\theta) = T_{\theta} - T_{20} = (T_0 - K_t(\theta)t) - ((T_0 - K_t(0)t)) = (K_{20} - K_t)t$$
(6)

or

$$\Delta T(\theta) = \Delta K(\theta)t \tag{7}$$

where $\Delta K(\theta)=K_{20}-K_t$, depending on the tilt angle. t is exposure time. It shows that the change of the average transmission, ΔT , increases linearly with time for a given coating. This agrees with Fig. 5 between two rainfalls. The ΔT corresponds to the effect of gravity on the RSCE of the sample. One can see that the larger the tilt angle, the greater ΔT with time. Additionally, ΔT is smaller for samples D than that of A, B, and C at any given time. That means that the effect of gravity is weakest for the superhydrophobic surface. This is because there is less interaction between the dust particles and the superhydrophobic surface [2,18,21,23]. Lastly, ΔT has a similar trend (Fig. 5) with time for all samples, indicating they experience similar dust pollution environments.

It is important to note that the RSCE data (3–4%) obtained from the average transmittance 200-day outdoor test (refer to Fig. 4) of the superhydrophilic coating (sample C) is consistent with the RSCE data (3

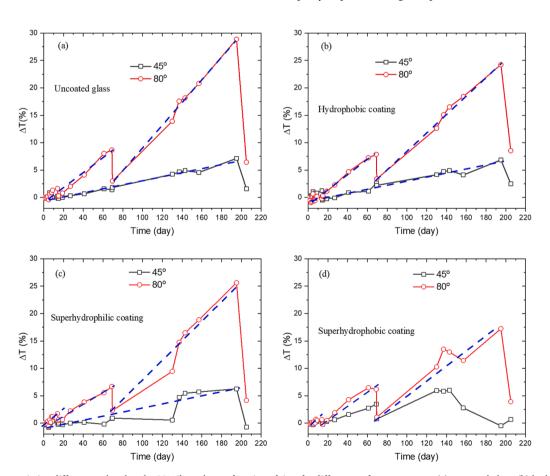


Fig. 5. Average transmission difference related to the 20° tilt angle as a function of time for different surface treatments. (a) uncoated glass, (b) hydrophobic coating, (c) superhydrophilic coating, and (d) superhydrophobic coating.

%) obtained from our recent 3-year long-term outdoor testing [28]. Similarly, the data acquired from the long-term testing of a 10-kW photovoltaic panel (with a 32-degree tilt angle and located at in Odessa, Ukraine) for 420 day, showing an increase of 2.8 % in power generation [29], is in line with our measurements. Therefore, utilizing the RSCE data obtained from the average transmittance as a quantitative evaluation method can be useful in assessing the self-cleaning performance or capability of surfaces and has reference significance.

4. Conclusions

We have evaluated the self-cleaning efficacies of different surface wettability coating, using a relative self-cleaning efficacy concept, based on the average outdoor light transmission of superhydrophilic, hydrophobic and superhydrophobic coatings over more than 200 days. We demonstrate that the *RSCE* is the greatest for the superhydrophobic coating, followed by the superhydrophilic coating; there is a small RSCE for the hydrophobic coating. Both the superhydrophobic and superhydrophilic rainfall-associated self-cleaning capacities are similar, although the superhydrophobic coating has a better *RSCE* if there is a rainfall shortage. However, the duration of the superhydrophobic coating is limited to 0.5–1 year [2], while the duration of the superhydrophilic coating can be up to more than 3 years [29].

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.solener.2023.112190.

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