



Fabrication of Superhydrophobic Coatings on Glass based on Poly(dimethylsiloxane) and Fumed Silica

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Abstract: This study reports the preparation of superhydrophobic covalently attached poly(dimethylsiloxane)(PDMS)/fumed silica composite coatings on glass via spin-coating and subsequent heating. The effect of PDMS molecular weight and fumed silica type (hydrophilic and three different hydrophobic) on the wettability and morphology were studied in detail, and the wettability and morphology of the coatings were characterized by contact angle measurements and scanning electron microscopy (SEM). Superhydrophobic coatings which have advancing contact angles of 172-174° and contact angle hysteresis (CAH) as low as 7-8° were obtained using 4% (w/v) PDMS modified hydrophobic fumed silica and all kinds of PDMS. The surfaces which were prepared by 4% (w/v) hydrophilic and dimethylsiloxyl-modified fumed silica had high advancing contact angles of 163-168°, but the CAH on these surfaces were higher than the ones prepared using PDMS modified fumed silica. It is due to the incompatibility of the hydrophilic and dimethylsiloxyl-modified fumed silica with the PDMS matrix also confirmed with the SEM results.

Keywords: poly(dimethylsiloxane), fumed silica, wetting, superhydrophobic coatings, contact angle, spin-coating

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INTRODUCTION

There is a great interest in the fabrication of artificial superhydrophobic surfaces over the last 20 years after Barthlott et al. reported the self-cleaning property of a lotus leaf in 1997, which is known as a "lotus effect" (1-5). Superhydrophobic surfaces, which have water contact angles (CA) larger than 150°, sliding angles lower than 5° or contact angle hysteresis (CAH) lower than 10°, can be used in various applications such as oil-water separation, self-cleaning, anti-icing, anti-fouling, and drop transport (5-7). There are two crucial factors when designing hydrophobic surfaces; (1) using a low energy material as a coating and (2) creating a rough surface, which will increase the hydrophobic surface character according to the Cassie Baxter wetting situation (8). Therefore, superhydrophobic surfaces can be prepared either by modifying rough

surfaces using low energy materials or by making a roughness using low energy materials (2-7,9).

To date, researchers used different techniques to create various superhydrophobic surfaces such as sol-gel, lithography, templating method, colloidal assembling and aggregation, layer-by-layer assembly, plasma method, chemical vapor deposition, phase separation, electrospinning and electrospraying, dip-coating and so on (4,10-11). These techniques are quite complicated and expensive due to multiple steps, specific equipment usage, and costly reagents. Besides, they can be applied to specific substrates. Overcoming such difficulties, different kinds of polymers, together with various nanoparticles, were used to fabricate superhydrophobic surfaces on different substrates where the nanoparticles were used to create surface texturing on the surfaces (12-18). Silica-based

nanofillers are the most used ones in the literature to create superhydrophobic and self-cleaning surfaces (14, 19-26). Manoudis et al. prepared superhydrophobic composite films containing hydrophilic silica nanoparticles and poly(methyl methacrylate) (PMMA) and commercial poly(alkyl siloxane) (Rhodorsil 224) on various substrates (glass, silicon, wood, concrete, aluminum, silk, marble) via spray coating (14). Superhydrophobic films were fabricated on glass via a drop-coating method using Sylgard 184 silicone (containing curing agent and poly(dimethylsiloxane) (PDMS) prepolymer) and silica particles (19). Chang et al. successfully prepared the superhydrophobic wood surfaces via a dip-coating method using PDMS Sylgard 184, curing agent, and hydrophobic silica particles (20). They also investigated the morphology, wetting behavior, and roughness of the coatings as a function of silica concentration (20). Superhydrophobic composite films containing hydrophobic fumed silica particles and different polymers (thermoplastic resin, PS, cross-linked epoxy resin, PDMS-urea copolymer, polyether-based polyurethane urea, PMMA, polycarbonate) were prepared on glass via spin-coating process or doctor blade coating method by Yilgor and coworkers (21-23). Cai et al. fabricated superhydrophobic fabrics via successive dip-coating of PDMS and silica suspensions and successive annealing steps (24). Also, bioinspired superhydrophobic cross-linked polyurethane-acrylate coatings containing hydrophobic fumed silica on glass were prepared via bar coating following UV curing by Fourmentin and coworkers (25). The superhydrophobic coating on cellulosic filter paper for oil-water separation was achieved via solution casting technique using PMMA copolymer and silica nanoparticles by Siriam et al. (26).

As noted above, several papers in the literature report the preparation of superhydrophobic coatings on various substrates such as glass, wood, aluminum, silicon wafer using PDMS, low surface energy material, and nanoparticles (14,19,20,24,27). Either these coatings are not covalently attached to the substrates (14), or they are prepared using curing agents (19,20,24, 27). In 2011, McCarthy and coworkers showed, for the first time, that linear polysiloxanes containing only Si-O-Si in their main chain react with inorganic surfaces containing hydroxyl groups upon heating to desired temperature (especially 100 °C for 24 h) (28). Furthermore, they reported that they obtained low hysteresis surfaces and concluded that they could control the thickness of the PDMS layers on silica by changing the molecular weight of the PDMS.

In the present study, we have prepared superhydrophobic PDMS/silica coatings which are covalently attached to glass via a simple spin-coating process and subsequent heating. The wettability and surface morphology of PDMS/silica

composites on glass were characterized by contact angle measurements (equilibrium, advancing and receding contact angle) and scanning electron microscopy (SEM), respectively. The effect of PDMS molecular weight, silica concentration, and silica type on coatings' morphology and contact angles were also systematically studied, and the results were compared with the findings reported in the literature.

EXPERIMENTAL DETAILS

Materials

Polydimethylsiloxane (PDMS) with three different weight average molecular weight (M_w) of 2000 (2k), 63000 (63k) and 117000 (117k) were obtained from Alfa Aesar (Massachusetts, USA). Amorphous hydrophobic fumed silicon (IV) oxide (~50 nm; PDMS modified, referred to as FS) was obtained from Alfa Aesar. Amorphous hydrophobic pyrogenic silica with different particle sizes and different surface modification (~40 nm; dimethylsiloxyl modified; referred as H15 and ~30 nm; PDMS modified; referred as H18) and amorphous hydrophilic pyrogenic silica (~25 nm; referred as N20) were obtained from Wacker (Burghausen, Germany). Toluene was obtained from Sigma-Aldrich. Ethanol, sulfuric acid (H_2SO_4 , 95-97%), hydrogen peroxide (H_2O_2 , 30%), and ultra-pure water were obtained from Merck (Darmstadt, Germany). All reagents were used as received without further purification. Glass microscope slides were obtained from ISOLAB GmbH (Wertheim, Germany) and cut into 25 mm x 35 mm x 1 mm pieces. Deionized water ($18.2 M\Omega cm^{-1}$) was obtained through a system including pretreatment, reverse osmosis, and ion exchange steps (Elga Option Q15).

Preparation of PDMS-Silica Coatings

Glass microscope slides were cleaned by ethanol and then activated by piranha solution before the coating procedure. Glass slides were immersed into freshly prepared piranha solution, which is a 7:3 (v:v) mixture of concentrated H_2SO_4 and 30% H_2O_2 . They were kept at 80 °C for 2 h and then left at room temperature overnight. Then the glass slides were washed with deionized water and dried at 125 °C for 30 min in an oven. Activated glass slides were used immediately for PDMS-silica composite film preparation.

The coating solution was prepared as follows; the desired amount of silica particles (to obtain x% w/v for the toluene volume, where x is 0, 0.4, 1.2, 2, 3, and 4) were added to 1 % w/v PDMS toluene solution. The polymer solution was mixed with a magnetic stirrer for 1 h and then sonicated for 30 min at room temperature. 10 μL of the homogeneous polymer solution was dropped onto the freshly activated glass slide and spin-coated at

2000 rpm for 60 s. Then the coatings were kept at 100 °C for 24 h in an oven (Figure 1).

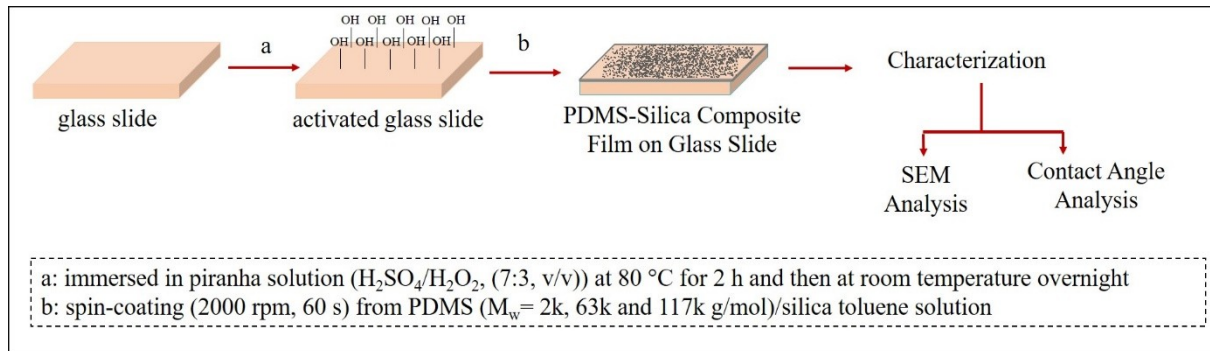


Figure 1: Schematic illustration of the PDMS-silica composite film preparation and characterization procedure.

Characterization of The Coatings

The surface morphology of the coatings was characterized by scanning electron microscopy (SEM, Philips XL 30 SFEG). PDMS-silica composite films were coated with a gold layer before SEM analysis to prevent charging effects. Contact angles were measured by a contact angle meter (KSV-CAM 200) with a PC controlled motorized syringe using ultra-pure water under air at room temperature. Static contact angles (Θ_e) were measured by forming 5 μL drops of water on samples, and the values recorded 2 s after the release of the needle. Advancing contact angles (Θ_a) were measured by increasing the volume of the water droplet on the surface from 3 to 8 μL , and receding contact angles (Θ_r) were measured by decreasing the volume of a water droplet from 10 to 3 μL by using the automatic dispenser through the needle. The needle was kept within the water droplet during the Θ_a and Θ_r measurements. The reported values were calculated by taking the averages of 5 different measurements made on the same sample.

Chemical stabilities of PDMS/silica composite coatings were tested by washing the samples with toluene for 5 minutes and dried at 100 °C for 24 h in the oven. SEM images were again taken, and contact angle (Θ_e , Θ_a , and Θ_r) values were again measured after washing with toluene, and the results were compared with the unwashed samples' results.

RESULTS AND DISCUSSION

In this study, covalently attached superhydrophobic PDMS/silica coatings were prepared on piranha-cleaned glass slides. The superhydrophobic coatings were prepared via a spin-coating of silica/PDMS dispersions and subsequent heating at 100 °C for 24 h. The covalent attachment was achieved via heating, as reported in the literature by McCarthy et al. (28). PDMS with three different molecular weights, hydrophilic, and three different hydrophobic fumed silica were used to prepare the coatings. The effect of hydrophobic fumed silica

concentration on surface morphology and wettability were also studied.

Table 1 shows the effect of fumed silica concentration on the equilibrium (Θ_e), advancing (Θ_a) and receding (Θ_r) contact angle and contact angle hysteresis (CAH) values of composite films. The composites films were prepared by two different molecular weights (63k and 117k g/mol) of PDMS and five different concentration of hydrophobic fumed silica (0.4, 1.2, 2, 3 and 4% w/v) where hydrophobicity is achieved by modification with PDMS (~50 nm; referred as, FS). As seen in Table 1, the Θ_e , Θ_a and Θ_r values of bare PDMS coating increase from $108 \pm 1^\circ$, $111 \pm 1^\circ$ and $96 \pm 2^\circ$ to $119 \pm 1^\circ$, $120 \pm 1^\circ$ and $103 \pm 2^\circ$ respectively as the molecular weight of PDMS increases from 63 k to 117 k. This trend is similar to the results reported in the literature [28]. The Θ_e , Θ_a , and Θ_r values of the coatings increased with an introduction of hydrophobic fumed silica (FS) as depicted in Table 1 regardless of PDMS molecular weight. As the concentration of hydrophobic fumed silica (FS) increased from 0.4 to 4 % w/v, and increase in Θ_e , Θ_a and Θ_r values were seen up to $169 \pm 2^\circ$, $173 \pm 1^\circ$, $166 \pm 1^\circ$, and $170 \pm 2^\circ$, $172 \pm 2^\circ$ and $164 \pm 1^\circ$ for the coatings prepared by 63k and 117k PDMS respectively (Table 1). The highest contact angle values and lowest CAH (7° for 63k PDMS and 8° for 117k PDMS) were obtained at 4 % w/v silica concentration for PDMS coatings prepared by different molecular weights. The highest Θ_e value (164°) of cross-linked silicone-based coatings prepared by spray coating using $\text{CaCO}_3/\text{SiO}_2$ particles on glass by Yang et al. (27) was lower than our results. Manoudis et al. reported that they obtained siloxane-based coatings via spraying on glass using 2% w/v hydrophilic silica particles with a Θ_a of 164° and CAH of 3° . The CAH hysteresis value (3°) is better than our values (7° for 63k PDMS and 8° for 117k PDMS), but the information on the chemical composition and the molecular weight of the poly(alkyl siloxane) which was used to prepare the coating was not given in the paper (14).

Table 1. Equilibrium (Θ_e), advancing (Θ_a) and receding (Θ_r) contact angle and contact angle hysteresis (CAH) values of PDMS/SiO₂ composite films prepared by hydrophobic fumed silica (FS) and PDMS with different molecular weights. The PDMS (1% w/v) was prepared by dissolving PDMS (63k or 117k) in toluene.

PDMS (M _w (g/mol))	Silica/toluene (% w/v)	Θ_e (°)	Θ_a (°)	Θ_r (°)	CAH (°)
63k	0	108±1	111±1	96±2	15
63k	0.4	119±2	124±1	108±2	16
63k	1.2	161±1	168±2	150±1	18
63k	2	167±1	171±1	155±2	16
63k	3	169±1	172±2	164±1	8
63k	4	169±2	173±1	166±1	7
117k	0	119±1	120±1	103±1	17
117k	0.4	129±2	146±1	107±1	39
117k	1.2	166±2	167±1	142±1	25
117k	2	167±1	168±1	134±1	34
117k	3	169±2	172±1	159±2	13
117k	4	170±2	172±2	164±1	8

The Θ_e , Θ_a , Θ_r , and CAH values of bare PDMS and composite coatings before and after washing with toluene were shown in Table 2. The coatings were prepared using PDMS with different molecular weights (2k, 63k, and 117k) and 4 % w/v hydrophobic fumed silica (FS). As seen in Table 2, the Θ_e , Θ_a , Θ_r values of the bare 2k PDMS and 63k PDMS coatings were slightly decreased (1-2°) or increased (1-2°) after washing with toluene, but the decrease in Θ_e , Θ_a , Θ_r values of the coating prepared using 117 k PDMS were higher compared to the other coatings. The increase in all the contact angle values was seen as the molecular weight of the PDMS rises, as shown in Table 2, and the trend did not change after washing with toluene. Similar to the bare PDMS coatings, there was a slight change (1-2°) in the Θ_e , Θ_a , Θ_r values of the composite coatings prepared using 4 % w/v silica (FS) after washing with toluene. The results in Table 2 clearly show that bare PDMS and composite coatings were covalently attached to the piranha cleaned glass slides, and the superhydrophobic character of the composite coatings did not alter after washing with toluene.

Figure 2 shows the SEM images of the coatings prepared by PDMS with different molecular weights PDMS/silica (FS) composite before and after washing with toluene. All the SEM pictures in Figure 2 a-c clearly show that the homogenous PDMS/silica (FS) composite coatings were obtained with a

similar morphology regardless of the PDMS molecular weight. No change in the homogeneity of the composite coatings was observed after washing with toluene. (Figure 2 d-f).

After investigating the effect of silica concentration on coatings' wettability, 4 % w/v silica concentration and 63k PDMS were chosen to study the effect of silica size and type on coating morphology and wettability (Table 3 and Figure 3). The Θ_e , Θ_a , Θ_r , and CAH values of PDMS/silica composite films prepared using different types and sizes of fumed silica were shown in Table 3. The Θ_e contact angles of the PDMS coating increased up to 159-172° with an addition of different types and sizes of fumed silica (Table 3). Although high Θ_a values were obtained for the coatings prepared by hydrophilic fumed silica (N20) (168±2°) and dimethylsiloxy modified hydrophobic fumed silica (H15) (N20) (163±2°), the surfaces were not superhydrophobic due to the high CAH hysteresis (24° and 16° respectively). Superhydrophobic composite coatings were obtained when two different sizes of PDMS modified fumed silica particles (FS; ~50 nm and H18; ~ 30nm) were used to prepare the coatings. The Θ_a , Θ_r , and CAH values of the coatings prepared using FS and N18 were measured as 173±1°, 166±1°, 7°, and 174±1°, 166±1°, 8° respectively. These differences can be due to the incompatibility of the hydrophilic (N20) and dimethylsiloxy modified hydrophobic (H15)

fumed silica and compatibility of the PDMS modified hydrophobic (FS and H18) fumed silica with the PDMS matrix.

Table 2. Equilibrium (θ_e), advancing (θ_a) and receding (θ_r) contact angle and contact angle hysteresis (CAH) values of PDMS/SiO₂ composite before and after washing with toluene. The composite films composed of hydrophobic fumed silica (FS) and PDMS with different molecular weights. The PDMS (1% w/v) was prepared by dissolving PDMS (2k, 63k, or 117k) in toluene.

PDMS (M _w (g/mol))	Silica/toluene (% w/v)	Washing	θ_e (°)	θ_a (°)	θ_r (°)	CAH (°)
2k	-	No	94±1	95±2	81±1	14
2k	-	Toluene	92±2	99±2	82±1	17
2k	4	No	170±1	173±2	166±1	7
2k	4	Toluene	168±1	172±1	162±1	10
63k	-	No	108±1	111±1	96±2	15
63k	-	Toluene	109±1	110±1	96±1	14
63k	4	No	169±2	173±1	166±1	7
63k	4	Toluene	168±1	172±1	165±2	7
117k	-	No	119±1	120±1	103±1	17
117k	-	Toluene	110±2	117±1	98±1	19
117k	4	No	170±2	172±2	164±1	8
117k	4	Toluene	171±2	173±2	165±1	8

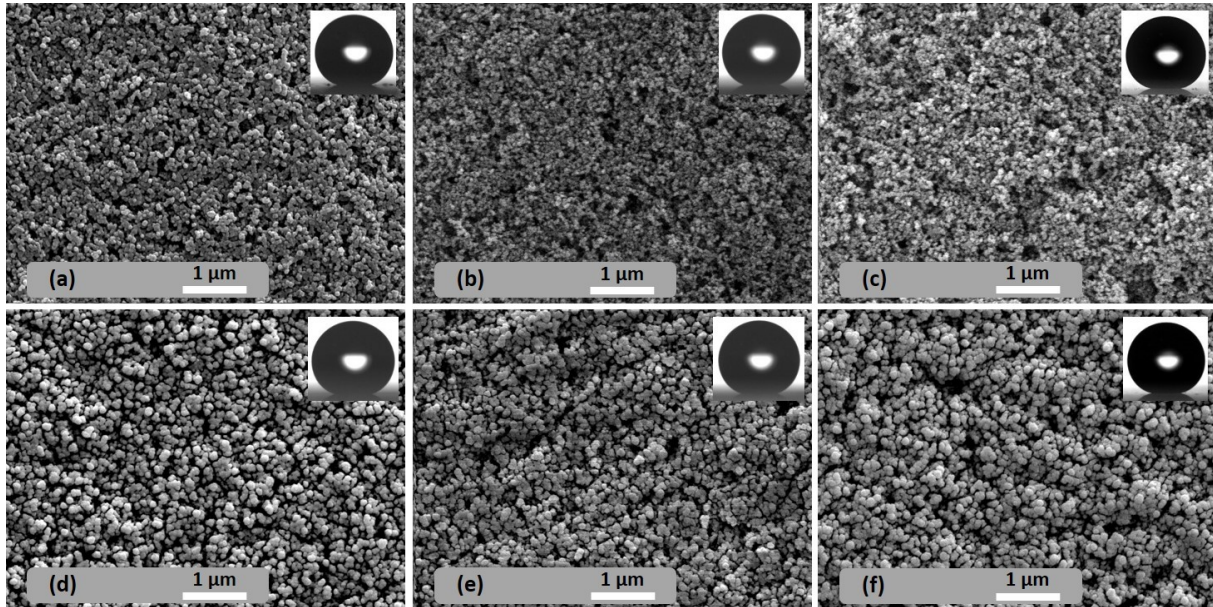


Figure 2. SEM pictures and water droplets of PDMS-silica composite films prepared with fumed silica (FS) and PDMS with different molecular weights : (a) 2k PDMS; (b) 63k PDMS; (c) 117k PDMS; (d) 2k PDMS, washed with toluene; (e) 63k PDMS, washed with toluene; and (f) 117k PDMS, washed with toluene (Silica concentration is 4 % w/v).

Table 3. Equilibrium (θ_e), advancing (θ_a) and receding (θ_r) contact angle and contact angle hysteresis (CAH) values of PDMS/SiO₂ composite films with hydrophilic and three different hydrophobic fumed silica and PDMS (63k). The PDMS (1% w/v) was prepared by dissolving PDMS (63k) in toluene.

Silica type	Silica/toluene (% w/v)	θ_e (°)	θ_a (°)	θ_r (°)	CAH (°)
-	-	108±1	111±1	96±2	15
FS	4	169±2	173±1	166±1	7
N2O	4	163±2	168±2	144±2	24
H15	4	159±1	163±1	147±1	16
H18	4	172±2	174±1	166±1	8

SEM pictures of the PDMS/fumed silica coatings which were prepared using 4% w/v PDMS modified hydrophobic fumed silica (FS and H18) and dimethylsiloxo modified fumed silica (H15) and hydrophilic fumed silica (N2O) are depicted in Figure 3 a, b, c, and d respectively. Figure 3 a and b shows that the coatings prepared using FS and N10 are homogenous and very small aggregates of the fumed silica were seen on the surfaces confirming

the compatibility of the PDMS modified fumed silica with the PDMS matrix. Distinct big aggregates of fumed silica, which is embedded in the PDMS matrix and non-homogeneous coating formation were observed as seen in Figure 2 c and d when dimethylsiloxo modified fumed silica (H15) and hydrophilic fumed silica (N2O) were used conforming the non-compatibility of the particles with the PDMS matrix.

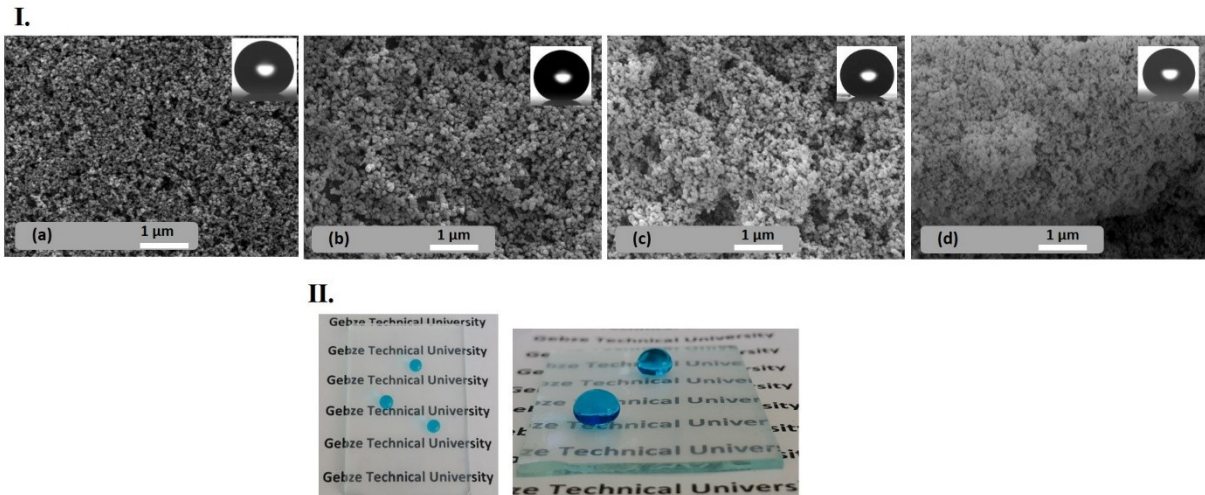


Figure 3. I. SEM pictures and water droplets of PDMS-silica composite films prepared with 63k PDMS and three different hydrophobic and hydrophilic fumed silica particles: (a) FS (b) H18, (c) H15 and (c) N20 (Silica concentration is 4% w/v). II. Photographs of superhydrophobic PDMS-silica composite film (63k PDMS, 4% w/v FS) coated glass slide (water droplets were dyed with methylene blue).

CONCLUSIONS

Summarizing, in this paper, we have demonstrated the preparation of covalently attached superhydrophobic PDMS/silica composite coatings on activated glass slides via an easy and straightforward spin-coating process. SEM and contact angle measurements were used to characterize the morphology and the wettability of the surfaces. The superhydrophobic coatings were obtained when PDMS modified fumed silica (FS and H18) were used, and the concentration of the fumed silica was 4 % w/v whereas the superhydrophobic surfaces were not obtained when hydrophilic (N2O) and dimethylsiloxy-modified (H15) fumed silica were used due to the high CAH. To obtain a superhydrophobic surface using PDMS and fumed silica via a spin coating process, the type and the concentration of the fumed silica need to be optimized.

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