Development of Self-cleaning Superhydrophobic Cotton Fabric through Silica/PDMS Composite Coating

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Development of Self-cleaning Superhydrophobic Cotton Fabric through Silica/PDMS Composite Coating

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Abstract

The lotus effect informs that self-cleaning superhydrophobic surfaces can be obtained by creating rough surface structures and modifying them with chemicals that have low surface energy. Herein, the composite of hydrophobic silica nanoparticles (SNPs) and polydimethylsiloxane (PDMS) was deposited on cotton fabric by multiple dip cycles. At optimal condition, the agglomerated SNPs in PDMS produces a hierarchical rough surface, as a result the coated cotton fabric has revealed a water contact angle (WCA) of 158.41 ± 1.58° and 4° of sliding angle. Due to negligible water adhesion to a superhydrophobic surface, coated cotton fabric reveals excellent self-cleaning behavior, which was tested by dust particles, muddy water and tea droplets. Furthermore, coated cotton fabric sustains superhydrophobicity over the mechanical robustness tests including adhesive

tape peeling test, sandpaper abrasion test, and ultrasonication. Therefore, such an approach may be applicable in textile industries for self-cleaning purpose.

Keywords: Superhydrophobic coating; hydrophobic silica nanoparticles; superhydrophobic cotton fabric; self-cleaning coating

1. Introduction

A lotus leaf is very well known as example of purity, the dust particles easily removed by rolling water droplets from the leaf surface. The presence of micro-scale papillae and nano-scale wax materials, the lotus leaf surface exhibits self-cleaning superhydrophobic characteristic [1]. A superhydrophobic surfaces have exhibits higher WCA as well as lower sliding angle, due to this reason water droplets are freely movable on their surface. The dust particles have strong adhesion towards water droplets than superhydrophobic surface. Hence, the rolling water droplets can easily collect dust particles on the way and cleaned surface itself, this performance is recognized as self-cleaning or lotus effect [2]. Till a date, the superhydrophobic surfaces have been used for self-cleaning [3], oil-water separation [4], anti-freeze [5], and anti-corrosion [6] applications. Many efforts have been made to fabricate superhydrophobic coatings using various fabrication techniques such as phase separation [7], spray coating [8], drop-casting [9], dip coating [10, 11] and so on [12, 13].

Cotton fabrics have softness, breathability, moisture absorption, and flexibility characteristics [14]. Cotton fabric is one of the essential material, utilized extensively in households and industries for a variety of reasons in our daily lives. The apparels that made of cotton fabrics are contaminated by various types of contaminants in indoor and outdoor movement. The contaminated apparels required regular washing. The frequent mechanical washing by detergents degrades the shine and lifetime of the cotton fabrics. Therefore, cotton fabrics must have the ability to clean themselves in order to withstand dust particles and other surface contaminants [15]. The self-cleaning superhydrophobic coating on cotton fabric can resolve these issues [16]. The cotton fabric has inherent roughness; hence the superhydrophobicity on cotton can be easily achieved by applying a layer of low surface energy materials [17]. Recently, Latthe et al. [18] have deposited a suspension of hydrophobic silica nanoparticles on cotton fabric shirt and fabric shoes via spray and dip coating method, respectively and obtained superhydrophobic surface. They also deposited hydrophobic silica nanoparticles suspension on body of a motorcycle, a little boat, a solar cell

cover glass, building wall, window glass, currency notes, metal, wood, sponges, plastic, and marble substrates. Such superhydrophobic coated substrates had showed outstanding self-cleaning properties. Eduok et al. [19] has prepared superhydrophobic cotton fabric by depositing a nanocomposite of zirconia/ polydimethylsiloxane through dip coating technique. The prepared superhydrophobic cotton fabric was showed WCA of 155.5 ± 0.5° and contact angle hysteresis of 2.5 ± 0.3°. The coated cotton fabric showed self-cleaning properties against chalk particles were taken as dust particles. Phuong et al. [20] have prepared superhydrophobic cotton fabric using silica nanoparticles and tetraethylorthosilicate (TEOS) by dip coating method. The silica nanoparticles agglomerated on the fibers endows the rough structure. As a result, the coated cotton fabric had showed WCA of 173°. Xiong et al. [21] have fabricated fluorine-free multifunctional superhydrophobic cotton fabric by using dip-coating method. Micro-sized SiO_2 and nano-sized TiO_2 particles were bound together by silane coupling agent. A hierarchical structure was formed due to SiO_2 and TiO_2 particles strongly bonded to the cotton fabric with silane. Such superhydrophobic cotton fabric exhibited a WCA of 161.7°. Pal et al. [22] have fabricated superhydrophobic cotton fabric by simple dip coating method using TiO_2 and 3-(trimethoxysilyl) propyl methacrylate. The fabricated superhydrophobic cotton fabric exhibited excellent self-cleaning behavior for rhodamine-B dye as a pollutant. The superhydrophobic cotton fabric was showed mechanical stability for 50 adhesive tape peeling cycles with the WCA reduced from 154.3° to 146.8°. In 20 cycles of washing durability test, the WCA decreased from 154° to 143.2°. Many researchers have used numerous chemicals, and complicated fabrication processes to develop self-cleaning superhydrophobic cotton fabric [23-27]. Weakly adhered hydrophobic material on cotton surface has revealed poor mechanical robustness. Consequently, production cost may increase and limited to their industrial scale applications. To avoid these drawbacks, a facile and cost-effective approach is essential for production of robust superhydrophobic coating on cotton fabric for large scale application.

In this study, a simple dip-coating approach was adopted to produce self-cleaning superhydrophobic coating on cotton fabric by depositing consecutive dipping layers of PDMS and hydrophobic SNPs. The PDMS act as an adhesive layer between the substrate and SNPs and inter-binding of nanoparticles so that the durability of the coating can be enhanced on the cotton fabric. The coated cotton fabric exhibits good mechanical robustness against adhesive tape testing,

sandpaper abrasion testing, heat treatment, and ultrasonication test. Also, the coated samples showed excellent self-cleaning ability.

2. Experimental

2.1. Materials

Polydimethylsiloxane (PDMS; viscosity 5 cSt) was purchased from Sigma-Aldrich, St. Louis, MO, USA. Hydrophobic silica nanoparticles (SNPs; ≥ 99.8% SiO_2; particle size 50 nm; AEROSIL R 9200) was purchased from Nippon AEROSIL Co. Ltd., Japan. Chloroform ($CHCl_3$; 99%; AR grade) purchased from spectrochem private limited, Mumbai, India. Pure cotton fabrics were procured from DKTE Society, Textile industry, Ichalkaranji, Maharashtra, India.

2.2. Preparation of the superhydrophobic coating on the cotton fabric

Initially, the cotton fabric was cut into small pieces of size 8 x 3 cm^2. The cotton fabric pieces were cleaned in distilled water and ethanol to remove surface contaminants using ultrasonication bath and it dried in oven at 80°C for 30 min. A 0.5 mL of PDMS was dissolved in 20 mL of chloroform by using magnetic stirrer for 30 min. Simultaneously, 100 mg of hydrophobic SNPs were dispersed in 20 mL of chloroform by continuous stirring for 30 min. The pre-cleaned cotton fabric was dipped in the PDMS solution for 10s and dried at room temperature for 5s. Thereafter, the cotton fabric was dipped in the SNPs dispersion for 10s and kept at room temperature for drying. In this, the dip and withdrawing speed (100 mm/min) was controlled by dip coating machine. The first layer deposition of PDMS and SNPs is referred as one dipping cycle of coating composite. Likewise, 5 dipping cycles were applied on cotton fabric and dried at 100°C for 30 min in hot air oven. Furthermore, for achieving optimal coating 10 and 15 dipping cycles were applied on fresh cotton fabric. The cotton fabric coated by 5, 10 and 15 dipping cycles are labeled as SC-1, SC-2, and SC-3 respectively. Figure 1 depicts the schematic of preparation of the superhydrophobic cotton fabric.

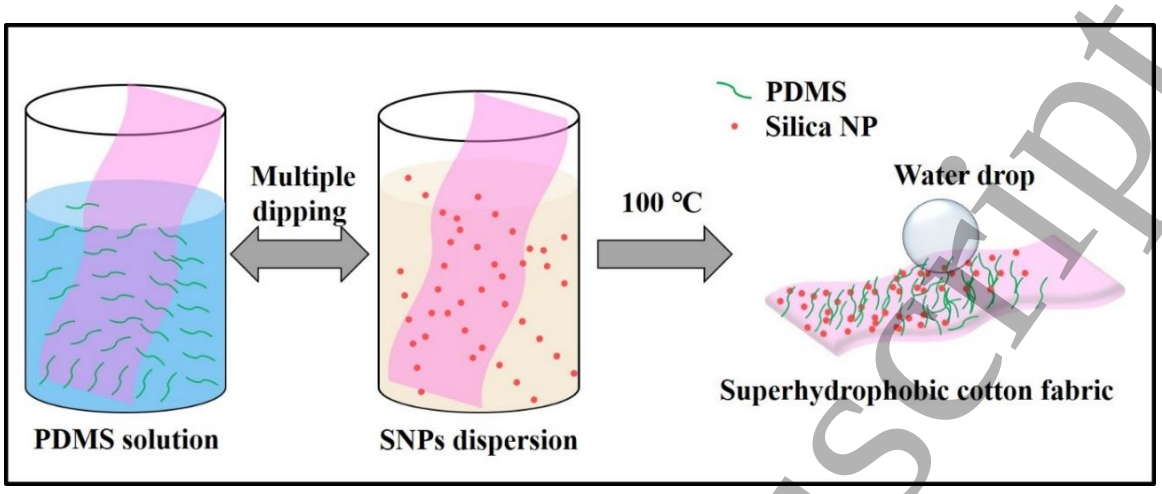

Figure 1: Schematic representation of fabrication of a superhydrophobic cotton fabric.

2.3. Characterizations

Surface morphology was studied by scanning electron microscopy (SEM, JEOL, JSM-7610F, Tokyo, Japan). Elemental analysis was performed using an energy dispersive spectroscopy (EDS, Oxford Instruments X-Max, England) instrument. The chemical bonding was represented using Fourier transform infrared spectroscopy (FTIR, Magna-IR 560, Nicolet). The water contact angle and sliding angle of coatings were measured using a contact angle meter (HO-IAD-CAM-01, Holmarc Opto-Mechatronics Pvt. Ltd., Kochi, India). Mechanical durability of coatings was evaluated by adhesive tape test, sandpaper abrasion test, heating, and ultrasonication. Scotch magic tape having 4 N/m of adhesion strength was purchased from 3M India limited to carry out the adhesive tape test. Sandpaper (Grit No. C-120) was purchased from John Okey & Mohan Ltd., India for the sandpaper abrasion test. Thermal stability of coatings was determined by keeping the coated cotton fabric in oven at 200°C for 2 h. For ultrasonication test, the cotton fabric was kept in ultrasonic bath for 20 min. A self-cleaning ability of coated cotton fabrics were checked by spreading dust particles as well as pouring muddy water and tea on it.

3. Results and discussion

3.1. Surface morphology, chemical composition and wettability

The surface and chemical structure analysis are an essential characterization for evaluating the superhydrophobic characteristic of prepared coatings. During the consecutive deposition of PDMS and SNPs layers on cotton fabric, SNPs embedded in polymer layer. After multiple deposition, SNPs get aggregated on cotton fabric surface. As shown in figure 2 (a-f) illustrates that the SEM

images of coated cotton fabrics. Figure 2 (a and d) depicts lower and higher magnified SEM micrographs for the 5 deposition cycles (SC-1 sample), coating composite covers cotton fabric surface and it makes hydrophilic cotton fabric becomes hydrophobic (WCA of 137.3 ± 1.37°). The 10 consecutive layered deposition of PDMS and SNPs (SC-2), the agglomeration of SNPs occurs and produces microscale cavities, consequently surface structure more roughened than SC-1 sample as shown in figure 2 (b and e). The value of WCA gradually increased. For SC-3 sample (15 deposition layer), the agglomerated SNPs and regular sized large number of cavities produces rough hierarchical surface structure as shown in figure 2 (c and f). The cross-linking between the PDMS and hydrophobic SNPs formed hierarchical rough surface structure, is essential for achieving superhydrophobicity [28]. The dual scale hierarchical rough surface structure enable to trap air layer in their protrusion, which minimizes the actual contact of water drop to surface [29]. Therefore, the SC-3 sample reveals higher WCA of 158.41 ± 1.58° and SA of 4°. After 15 deposition cycles WCA becomes almost steady at around 158.5 ± 1.58° with number of deposition cycles. While depositing 20 and 25 layers of PDMS and SNPs on cotton fabric, the surface revealed WCA of 158.3 ± 1.58° and 159.32 ± 1.59°. However, due to deposition of layers above 15, the surface depicted powdery appearance of coating material which was not found stable and ripped out by gentle fingertip touching. Hence, 15 deposition cycles of PDMS and SNPs is an optimum for achieving superhydrophobicity on cotton fabric. The optical image of colored water droplets on SC-3 sample is shown in figure 2h and their contact angle image shown in inset of figure 2h. The EDS spectra of coating composite reveals three major peaks that related to carbon (C), oxygen (O), and silicon (Si) elements. The larger proportion of C is related to the polymeric structure of PDMS and the elements Si and O present in composite coating owing to PDMS and SNPs. This proportion of C, O and Si confirms that the coating composite of PDMS and SNPs present on cotton fabric surface. The mass percentage of Si, C and O elements are given in Table 1.

Table 1: The mass percentage of elements present in coatings.

Sample	Si –K (Mass %)	C-K (Mass %)	O-K (Mass %)
SC-1	7.46 ± 0.93	44.79 ± 1.63	47.74 ± 2.91
SC-2	6.81 ± 0.92	47.68 ± 1.70	45.51 ± 3.02
SC-3	5.78 ± 0.90	49.54 ± 1.74	44.68 ± 3.10

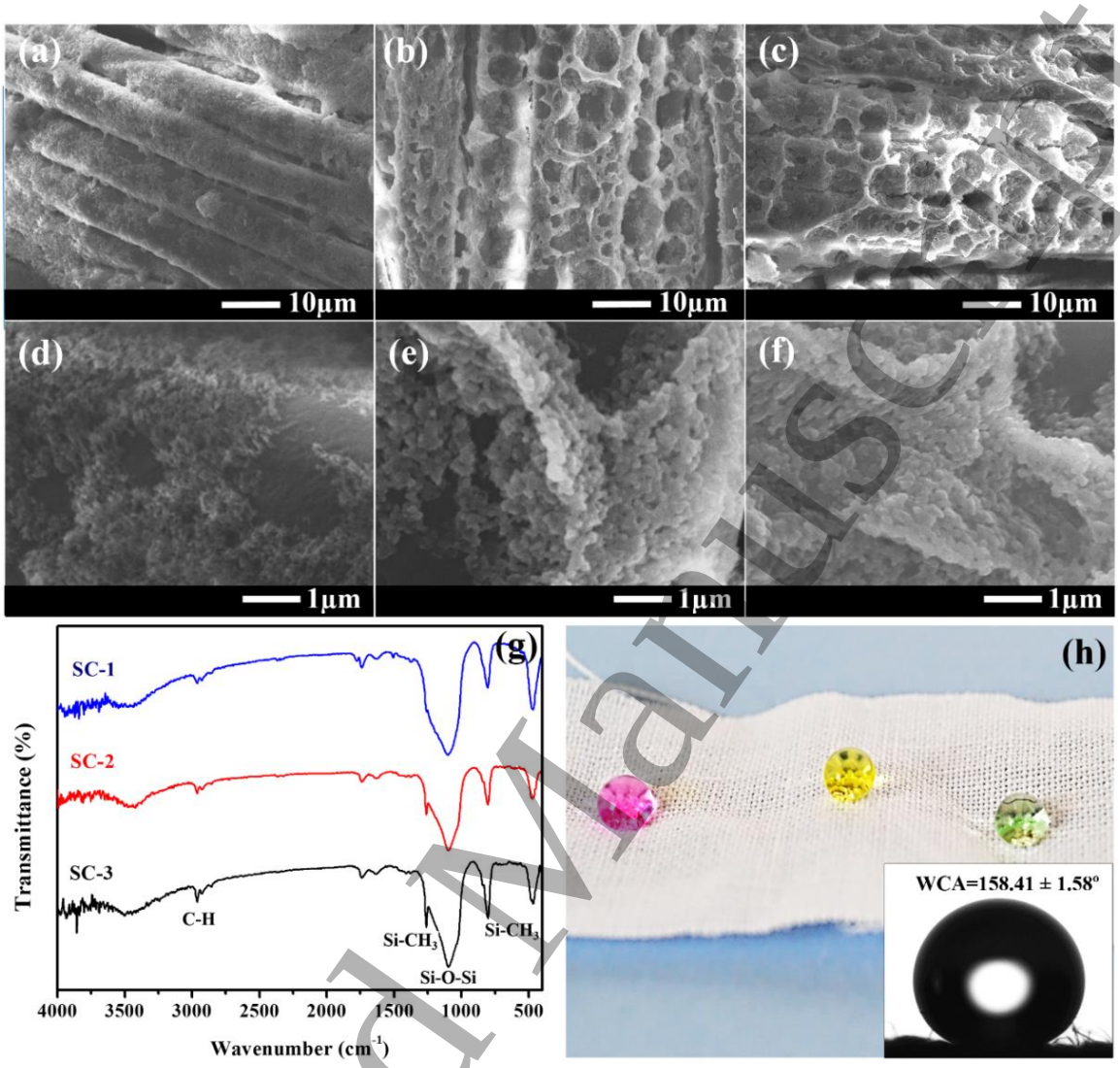

Figure 2: (a-c) lower and (d-f) higher magnified SEM images of SC-1, SC-2 and SC-3 samples, respectively. (g) FTIR spectra of SC-1, SC-2 and SC-3 samples. (h) Photograph of the colored water droplets on the SC-3 sample and in inset the optical water drop image on SC-3 sample.

The chemical bonding of the coatings (SC-1 to SC-3) is illustrated in figure 2g using the FT-IR spectroscopy. Due to the presence of PDMS and hydrophobic SNPs the strong peaks are seen in the region of 1100-470 cm^{-1}. The peaks at 800 cm^{-1} and 476 cm^{-1} are represented the asymmetric and symmetric deformation of the PDMS. The peaks at 1260 cm^{-1} and in the range 800-470 cm^{-1} are represents $Si\text{-}CH_3$ group [30, 31]. The peak at 1092 cm^{-1} is assigned to asymmetric stretching vibration of Si-O-Si bond [32]. The peaks presence at 2963 cm^{-1} and 2904 cm^{-1} due to C-H

asymmetric and symmetric vibrations, respectively [32]. Hence, the FTIR analysis concluded that the PDMS and hydrophobic SNPs exist in the coating.

3.2. Mechanical durability

Adhesive tape test was performed to evaluate adhesiveness of the coating materials on the cotton fabric [33]. The SC-3 sample revealed excellent superhydrophobic properties, therefore the mechanical durability was checked for this sample. Adhesive tape was adhered to the SC-3 sample and gently pressed by a fingertip to make adequate contact between coated fabric and adhesive tape [34, 35]. The tape was peeled off to complete the adhesive tape test. In a similar manner, the adhesive tape test was performed 30 times. After completion of every 5 cycles of adhesive tape test the WCAs were recorded. As shown in figure 3a, the WCA gradually decreases with number of adhesive tape test. The SC-3 sample retain their superhydrophobic property up to 15 cycles of adhesive tape test with exhibiting WCA of 150.46 ± 1.5° and 7° of SA. Hence it ensures that the coating material strongly adhered to cotton fabric and surface structure slightly disrupted as shown in figure S1 (a). The presence PDMS as a binder in coating composite has created strong bonding towards the cotton fabric surface [36]. The WCA reduced to 138 ± 1.38° after completion of 30 tape peeling cycles, indicates that coating material moderately detached from cotton fabric. The screen snaps of adhesive tape test are shown in inset of figure 3a.

Due to fragile, brittle surface structure and poor mechanical resistance, the superhydrophobic surfaces are disregard from practical applications. A sandpaper abrasion test is a common method to determine the wear resistance of the coatings [33]. The SC-3 sample was put on the sandpaper and a weight of 40 g imposed on it. The sample was dragged for 5cm over the sandpaper with 5 $mm.s^{-1}$ of speed; it is referred as one-cycle of sandpaper abrasion test. This process was repeated for 50 times. As shown in figure 3b, the WCA continuously drops as sandpaper abrasion tests increased. The SC-3 sample sustained its superhydrophobicity up to 30 cycles of sandpaper abrasion test with WCA of 153.5 ± 1.53° and 6° of SA. Figure S1 (b) shows that the microscale cavities were wrecked and formed new rougher structure with presence of microsized aggregated SNPs on SC-3 sample, hence it proved that the coating became tougher and resistant to mechanical damage [37]. Further, increasing sandpaper abrasion test to 50 cycles, the WCA decreased to 134 ± 1.34°. This is due to detaching the weakly bounded SNPs from the PDMS film, which impacts overall surface roughness. The experimental arrangement of sandpaper abrasion test is shown in inset of figure 3b.

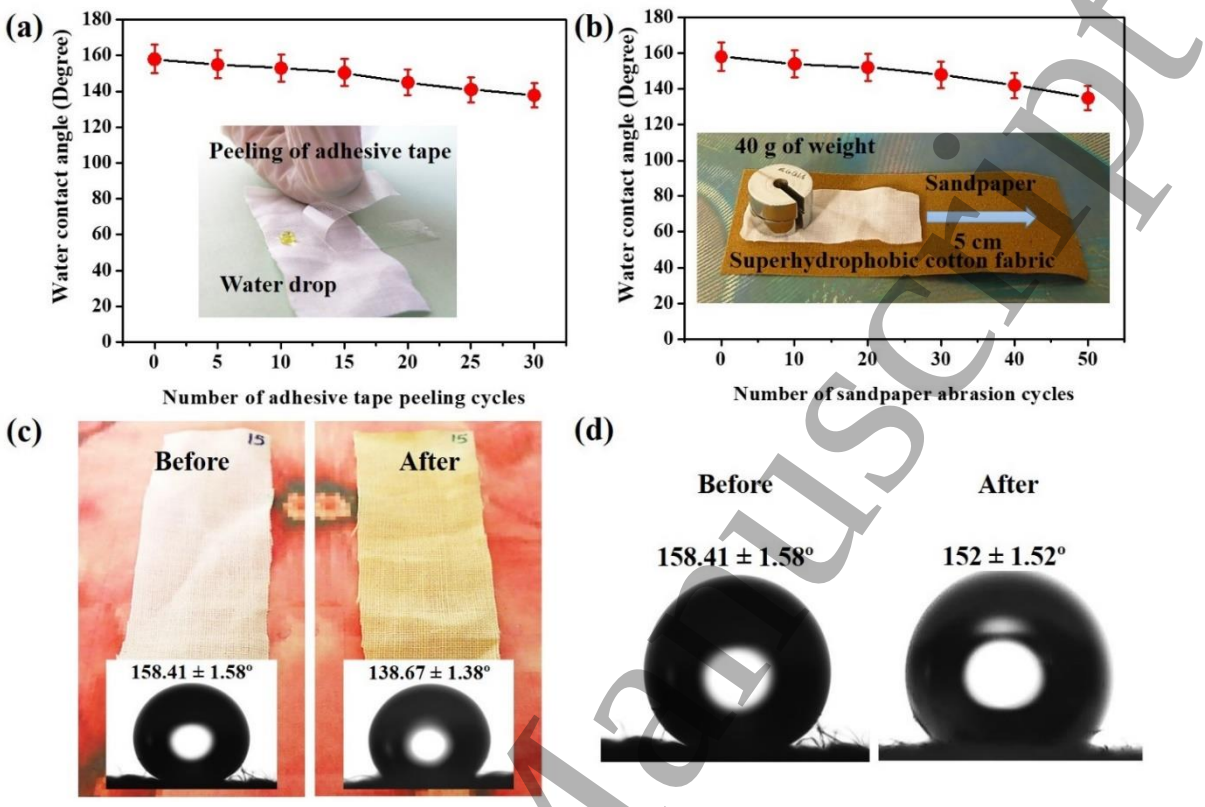

Figure 3: (a) The change in WCA with respect to number of adhesive tape peeling cycles for SC-3 sample and in inset the screen snaps of adhesive tape peeling test. (b) The variation of WCA with respect to number of sandpaper abrasion cycles for SC-3 sample and in inset the optical image of experimental setup of sandpaper abrasion test. (c) The optical photographs and optical water profile image before and after heating the SC-3 sample (d) The optical water profile image before and after the ultrasonication treatment.

The SC-3 sample kept in oven at 200°C for 2 h to evaluate thermal stability of coating. As shown in figure 3c, the WCA of SC-3 sample significantly reduced to 138.67 ± 1.38°. This can be due to the fact that surface structure might have restructured by increasing temperature and subsequent decomposition of hydrophobic materials [38]. In another method of evaluating mechanical stability of coatings, the SC-3 sample was kept in water filled ultrasonication bath for 20 min. Ultrasonication is regarded as a more abrasive process than laundering [39]. Immediately the WCA was measured after treatment, found that WCA reduced to 152 ± 1.52° and SA decreases with small value (figure 3d). Ultrasonication test confirms that the prepared coating on cotton fabric is highly durable and sustain their superhydrophobicity up to 20 min of ultrasonic treatment [40].

The effective results of this work against other literatures are comparatively summarized in Table 2.

Table 2. Comparative study of mechanical robustness between current work and existing literatures.

Materials	Coating Method	Adhesive tape peeling test	Sandpaper abrasion test	Thermal stability test	Ultra-sonication treatment	Ref.
zirconia nanoparticles and polydimethylsiloxane	Dip	-	-	-	-	[16]
Zirconium (IV) n-propoxide (ZP) and 1 H, 1 H, 2 H, 2 H-perfluorodecyltriethoxysilane	Dip	-	20	-	-	[41]
Zirconium (IV) n-propoxide (ZP), hexadecyltrimethoxysilane, silver nitrate, cetyltrimethylammoniumbromide	Dip	30	30	-	-	[34]
Triethoxyvinylsilane, polydimethylsiloxane, silver nitrate, cetyltrimethylammoniumbromide	Dip	20	10	-	-	[42]
Tetraethylorthosilicate, Dopamine hydrochloride, Tris(hydroxy methyl)aminomethane, Dodecyltrimethoxysilane	Dip	-	-	-	40 min	[43]
Hydrophobic silica nanopartilces and polydimethylsiloxane	Dip	15 cycles	30 cycles	200℃	20 min	Present work

3.3. Self-cleaning ability

Every solid surfaces in an exterior application are contaminated by different types of dust particles. Frequent cleaning requires the use of detergent, hazardous chemicals, time, and labor. Multiple mechanical cleanings reduce the initial shine of the surface. As prepared samples have extreme

non-wetting property due to high WCA 158.41 ± 1.58° and SA of 4° that allows them to clean themselves. The self-cleaning ability of superhydrophobic cotton fabric was observed by spreading dust particles as well as pouring muddy water and tea. As shown in figure 4 (a-b), the SC-3 sample was kept in petri dish and the fine soil dust randomly spread on sample surface. When water poured on the dust contaminated sample, water droplets become ball like shape and roll down in downward direction. The rolling water droplets gather dust from the path and gently cleaned the surface. During the rainy season, many objects endured mud problems. Therefore, muddy water prepared by adding of 10 g of fine soil particles in 20 mL of water and poured on SC-3 sample (figure 4c-d). The muddy water drops roll down from superhydrophobic cotton fabric similar ordinary water droplets. Furthermore, tea was dumped onto a superhydrophobic cotton fabric (SC-3 sample), and tea droplets rolled down without adhering to the surface (figure 4e-f).

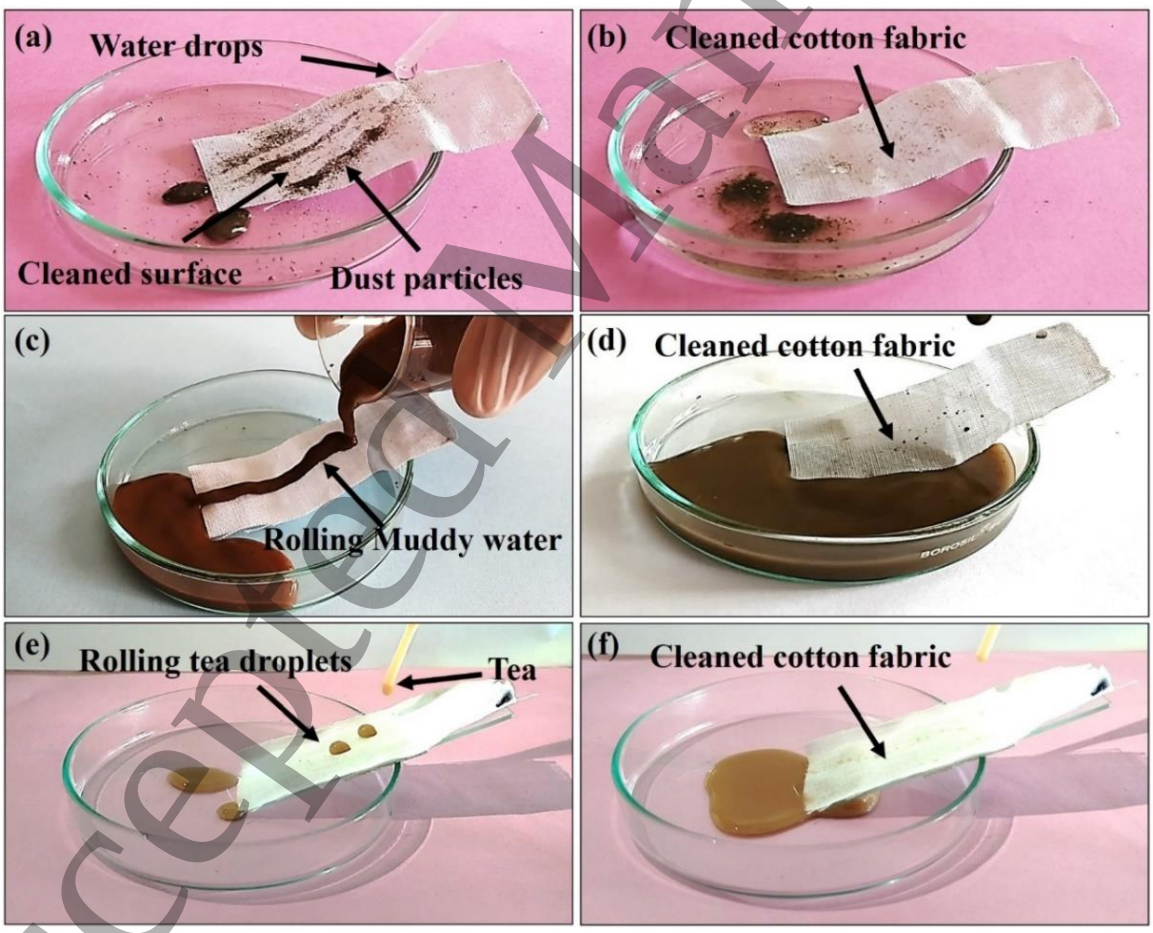

Figure 4: The self-cleaning effect against (a-b) dust particles, (c-d) muddy water and (e- f) tea.

4. Conclusions

In summary, a simple low-cost dip-coating approach was employed to prepare superhydrophobic cotton fabric using polydimethylsiloxane (PDMS) and hydrophobic silica nanoparticles (SNPs). During the preparation, SNPs aggregated in low surface energy PDMS forms regular shaped micro cavities, produces high scale roughness. As a result, coated cotton fabric (SC-3 sample) exhibited WCA of 158.41 ± 1.58° and SA of 4°. The chemical composition analysis indicates that the SNPs-PDMS coating composite covers the cotton fabric. The SC-3 sample was revealed good mechanical stability against 15 cycles of adhesive tape test, 30 cycles of sandpaper abrasion test and 20 min of ultrasonication treatment. The prepared SC-3 sample demonstrated excellent self-cleaning ability in a variety of situations. Therefore, such an approach may be relevant for self-cleaning applications in the textile industries.

Data availability statement

The data that support the findings of this study are available upon request from the authors.

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Declaration of interests

The author(s) declare no conflicts of interest with respect to the research, authorship, and publication of this article.

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