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## Self – cleaning superhydrophobic coatings: Potential industrial applications

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### ABSTRACT

Many technologies have surfaced through careful observation and investigation of unusual features of various species found in Nature. Among which, the remarkable non – wetting properties of lotus leaf has hugely occupied the minds of students, researchers and industrialists from last two decades. Due to high contact angle ( $> 150^\circ$ ), water drops readily roll off the lotus leaf surface compiling dirt particles. This self – cleaning lotus effect has found huge attention in daily life. Many surfaces in day-to-day life eventually get contaminated due to the accumulation of dust/dirt or through air pollution. A huge amount of money, labor and energy is wasted in their restoration. The self – cleaning superhydrophobic coating is one of the best options for this problem. In this study, the suspension of hydrophobic silica nanoparticles was dip and/or spray coated on the body of motorcycle, building wall, mini boat, solar cell panel, window glass, cotton shirt, fabric shoes, paper (currency notes), metal, wood, sponges, plastic and marble. Every coated substrate exhibited superhydrophobicity with water contact angle nearly  $160^\circ$  and sliding angle less than  $6^\circ$ . The self-cleaning performance of the superhydrophobic coating applied on various substrates was thoroughly evaluated. The specific purpose of this article is to explore the possible industrial applications of self – cleaning superhydrophobic coatings.

### 1. Introduction

The word ‘superhydrophobic’ has now become well – known to common people. On the superhydrophobic surfaces like lotus leaf, water drops refuse to stay and roll off at slight tilting due to very high contact angle ( $> 150^\circ$ ). The extreme non – wettability of lotus leaf is due to its low surface energy along with rough hierarchical micro/nanostructure. The air trapped inside the rough protrusions disables water to impregnate by maintaining stable liquid – air interface with minimum solid fraction in contact with water drop [1]. Unlike on normal smooth surface (glass), the rolling water drops compile the dust from rough superhydrophobic surface performing self – cleaning phenomena. Apart from researchers, the self – cleaning ability of superhydrophobic surface (*Lotus Effect*) is attracting the attention of common people also. This self – cleaning superhydrophobic coating can be applied on many day-to-day used surfaces like windshields of vehicles, entire body of vehicles, window and door glasses, skyscrapers, solar cell panels, fabrics, sport shoes, metals, papers, sponges, woods, marbles and the list is ceaseless [2–5].

Over the last two decades, tremendous research work has been pursued in developing the superhydrophobic surfaces for efficient self – cleaning applications. So far, numerous effective methods to prepare superhydrophobic coatings have been investigated [6]. Some methods are fast, easy and economical, whereas few needs costly chemicals and exclusive equipment. Hence, the topic of discussion here are the various interesting research work on the fabrication of superhydrophobic coatings on different substrates using simple and robust techniques. The transparent and durable superhydrophobic coatings on glass finds high demand in industrial sectors. Xu and He [7] prepared highly transparent superhydrophobic coating on glass using modified hollow silica nanoparticles. The hollow silica nanoparticles modified by aminopropyltriethoxysilane were dip – coated on glass substrates and post surface chemical modification was completed by chemical vapor deposition of perfluorooctyltrimethoxysilane. Liu et al. [8] utilized sol – gel processing of long chain (Heptadecafluoro-1,1,2,2-tetrahydrodecyl) trimethoxysilane (17FTMS) and obtained transparent superhydrophobic film on glass substrate by simple dip coating. Yang et al. [9] prepared transparent superhydrophobic coating by simple spin or

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dip coating the fluorosilane modified silica nanoparticles on glass. In addition, the self-cleaning superhydrophobic fabrics are also greatly appealing to the researchers. Gao et al. [10] prepared superhydrophobic cotton and polyester fabrics by depositing silica sol and subsequent surface modification by hexadecyltrimethoxysilane (HDTMS). The superhydrophobicity of the fabrics was preserved for almost 30 laundering cycles. Xue and his research group [11] deposited  $\text{TiO}_2$  sol on cotton fabric and its surface energy was lowered using stearic acid to achieve superhydrophobicity. Moreover, this superhydrophobic cotton fabric revealed good UV-shielding characteristics.

The corrosion of metallic materials is one of the crucial challenges and that can be inhibited by applying superhydrophobic coating. Wu and researchers [12] adopted single step electrodeposition technique to deposit hybrid silica sol-gel film on mild steel (MS). Through finely adjusting the deposition potential and time, the superhydrophobicity was achieved which revealed the improved corrosion resistance in NaCl solution. Bayer et al. [13] have spray deposited four to six alternating layers of perfluoroalkylmethacrylic copolymer (PMC) and hydrophobic silica nanoparticles on aluminum substrate and the silica nanoparticles were silica nanoparticles were embedded into the polymer matrix by inducing thermal treatment after each alternating layers. The prepared superhydrophobic coating revealed excellent mechanical durability and effectively prevented aerodynamic insect fouling. Bayer and researchers [14] also used thermal welding treatment to attain highly abrasion resistant superhydrophobic silica nanoparticle-polymer coating on aluminum substrates. Our research group [15] lowered the surface energy of stainless steel (SS) by etching it in sulfuric acid and modifying with methyltrichlorosilane. The superhydrophobic SS showed excellent self-cleaning and anti-corrosive properties. Furthermore, the wettability of the coating was tested and found to be intact under harsh bending, water jet impact and abrasion. Cellulose papers are prone to capture moisture from surroundings and eventually become wet. Ogi-hara et al. [16] obtained transparent and superhydrophobic paper by simply spray coating the suspension of hydrophobic silica nanoparticles on the hydrophilic paper. The effect of different alcohols on the superhydrophobicity of the silica coating was studied, whereas the coatings with silica nanoparticles suspension in ethanol exhibited superhydrophobicity due to the short length of hydrocarbon chain in ethanol. The superhydrophobic paper maintained its wetting state after bare finger pressing, rolling and folding. The same research group [17] has hydrophobically modified the  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  nanoparticles using silane coupling agents and spray coated on the paper. The paper coated with modified  $\text{SiO}_2$  nanoparticles depicted superhydrophobic behavior because the  $\text{SiO}_2$  has the lowest amount of residual hydroxyl groups, which were quickly replaced by the hydrocarbon groups. Zhang et al. [18] prepared transparent superhydrophobic coating on paper through wet deposition of the emulsion of beeswax and carnauba wax followed by annealing.

Wood degradation arises in continuous contact with water. Recently, Jia and researchers [19] prepared a durable superhydrophobic coating on the wood surface using sol-gel process. The vinyl modified silica nanoparticles were applied on the wood by simple solution immersion method that demonstrated good mechanical stability against sandpaper abrasion test. Wang et al. [20] fabricated superhydrophobic wood surface through cultivating the ZnO nanorods on wood surface with subsequent stearic acid modification. Chang et al. [21] used the suspension of hydrophobic silica nanoparticles in polydimethylsiloxane (PDMS) solution to dip-coat the wood surface. The prepared  $\text{SiO}_2$ -PDMS composite coating on wood revealed excellent superhydrophobicity along with good mechanical durability against abrasion and ultrasonic washing. Oil spill in the environment is yet another serious issue faced around the world. Superhydrophobic-superoleophilic sponges can be utilized for efficient oil-water separation. Zhang et al. [22] fabricated superhydrophobic-superoleophilic polyurethane (PU) sponges by incorporating methyl modified silica nanoparticles in porous PU matrix with the aid of PDMS as adhesive. The

modified sponges could continuously and efficiently collect oil from the water surface with high speed. Furthermore, the sponges revealed excellent wetting stability against the abrasion and acid/alkali immersion. Several research groups have used hydrophobic silica nanoparticles to modify sponges for efficient oil-water separation application [23–25]. The superhydrophobic silica coatings were also utilized for conservation of marbles [26], plastics [27] and to effectively reduce the frictional drag [28]. In the present research article, the hydrophobic silica nanoparticles suspension was coated on the various substrates including body of motorcycle, building wall, mini boat, solar cell panel, window glass, cotton shirt, fabric shoes, paper (currency notes), plastic, metal, wood, marble and sponge. The coating was applied on the substrates using simple dip and spray techniques. All the coatings prepared on different substrates exhibited strong repellent behavior towards water with contact angle nearly  $160^\circ$  and sliding angle less than  $6^\circ$ . Though, the prepared superhydrophobic coatings lacks optical transparency and mechanical durability, the main objective of our research article is to explore the feasible industrial applications of self-cleaning superhydrophobic coatings.

## 2. Experimental section

### 2.1. Materials

Hydrophobic silica nanoparticles (Surface area  $210\text{ m}^2/\text{gm}$ , AEROSIL Company, RX 300-5, Japan) and hexane (Puriss for synthesis, Spectrochem, Mumbai, India) were bought. All the substrates like a motorcycle, glass, metal, mini boat, solar cell panel, wood, cotton fabric, window glass, paper, plastic, shoes, marble and melamine sponge were purchased from local market.

### 2.2. Preparation of superhydrophobic coatings

All the substrates were cleaned by water and detergent to remove dust and organic components. Hydrophobic silica nanoparticles were simply dispersed in hexane ( $5\text{ mg/ml}$ ), ultrasonicated for 30 min. and applied on various substrates by dip or spray coating technique depending on the shape of the substrate. While the dip coating process, the effect of deposition time and deposition layers on the wettability of the coatings was studied in detail. The 10 layers were applied with the deposition time of 1 min for each deposition layer. Whereas during spray coating, 12 spray layers from the distance of 15 cm were sufficient to achieve superhydrophobicity. After deposition, the coatings were dried overnight at room temperature ( $\sim 32^\circ\text{C}$ ).

### 2.3. Characterizations

The surface morphology of the coated substrate was studied by Field Emission Scanning Electron Microscopy (JEOL, JSM-7610 F, Japan). The water contact angles and sliding angles were measured for five times at different positions and their mean was taken as the final value using a contact angle meter (HO-IAD-CAM-01, Holmarch Opto-Mechatronics Pvt. Ltd. India). The self-cleaning property of the coated surface was checked by spreading carbon black powder (hydrophobic dust) or fine soil (hydrophilic dust) on the surface with subsequent cleaning by merely rolling water drops and muddy water test. The mechanical stability of the coating was qualitatively analyzed by normal fingertip touching, water jet impact, adhesive tape and sandpaper abrasion tests.

## 3. Results and discussion

### 3.1. Primary coating on small scale

Attempts have been made to coat superhydrophobic coating on several substrates for efficient self-cleaning applications. At first, the

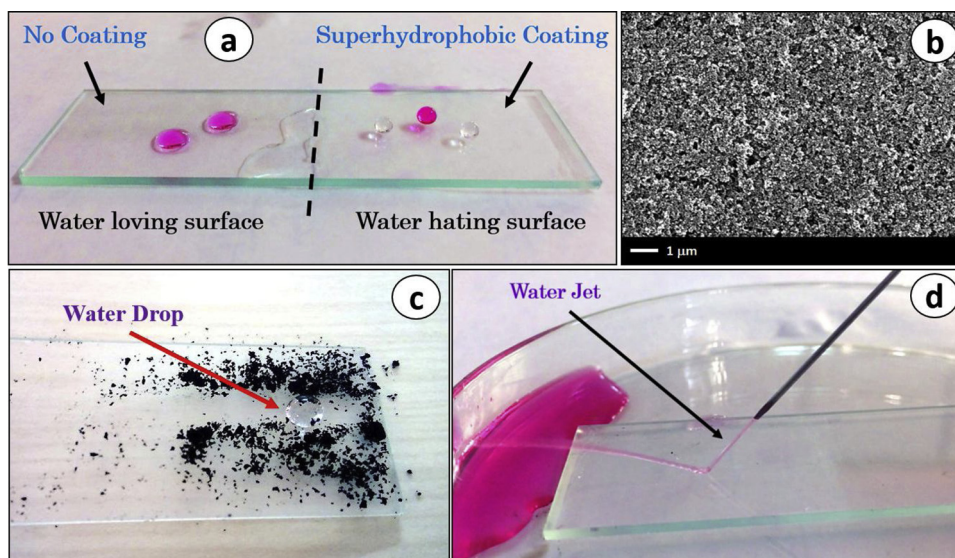


Fig. 1. (a) Optical photograph of water drops on non-coated and coated glass substrate, (b) SEM image, (c) self-cleaning ability, and (d) water jet bouncing on the superhydrophobic coating.

deposition was carried out at laboratory scale on a small piece of glass substrate using dip coating. As shown in Fig. 1a, the half part of the glass was dip-coated and half was left as it is. On the prepared coating, the water drop achieved the contact angle  $\sim 160^\circ$  and sliding angle less than  $6^\circ$ , confirming its superhydrophobic wetting state. Whereas, the uncoated glass surface showed strong wettability towards the water. The SEM image confirms the rough porous microstructure originated from the aggregated silica nanoparticles (Fig. 1b). Moreover, the SEM images captured at different magnifications confirmed uniform, rough and porous morphology of superhydrophobic coating as a result of aggregated silica nanoparticles (Fig. S1). The air trapped underneath the water drop inside the rough microstructure formed by the close-packed hydrophobic silica nanoparticles is responsible for the extreme non-wettability of the coatings [1]. On the self-cleaning superhydrophobic surface, the adhesion amongst dirt and surface is weaker than that between water droplet and surface. A water drop could effortlessly take away all the carbon black particles along its path confirming the self-cleaning ability of the prepared superhydrophobic coating (Fig. 1c). The mechanical durability of the coating structure was qualitatively evaluated by water jet bouncing test and adhesive tape test. The water jet was immediately repelled off the coating surface and the steady local hitting could not damage the surface as well as its wetting state (Fig. 1d).

A  $90^\circ$  adhesive tape test was carried out with tape adhesion strength of 4 N/m. The adhesive tape was gently applied and slowly pressed to remove any trapped air inside the coating surface [29]. After peeling off the adhesive tape (speed  $\sim 2$  mm/s), we observed that a small amount of coating material remained attached on the tape which was detached from the coating surface (Fig. 2). However, the coating still showed superhydrophobicity with slight devaluation in contact angle ( $\theta \sim 157^\circ$ ). In fact, the uppermost loosely bounded silica nanoparticles might have detached from the 10 times deposited coating. The superhydrophobicity has perished after 4 cycles of adhesive tape test. In the recent research paper, the spray coated hydrophobic silica nanoparticles from metals were completely removed after single adhesive tape test and the water contact angle value greatly devaluated from  $155^\circ$  to  $118^\circ$  [30]. In our case, the mechanical stability was further evaluated by performing robust sandpaper abrasion test [13]. The superhydrophobic glass plate loaded with 50 g weight was placed on the sandpaper (600 grit silicon carbide sandpaper) and linearly moved with a speed of 5 mm/s for 30 cm. We observed the complete removal of coating after sandpaper abrasion test, which confirmed poor adhesion

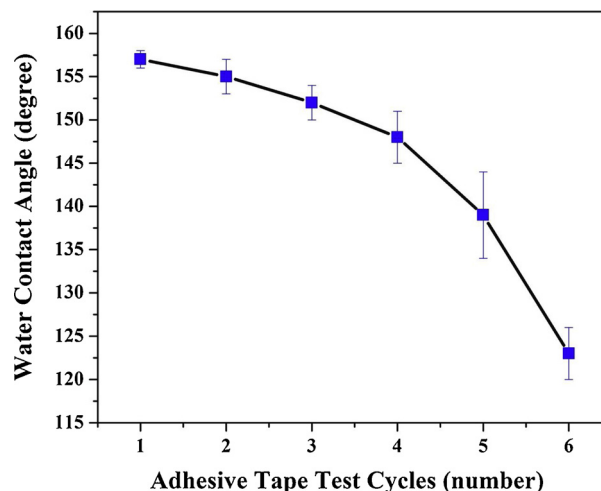


Fig. 2. Effect of adhesive tape test on the durability of the superhydrophobic coating.

of silica nanoparticles on glass. The unique deposition procedures adopted by Bayer et al. [13,14] can be very useful to attain mechanically durable polymer-nanoparticle superhydrophobic coatings on glass and metals.

### 3.2. Superhydrophobic coating on motorcycle (Honda, Activa-3G)

The vehicles can easily become dirty due to the accumulation of dirt, grime and organic pollutants from surrounding. Daily vehicle washing is a hectic process and requires costly liquid soaps, large amount of water, microfiber gloves, labor and time. This may reduce original shine and life of the vehicle. The transparent and durable superhydrophobic self-cleaning coatings are now gaining a lot of public attention. In the present study, the body of motorcycle was coated with suspension of hydrophobic silica nanoparticles by spray technique. The spray deposition was carried out at room temperature and the vehicle was dried overnight. To check the self-cleaning performance, a fine soil was dispersed on the body of vehicle and by merely pouring the water, the soil was eventually washed out (Fig. S2). Whereas, on non-coated motorcycle, the soil mixes well with water and leave muddy impression on the body. However, the superhydrophobic motorcycle

was thoroughly cleaned out with the simple water wash. A short video demonstrating spray coating and self – cleaning process on motorcycle is provided in the ESI. A gentle fingertip touching could not harm the coating, whereas the self – cleaning ability was intact for almost 5 months. Thorough removal, cleaning and re – application of superhydrophobic coating would again last for the same period.

### 3.3. Superhydrophobic coating on building walls

The building walls lose their shining appearance due to frequent accumulation of dust and carbonaceous particles from the surroundings. The polymer based house paint can only partially fulfill the self – cleaning ability, hence application of robust self-cleaning superhydrophobic coating may keep the building walls clean and shining. A natural rain shower or normal mechanical water spraying may efficiently self – clean the superhydrophobic building walls. We have developed the self-cleaning superhydrophobic building wall (1 m × 1 m) by simple spray coating the silica nanoparticle suspension (Fig. S3a). As depicted in Fig. S3 b–d, the superhydrophobic building wall exhibited strong repellency towards muddy water, whereas the ordinary wall eventually got dirty. A short video of self-cleaning behavior of superhydrophobic building wall is provided in ESI.

### 3.4. Superhydrophobic coating on cotton shoes

Sports shoes frequently gets dirty due to walking/playing in dusty and muddy areas. Regular washing can adequately degrade the lifespan of shoes. Superhydrophobic coating is one of the best choices to have self – cleaning sport shoes. The cotton shoes were spray coated and dried overnight at room temperature. A muddy water was prepared by adding fine soil in water. As shown in Fig. S4, the muddy water poured on the superhydrophobic cotton shoes was strongly repelled and no adverse impression of mud was observed on the surface of shoe. Liters of muddy water was continuously poured which could not deteriorate the superhydrophobicity of the shoes. Although the shoes were immersed in muddy water for several hours, still they kept repelling muddy water. A short video of muddy water repellent superhydrophobic cotton shoes is provided in the ESI.

### 3.5. Frictional drag reduction

The air trapped in rough micro/nanostructure of superhydrophobic surface effectively reduces the frictional drag enabling less energy consumption especially in submarines and watercrafts. Here, we spray coated the bottom of mini boat (weight ~ 30 g) by suspension of silica nanoparticles and compared the results of frictional drag between normal superhydrophilic and superhydrophobic-coated mini boat (Fig. S5). As shown in Fig. S5, the normal superhydrophilic and superhydrophobic-coated mini boat were placed on water-filled tub. The distance covered by both the mini boats in approximately 30 s were noted. A normal superhydrophilic mini boat travelled a distance of nearly 368 cm, whereas superhydrophobic-coated mini boat covered a distance of almost 506 cm confirming effective reduction in frictional drag. The reduction in frictional drag can eventually reduce the fuel consumption by submarines and watercrafts. A short video of frictional drag reduction by superhydrophobic-coated mini boat is provided in ESI.

### 3.6. Superhydrophobic coating on metal

The industrial use of metals and alloys are found in the construction of ships, aircrafts, electric wires, pipelines, buildings and many more. The deterioration of metals through oxidation and electrochemical process would be initiated in aqueous medium causing immense reduction in its life-span [31]. For instance, the frequent corrosion of ships and docks in seawater is one of the major issues associated with

the wear and tear of such automobiles. The superhydrophobic coating on metal can inhibit the corrosion by restricting the diffusion of water molecules. The metal currency coins and scale were dip coated and as shown in Fig. S6, the superhydrophobic metals strongly repelled the water drops. Immersion of these surfaces for almost 6 h in 3.5 wt. % of NaCl solution could not affect the wettability. However, slight reduction in water contact angle to  $\sim 151 \pm 4^\circ$  was observed for immersion in NaCl solution for 1 day. A short video of water drop bouncing on superhydrophobic metal currency coins is provided in the ESI.

### 3.7. Superhydrophobic coating on fabrics

Unlike other surfaces, the apparels are subjected to regular washing. Frequent washing of cloths requires copious amount of water, detergent, energy and machine or mechanical wash. Also laundry detergent waste adds secondary pollution to the environment. On the other hand, regular washing reduces usual shine and life of the cloths. Herein, we spray coated the cotton shirt by hydrophobic silica nanoparticles. As shown in Fig. S7, the colored water was poured on the superhydrophobic cotton shirt, perhaps water could not wet the shirt (A short video is provided in ESI). Each and every drop of water was easily collected back and no color impression was observed on the superhydrophobic shirt. In another self – cleaning study, the fine soil was spotted on the superhydrophobic shirt, immersed in bucket of water and taken out. The fine soil particles were eventually detached from the superhydrophobic shirt revealing its excellent self – cleaning ability. Thus, the superhydrophobic apparels can be cleaned merely by water dipping without the use of detergent, machine or mechanical wash.

### 3.8. Superhydrophobic coating on paper

The papers are mainly used for the preparation of notebooks, books, newspapers in addition to kitchen items like cups, bowls, mugs and plates. Cellulose papers are prone to wetting by liquids or by capturing moisture from surroundings. Currency notes are subject to wetting especially due to the sweat of human and so they can be teared apart easily under wet condition. By keeping this in mind, the wetting properties of currency notes (Rs. 100 /-) were modified by dip – coating. The optical appearance of the currency note was not altered by coating. As shown in Fig. S8, the different colored water was poured on the superhydrophobic currency notes with the help of syringe and beaker. The water drops as well as water stream were flawlessly bounced off the surface without wetting the surface. Moreover, the superhydrophobic currency note was kept under water for almost 2 h (Fig. S8d) that could not alter the superhydrophobic wetting state. Non – wetting currency notes are possible in coming future with the help of superhydrophobic coating. A short video of non – wettability of superhydrophobic currency note is provided in the ESI.

### 3.9. Self-cleaning coating on glass sheets

Glass is abundantly used material in our day to day life including windshields of automobiles, solar cell panels, window and door glasses, skyscrapers, tableware, glass furniture's, electronic appliances, and the list is unending. Self – cleaning glasses are immensely popular in public due to hectic cleaning procedures associated with normal glass, which requires labor, detergent, time and energy. Few accidents were also reported in the news while cleaning the skyscrapers. To get rid of it, the transparent and durable self – cleaning superhydrophobic coating is better option. The dirty superhydrophobic glass surfaces can be ultimately cleaned out by merely sprinkling of water. We have coated the rough and smooth window glass sheets by spray coating. Fig. S9 shows the optical photographs of spherical water drops on superhydrophobic window sheets. A short video of water drops rolling on superhydrophobic window sheets and plastic is provided in the ESI.

### 3.10. Superhydrophilic channels on superhydrophobic surface

The wings of Namib Desert beetle exhibit hydrophilic–superhydrophobic patterns through which it collects drinking water from fog-laden wind [32]. The fog droplets from atmosphere accumulate in hydrophilic channels and after growing in bigger shape experiences strong push from surrounding superhydrophobic surface resulting in rolling down directly in the mouth of beetle [33]. We have fabricated representative superhydrophilic – superhydrophobic patterns by masking the pre-cleaned glass plate with plastic mask (superhydrophilic channel width ~ 3 mm) and spraying the silica nanoparticle suspension on it. As shown in Fig. S10, the water was efficiently guided through the superhydrophilic channels, where poured water on superhydrophobic surface rolled off quickly and meets the superhydrophilic channel. Such superhydrophilic–superhydrophobic patterned surfaces find useful application in harvesting rainwater in draught-prone regions around the world. A short video of water collection through superhydrophilic – superhydrophobic patterned surface is provided in ESI.

### 3.11. Self-cleaning coating on solar cell panel

Solar cell converts solar energy into electrical energy under exposure to light. The solar cell efficiency can be greatly reduced due to the accumulation of dust on solar cell panels. It is the common issue faced around the world and various time consuming and costly mechanical/chemical methods are employed to clean the solar cell panels regularly. Optically transparent, durable and self-cleaning superhydrophobic coating on solar cell panels can be applied to improve the solar cell performance. We have spray coated the self-cleaning superhydrophobic coating on small solar cell panel as shown in Fig. 3. The dust particles were randomly spread on the superhydrophobic solar cell panel (Fig. 3a) and the rolling water drops could take away all the dust particles in a single attempt (Fig. 3b). Our study is only limited to check the self-cleaning performance of superhydrophobic solar cell panels. Dusty solar panels can be cleaned by merely pouring the water drops that eventually reduces the time, labor and chemical costs. A short video depicting the self-cleaning ability of superhydrophobic solar cell panel is provided in ESI.

### 3.12. Superhydrophobic coating on wood

Wood is generally used to construct houses, decorative furniture's, packaging boxes, boards, flooring and in many other things. However, the degradation of wood takes place upon contact with water. The wood can be protected against degradation by applying non-wettable superhydrophobic coating. A demonstrative wooden car was spray coated using hydrophobic silica nanoparticles. Fig. S11 shows the extreme water repellency of superhydrophobic wooden car (A short video is provided in ESI). Aforesaid superhydrophobic wood can last long compared to normal wood.

### 3.13. Self-cleaning superhydrophobic sponges

An enormous oil spill in the environment especially into the ocean is one of the serious issues tackled around the world. Oil removal and collection is a tedious process. Many expensive and time-consuming physical, chemical and biological methods were adopted for oil spill clean-up like filtration, in-situ burning, gravity separation, centrifuge and so forth [34]. Usually, sponges absorb any kind of liquid and modifying it by hydrophobic materials can significantly change its wettability to superhydrophobic and superoleophilic at the same time. Such modified sponges can strongly absorb any kind of oil by repelling water concurrently. The modified superhydrophobic sponges can be used with high separation selectivity, absorption efficiency and reusability. We have modified the melamine sponges by simply immersing it in the suspension of silica nanoparticles for 30 min and subsequent overnight room temperature drying. Fig. S12 depicts the superhydrophobic nature of the modified sponge. The water drops were strongly repelled, whereas oil drops were quickly absorbed inside the superhydrophobic sponge. The superhydrophobic sponges could freely float on the surface of the water.

The superhydrophobic sponge revealed oil uptake/absorption capacity of 27 times of its own weight. The oil-water separation ability of the prepared superhydrophobic sponge was performed (Fig. 4). All 10 ml diesel mixed in the 20 ml water was quickly absorbed by the superhydrophobic sponge with high selectivity. It was observed that nearly all diesel was recovered after squeezing the sponge in another beaker. Further, the recyclability of the superhydrophobic sponge was tested against absorption-squeezing cycles [4]. The sponge was rinsed in ethanol and dried after each absorption-squeezing cycle. The superhydrophobic sponge revealed oil-water separation capability for almost 25 absorption-squeezing cycles. Further, the oil absorption capacity and hence the oil-water separation capability of the sponge was decreased confirming the loss of silica nanoparticles during the oil-water separation cycles. A short video of strong water repellent characteristic of modified sponge and efficient oil-water separation process are provided in the ESI.

### 3.14. Self-cleaning coating on marble

Marble is the most popular among all items used in flooring, building wall, bathroom, mosaic, monuments and statues, kitchen-countertop, interior decoration accessories, stairs and pavements etc. Marbles eventually get dirty due to accumulation of dust particles and organic pollutants every day. Due to firm crystalline structure and slightly porous properties, marbles can easily absorb organic pollutant and other polluted liquids. Huge sums of money, energy and time is needed to clean the marbles by using clay, hydrogen peroxide and baking soda etc. In this type of cleaning, marbles also lose their natural shine. In the present work, we applied the superhydrophobic coating on marble, which was easily cleaned by just pouring the water on it. Fig. S13 depicts the superhydrophobic marble surface. A word 'SUPERHYDROPHOBIC' was masked before spray coating of silica nanoparticles.

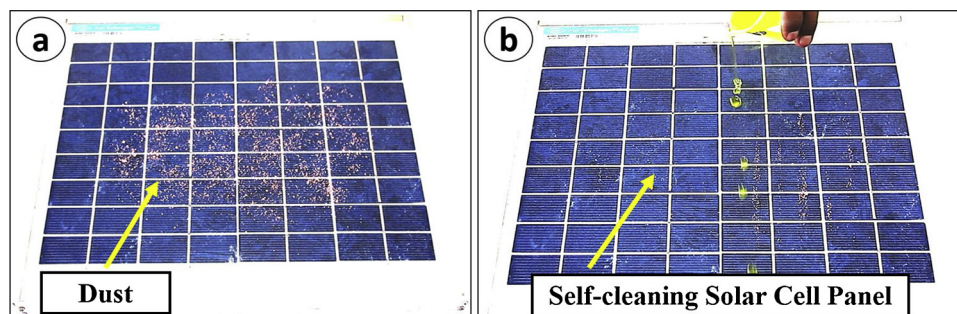
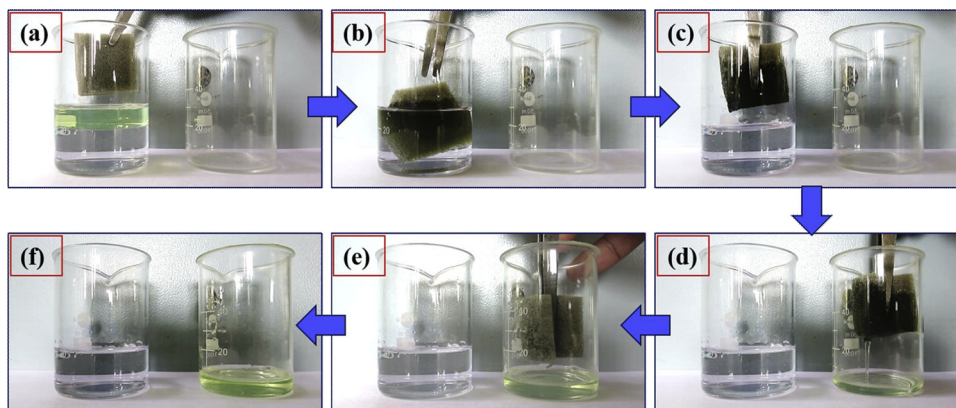


Fig. 3. Photographs of (a) dusty solar cell panel and (b) self-cleaning ability of superhydrophobic solar cell panels.



**Fig. 4.** Oil – water separation process; (a) superhydrophobic sponge dipped in the mixture of diesel and water, (b, c) sponge absorbs diesel, (d, e) diesel containing sponge squeezed in another beaker, (f) diesel was effectively separated from water.

**Table 1**

Comparison between the superhydrophobic coatings applied on various substrates.

Materials	Methods	Type of Substrates	Ref.
Styrene-b-(ethylene-co-butylene)-b-styrene elastomer (SEBS)	Casting	Glass, non-flat polypropylene, aluminum foil	[35]
Perfluorinated polydopamine	Oxidative polymerization	Gold, glass, zinc foil (Zn foil), polydimethylsiloxane (PDMS), vanadium foil (V foil), titanium dioxide (TiO <sub>2</sub> ), polyethylene terephthalate (PET)	[36]
ZnO	Modified Hydrothermolysis (MHT)	Glass, indium tin oxide (ITO) coated glass, Si substrate, Transparency sheet, Cotton wool	[37]
Fluoro-containing silica nanoparticles (Sol – gel process)	Dip, spin and spray coating	Glass, Polyester fabric, wool fabric, cotton fabric, silicon wafer, electrospun nanofibre mat, filter paper,	[38]
Poly(methyl methacrylate) (PMMA)-SiO <sub>2</sub> and Siloxane-SiO <sub>2</sub>	Spray Coating	Glass, silicon wafer, concrete, aluminum, silk, wood, marble	[39]
Polydimethylsiloxane (PDMS)-coated silica nanoparticles	Dip Coating	SS mesh, paper, cotton fabric, umbrella fabric, polytetra-fluoroethylene (PTFE) membranes	[40]
Polyaniline (PANI) / Copper oxide (CuO) nanocomposite	Polymerization and Chemical vapor deposition (CVD)	Glass slides, polystyrene (PS), Poly (ethylene terephthalate)	[41]
Polydimethylsiloxane (PDMS) / Silica nanoparticles	Aerosol-assisted chemical vapour deposition (AACVD)	Glass, cotton, copper mesh, copper block	[42]
Hydrophobic silica nanoparticles	Dip and Spray Coating	Motorcycle, building wall, mini boat, solar cell panel, window glass, cotton shirt, fabric shoes, paper (currency notes), metal, wood, sponges, plastic, marbles	Present Study

The superhydrophobic coating keeps the original shining of marble for a long time. A short video depicting behavior of water droplets on superhydrophobic marbles is provided in the ESI. Table 1 illustrates the comparison between the superhydrophobic coatings applied on various substrates in previous reports.

#### 4. Conclusion

The biomimicry of self-cleaning superhydrophobic lotus leaf can be attained to develop several useful commercial products. Self-cleaning superhydrophobic silica nanoparticle coating were applied on various substrates without much ruining their natural optical appearance. The dip and spray deposition layers were optimized to achieve the superhydrophobicity. A single coating approach fitted perfectly well for all shapes, sizes and types of the tested substrates. The tested surfaces include motorcycle (Honda, Activa-3 G), building wall, mini boat, solar cell panel, currency note (Rs. 100/-), sports shoes, currency coins, metal scale, cotton shirt, wooden car, big rough and smooth window glasses, plastic, sponges and marbles. The durability and optical transparency are the major issues associated with the superhydrophobic coating research and hence the commercial applications of self-cleaning superhydrophobic coatings are still awaiting. The efforts on improving the transparency and mechanical durability of the coating is the top priority and the subsequent experiments are still under process in our research laboratory. The superhydrophobic coatings attained by merely hydrophobic nanoparticles are lacking in durability and hence the

polymer-nanoparticle composites can be considered. We believe that, this research article will help early career researchers to understand the potential industrial applications of self-cleaning superhydrophobic coatings.

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#### Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.porgcoat.2018.12.008>.

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