

Original research article

The study on the self-cleaning property of hydrophobic polydimethylsiloxane/nano-CaCO₃ coating

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Abstract: Dust depositions always degrade the building's value due to the growth of mold and fungi. Manual cleaning is required to clean the building glass frequently which is time-consuming and costly. Self-cleaning coating is an effective and practical way to reduce dust accumulation. Using the sol-gel method, this study synthesized the self-cleaning polydimethylsiloxane (PDMS)/nano-calcium carbonate (CaCO₃) coating. The coating recorded the water contact angle (WCA) of $99.5^\circ \pm 0.5^\circ$, indicating the hydrophobic property. The embedded nanoparticles create air pockets that reduce the adhesion between water droplets and glass surface. The hydrophobic coated glass showed the self-cleaning effect where the coating achieves a low dust haze of about 32% at the sliding angle 25° . The water contact angle (WCA) of the coating system degrades to $98.2^\circ \pm 1.2^\circ$, indicating the coated glass can maintain its hydrophobic property in the real environment after 1 month of outdoor exposure. In comparison, the bare glass showed large dust haze value above 50% due to the dust accumulation impact.

Keywords: calcium carbonate; dust haze; hydrophobic; polydimethylsiloxane; self-cleaning

Received: 19 July 2023; **Accepted:** 5 September 2023; **Available online:** 1 December 2023

1. Introduction

Dust depositions and dirt promote the formation of deep watermark stains on the glass surface, especially when the location of building is near the factory's areas. In order to keep the building value, the building glass should be cleaned frequently, it is noted that the detergents create a translucent surface. Manual cleaning is time-consuming, costly, and hazardous to the environment. The use of gondolas or ropes in mid-air may face a falling risk, especially during the wind gust and earthquakes. The falling of ladders and heavy equipment would cause fatal to the workers. Several precautions must be taken to minimize the risk such as several stages of cleaning and additional cleaning period time. The stats have shown that an average of 30 cleaning workers are facing critical injuries annually in the UK^[1]. The Korea stats showed more than 15 cleaning workers lost their lives^[2], and not limited to the physical and mental health problems such as respiratory and dermatologic diseases, musculoskeletal disorders, and other conditions^[3].

The wall-cleaning robots could effectively cleanup the building automatically and reduce the dependency on the manpower. Zhang et al.^[4] have developed cleaning robot namely Sky Cleaner 3 which is employed to clean up non-uniform building structure of Shanghai Science and Technology Museum, China using brush cylinders and vacuum pump. The cleaning robots is supported by cable and equipped with the programmable logic controller (PLC) as signal sensor. However, the uses of cleaning robots are expensive and ineffective during climate changes especially heavy rain. Moon et al.^[5] have invented the cleaning tool systems with water

circulation and pulse width modulation (PWM) flow control. The focus of invention is to reduce the excessive water usage by 20%. The PWM flow control enables open-and-close control of nozzle during water spraying process, while the water circulation uses two-phase injection nozzles. The limitations of this systems are it is strongly depending on the water supply from the nozzle; besides it requires excessive electric current and heat to operate the nozzle systems.

Coating system is the reliable systems to prevent dust accumulation on the glass surface. This is because the coating system is a green technology which does not consume any energy to clear up the dust adhesion. The durability of self-cleaning coating under outdoor environment is real challenges since the impact of real dust accumulation is varied with the weather condition such as organic contaminants, rain climate and UV-radiation would deteriorate coating's hydrophobicity and functionality^[6]. From the previous studies^[7-10], the self-cleaning coating have improved the efficiency, output voltage and output current of photovoltaic (PV) panel at inclination angle around 20°. The embedded nanoparticles provide the sheeting effect in which the rain flows the dust particles and organic contaminants away, then reduce the dust accumulation on the cover glass^[11]. In comparison, the development of self-cleaning coating on outdoor building glass has been given less attention by several studies, since most of research studies focus on lab test. Prior to this issue, Chabas group^[12] have analyzed the self-cleaning effect of titanium dioxide (TiO₂) coated glass under outdoor environment for 24 months. They have observed that the dust haze values of self-cleaning glass are about 2.6% and 3.7% meanwhile the dust haze value of bare glass is about 5%. This is because the self-cleaning glass destroyed 48% of particulate organic matter (POM) compared to bare glass. Powell et al.^[13] have reported that their monoclinic vanadium(IV) oxide (VO₂)/SiO₂/TiO₂ films diminished the organic contaminants by 30%–40% on the window panel due to the photo-activation of TiO₂ photocatalyst under UVA irradiation. However, the photocatalysis reaction is restricted to sufficient irradiation of UV-light with wavelength below 380 nm^[14].

The hydrophobic coating means the coated surface has lower adhesion to dust and water on glass surfaces. The water droplets on the well-dispersed nanoparticles will roll the dust particles, dirt and other concentrated organic contaminants off glass surface easily. The developed self-cleaning coating based on hydrophobic polytetrafluoroethylene (PTFE) polymer achieves the superhydrophobic property above 150° and great self-cleaning effect^[15-20]. High hydrophobicity is attributed to the carbon-fluorine bonds of PTFE backbone which creates the hydrophobic sheets and low surface energy onto glass surface^[21]. Since the PTFE resins possess non-stick properties or low adhesion, it requires special treatment to fabricate the PTFE coating onto substrate^[22].

The hydrophobic polydimethylsiloxane (PDMS) polymer is the promising polymer which is high transparent, low toxic, great chemical stability, and price competitive materials^[23,24]. In this project, the coating developed in our lab have been synthesized by using hydrophobic polymer, polydimethylsiloxane (PDMS) and nano-calcium carbonate (CaCO₃) particles. The hydrophobic coating is developed by simple processing method that can be applied large glass panel^[25]. In comparison, the previous self-cleaning coating systems has complicated processing, applicable on small glass substrate only, translucent and requires specific equipment for drying process. The nano-CaCO₃ particles has been selected as nano-fillers because of its great compatibility within organic matrices^[26], economical materials^[27] and creates a nano-effect in binder resin^[28]. The developed coating has high potential in the commercialization industries since the novel synthesized coating can be applied on large glass panel via spray-coating technique and can be cured at ambient temperature. High transmission of coating gives large absorption of visible light for energy efficiency of building.

2. Experiment(s) and characterization(s)

2.1. Materials

Hydroxyl terminated polydimethylsiloxane (PDMS) is used as hydrophobic polymer, Silicone Elastomer, SYLGARD 184 was employed as curing agent, absolute ethanol (analytical grade), C₂H₆O is used as solvent, 3-Aminopropyltriethoxysilane (APTES) was used as binder coupling agent. The calcium carbonate, CaCO₃ nano powder with average size of 50 nm is used as fillers in the coating system.

2.2. Synthesis of PDMS/nano-calcium carbonate (CaCO₃) coating

The hydrophobic polymer, polydimethylsiloxane (PDMS) is blended with SYLGARD 184 elastomer resin at the weight ratio of 10:8. The incorporation of SYLGARD 184 as curing agent to reduce the concentration of PDMS resin to the temperature of 50 °C. The 10wt.% of PDMS/SYLGARD mixture is then stirred inside the volume of ethanol solvent at 250 mL for 1 h. The transparent ethanol solvent turns cloudy after the stirring process without left behind the insoluble PDMS/SYLGARD precipitates. The second stage is preparing the dispersed calcium carbonate (CaCO₃) nanoparticles. The 10 g of CaCO₃ nanoparticles were dispersed inside the 100 g of ethanol solvent that gives the concentration of solution is 10wt.%. The nanoparticles were dispersed inside the ultrasonic bath at 50 °C for 30 min, then subjected to stirring process inside the PDMS/SYLGARD resin at 600 rpm. The silicone binder, 3-Aminopropyltriethoxysilane (APTES) is used as coupling agent for both organic PDMS/SYLGARD resin and inorganic nano-CaCO₃. The amount of silane treatment is directly proportional to the weight percentage of nanoparticles fillers, therein 5 mL of APTES is injected slowly into prepared PDMS/nano-CaCO₃ solution and continuously stirring for another 1 h. The prepared PDMS/nano-CaCO₃ resin solution then has been fabricated on the glass substrates by dip-coating technique. The coating system is left dry at ambient temperature before subjecting to testing(s) and characterization(s).

2.3. Characterization(s)

The static water contact angle (WCA) of coating has been measured using 15EC optical contact angle machine. The 5 μL of water droplet has been dispensed at velocity of 2 μL/s by using the automatic dispensing system. Five measurements have been conducted to get the average static CA value. The surface morphology of coating system has been observed under the field emission scanning electron microscope (FESEM) at the magnification of 10 μm. The self-cleaning test has measured using the dust solution and the haze value of the coated glass has been measured to check the efficiency of self-cleaning effect. The outdoor test has been conducted for 1 months to observe the durability of glass. After the outdoor test, the WCA and surface defects has been measured respectively.

3. Result and discussion

3.1. Water contact angle (WCA) of coating system

The hydrophobicity of contact angle (WCA) of coating system has been measured as shown in **Figure 1**. The water contact angle of the bare glass has recorded at $57.6^\circ \pm 0.4^\circ$ as the bare glass possesses hydrophilic property. The hydrophilic property means the tendency of bare glass to attract the water droplets due to strong attraction force. It has been observed that the static contact angle of coating is about $99.5^\circ \pm 0.5^\circ$, indicating high hydrophobicity. The low surface tension of polydimethylsiloxane (PDMS) contains the methyl groups, CH₃ that is non-polar groups, where the non-polar CH₃ has low reaction with water. The high hydrophobicity is also supported by the embedded nano-calcium carbonate (CaCO₃) particles which create the air-pockets

within the nanoparticles. The air-pockets reduces the contact area between surface and water droplets, therein suspends the water droplet at the solid-liquid-air interface.

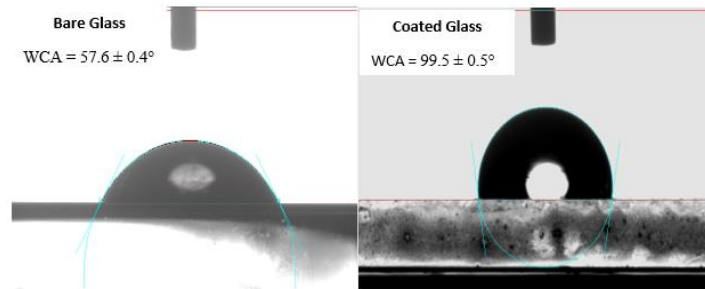


Figure 1. The water contact angle (WCA) of bare glass and PDMS/nano-CaCO₃ coating system.

3.2. Surface morphology of coating system

Figure 2a shows the images of coated surface morphology under the field emission scanning electron microscope (FESEM). The magnification has been adjusted to 10 μm. From the figure, the embedded nano-CaCO₃ particles were agglomerated in the polymer matrix as the nano-CaCO₃ has great compatibility with the organic PDMS polymer. As a result, the nano-CaCO₃ particles are dispersed well and demonstrate low visibility under the FESEM images. **Figure 2b** presents the chemical composition of coating system that been detected by energy dispersive X-ray (EDX) software. There are several elements have been detected on the coated surface namely silicon (Si), oxygen (O), carbon (C), nitrogen (N), and calcium (Ca).

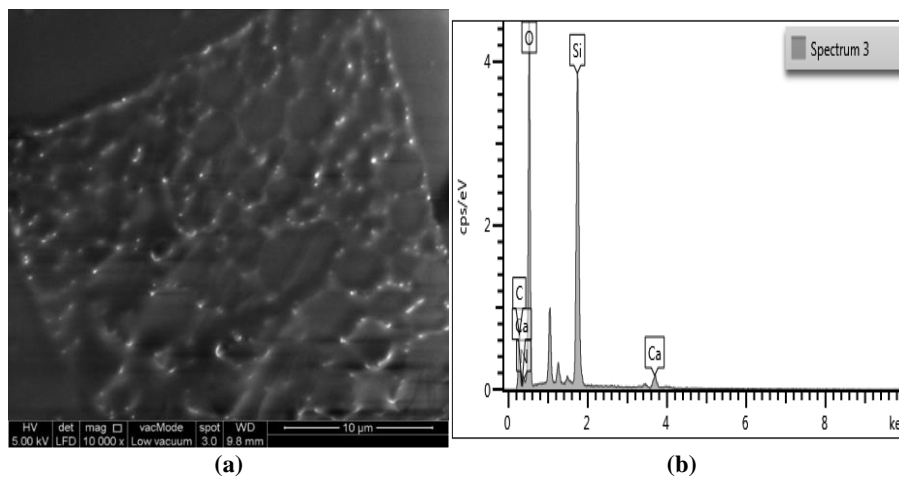


Figure 2. (a) the surface morphology; and (b) the chemical composition of PDMS/nano-CaCO₃ coating system.

Table 1. The weight percentage of chemical elements on the PDMS/nano-CaCO₃ coating system.

Element	Line type	wt.%
C	K series	16.00
N	K series	3.39
O	K series	44.69
Si	K series	31.24
Ca	K series	4.67
Total:		100.00

In the EDX spectra, the O element has recorded the highest weight percentage (wt.%) that is about 44.69%. The highest wt.% is attributed to the surface area of coating system that been covered by the siloxane bond, Si-O-Si of PDMS/SYLGARD layer. Then, it is followed by 31.24% of silicon (Si) and 16% of carbon (C) elements respectively. The presence of C contributes to the high density of methyl groups, CH₃ at the solid-liquid-air interface. As the coating surface showed the less rough surface, the presence of nanoparticles is not significantly observed under FESEM image. The wt.% of Ca and N elements has recorded about 4.67% and 3.39%, respectively. The details of wt.% of chemical elements has been presented in **Table 1**.

3.3. Self-cleaning testing

The self-cleaning test has been conducted using the concentrated ketchup solution. The concentration has been adjusted to 70wt.% by dissolving the 70 g of ketchup solution inside the 100 mL of water. The 5 mL of concentrated ketchup solution was injected onto the glass surface, both bare glass and coated glass. The ketchup solution is employed as dirt and the bare glass is used as reference. The surface energy of adhered concentrated ketchup solution is 40 mN/m. **Figure 3a,b** has presented the adhesion of dirt on the glass surface. As observed on the bare glass, the concentrated ketchup solution tends to spread as thin film layer. On the other hand, the coated glass demonstrates the hemispherical shape of dirt solution due to low adhesion contact. The trapped air pockets effectively reduce the adhesion between contaminants and coating surface.

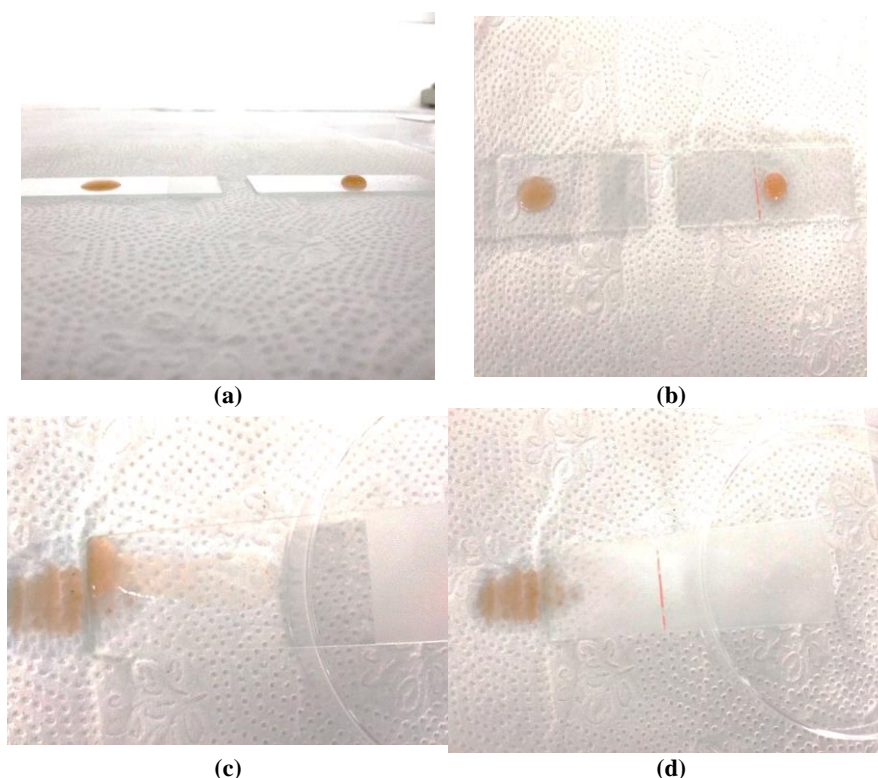


Figure 3. (a) and (b) the adhesion of concentrated ketchup solution on both glasses surface; the formation of dirt streaks on (c) bare glass surface; and (d) the coated glass surface.

The bare glass possesses the hydrophilic property which has strong adhesion force to the dirt, thereby cause the dirt streaks on the glass surface. The adhesion of dirt layer is attributed to the strong Van Der Waals Forces between the hydrophilic surfaces and dirt. In the **Figure 3d**, the hydrophobic PDMS/nano-CaCO₃ coated glass successfully expels the dirt contaminants since the rougher surface reduces the Van Der Waals

Force at the separation distance above 10 nm. The coated glass showed a clear surface without the presence of dirt streaks, indicating the hydrophobic surface has great self-cleaning effect.

3.4. Outdoor performance of coating system

The outdoor test has been performed in the UMPEDAC solar garden for 1 month. Both glass substrates have been exposed to the real environment according to Malaysian climate. Three parameters: haze value, water contact angle and surface morphology of coating system have been taken for the investigation on the self-cleaning effect and durability of coating. **Figure 4** shows the graph of dust haze value of glasses after 1 month of exposure in real environment. Three coated glasses have been used to measure the consistency of haze value and it has showed that the coated glass showed the lowest dust haze value about 21.01%. Coated glass 2 and 3 showed the dust haze value about 47.13% and 27.94%, respectively. The obtained data is different compared to the bare glass where the dust haze value has been recorded above 50%. The bare glass 1, 2, and 3 have recorded the dust haze value at 55.7%, 58.45% and 65.03%, respectively. Overall, the self-cleaning effect has taken place at the inclination angle of 25°. At the horizontal position, the glass tends to attract the dust particles due to gravity effect. The high dust haze value clearly informed the high dust concentration on the glass surface, which cannot be cleaned effectively by rain droplets.

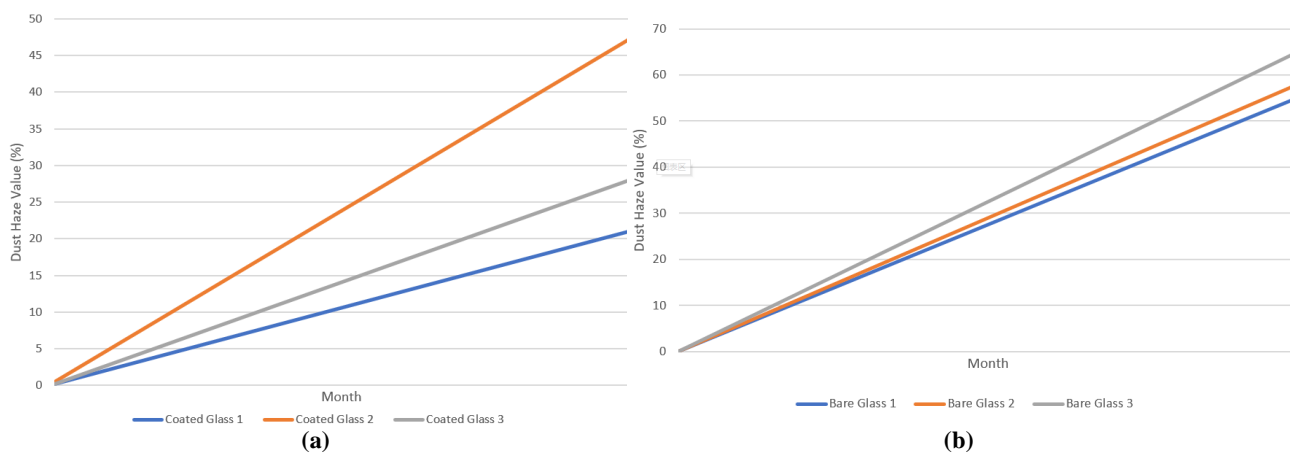


Figure 4. The graph of dust haze value over the month for (a) coated glass; and (b) bare glass.

Table 2. The dust haze values of self-cleaning glass after 1 month of outdoor exposure.

Test sample	Dust haze value (%)		Average value (%)
	Before	After	
Bare glass 1	0.1	55.70	59.72
Bare glass 2	0.1	58.45	
Bare glass 3	0.11	65.03	
Coated glass 1	0.18	21.01	32.02
Coated glass 2	0.44	47.13	
Coated glass 3	0.17	27.94	

On the other hand, the coated glass showed less dust impact since its hydrophobicity drives to great self-cleaning effect. The water droplets move in elastic motion that bounces on the surface then carries the dust away via sliding motion. The water droplet starts to slide at the inclination angle of 25°, as the rain droplets accelerate due to low surface tension and low attractive force. The average dust haze value that been recorded

by coated was 32.02%, meanwhile the bare glass has recorded the dust haze value above 59%. The details of dust haze value have been presented in the **Table 2**.

3.5. The degradation of coating system in real environments

The water contact angle (WCA) of coating system has been observed to measure the degradation in hydrophobicity after prolonged outdoor exposure as shown in the **Figure 5**. The coating system showed less degradation in WCA where the static contact angle was degraded to $98.2^\circ \pm 1.2^\circ$, with less degradation rate has been recorded about 1.305%. This is attributed to the great durability of PDMS/nano- CaCO_3 coating system in real environment since the strong silicone backbone of PDMS is not degraded by UV radiation and moisture effect. Apart from that, the PDMS/SYLGARD hydrophobic coating can retains its hydrophobic property in real outdoor environment^[29]. The heavy rainfall impact causes the liquid splashing that contributes to surface deformation at large diameter and bombardment of coated surface. The droplet spontaneously flattens then converts its kinetic energy into surface energy, that dissipated the surface energy of coating system^[30].

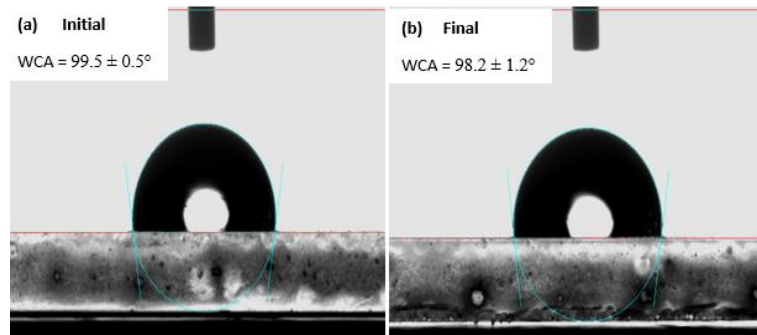


Figure 5. The WCA of coating system (a) before the outdoor exposure; and (b) after the outdoor exposure for 1 month.

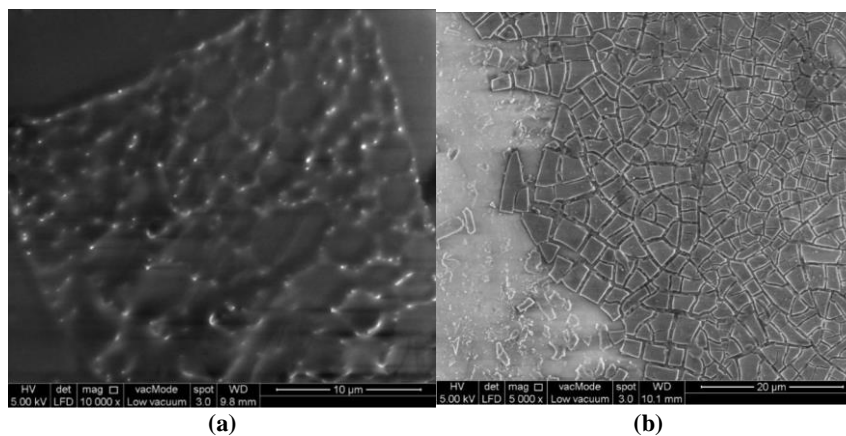


Figure 6. The surface morphology of coating system (a) before the outdoor exposure; and (b) after the outdoor exposure for 1 month.

The surface morphology of coating system after the exposure has been observed and analyzed. From the **Figure 6**, it has been observed that the coating experiences the cracking effect. Cracks propagation occurs under the prolonged hot exposure as well as the rainfall impact, the heavy rains cause moisture penetration, efflorescence, and water stains. In the long-term impact, the coating will be brittle which deteriorates its material properties. It is worth stating that the methyl groups on the hydrophobic PDMS surface ameliorates the restriction towards the moisture. The embedded nano-calcium carbonate particles suspends the fog droplets

then slide it away^[31]. Some nano-CaCO₃ interact with common pollutant such as sulfur dioxide then convert the nano-CaCO₃ into calcium sulfate, which degrades the hydrophobic property of coating system.

4. Conclusions

The hydrophobic self-cleaning coating has been developed using the hydrophobic polydimethylsiloxane (PDMS) polymer and nano-calcium carbonate (CaCO₃) particles. The hydrophobic PDMS contains the methyl groups, CH₃ that reduces the surface tension of coated glass. In addition, the embedded nanoparticles create the air-pockets within the nanostructures that ameliorates the hydrophobic property. The static water contact angle (WCA) of coating has been measured at $99.5^\circ \pm 0.5^\circ$, with the sliding angle of 25° . Its hydrophobic property drives the water droplets to slide the dust particles away. The developed PDMS/nano-CaCO₃ coating system demonstrates low dust concentration on its surface where the dust haze value is around 32% meanwhile the bare glass has recorded the dust haze value above 59%. The rainfall impact and prolonged outdoor exposure causes the cracking effect on the coated surface, then reduce the WCA of PDMS/nano-CaCO₃ coating system to $98.2^\circ \pm 1.2^\circ$. It can be concluded that the developed self-cleaning coating system has high potential in the building industry and photovoltaic (PV) panel sectors. High durability of coating in the outdoor environment increases the life-span of glass, since the coating can protect the glass from wears, rainfall impacts and dust scratches. In the PV panel sectors, this self-cleaning coating is useful to reduce dust accumulation on PV glass, then improves their electrical efficiency.

Acknowledgments

This work has been supported by UM Power Energy-Dedicated Advanced Centre (UMPEDAC) and the authors are thankful to the Higher Institution of Excellence (HICoE) Program Research Grant, UMPEDAC—2021 (MOHE HICOE-UMPEDAC) for their assistance in terms of technical, facilities and financial.

Conflict of interest

The author declares no conflict of interest.

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