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Wettability of Liquid Mixtures on Porous Silica and Black Soot Layers

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ABSTRACT

ophisticated manipulation of surface roughness and solid surface energy are widely used to design super-hydrophobic layers. In this work, we designed highly porous silica layer with contact angle (CA) of 145°, and its robustness was promoted with thermal treatment. Wettability of coated layer is studied with CA measurement for different liquid surface tensions using diluted organic solvents (ethanol, acetone or ethylene glycol) with distilled water. Zisman plot is also used to estimate the surface energy of the porous silica. The results are compared with coated black soot as a hydrophobic layer. Moreover, thin layer of Au (~10 nm) was coated on the porous silica layers to study the effects of chemical properties on the wetting behavior. The thickness of the Au thin layer is negligible compared to the surface roughness, so the drastic decrease in CA rates can be related to the layers' porosity and the affinity of solvents to liquid/air and liquid/solid interfaces. Prog. Color Colorants Coat. 13 (2020), 239-249© Institute for Color Science and Technology.

1. Introduction

Recently, tremendous efforts have been made to develop various artificial super-hydrophobic surfaces through mimicking and inspiring surface morphologies available in nature. Historically speaking, initial attempt through reaching contact angle near 180° was done by Ollivier via coating a substrate with soot in 1907 [1]. One of the common characteristics of superhydrophobic surfaces is the self-cleaning property which means that the drops roll-off easily on the surface and thus remove any contamination [2]. When water droplet sits on the apex of nanostructures, an air layer is trapped within the roughened or textured surface. Thus, it causes a substantial increase in contact angle and a significant reduction in roll-off angle and contact angle hysteresis [3]. This feature has been widely used by plants surfaces, animals, and insects to survive from many nature hazards [4-6]. In addition, these surfaces have attracted a lot of interest both in academia and industrial applications such as airplane, wind turbines, photovoltaic devices [7], electric power lines [8], ceramic pigments and coatings [9, 10] and ships because of their anti-corrosion [11], anti-frosting and anti-icing [12] which lead to energy-saving [13].

When a liquid drop is located on a solid surface, its edge usually forms a defined angle that on liquid and surface properties. This angle is called contact angle, CA [14]. Figure 1 presents a graphical representation of a droplet on a surface. Surfaces with contact angles above 150° and low contact angle hysteresis, which able to repel water, ethanol and oils, are known as super-omniphobic surfaces. Nevertheless, designing a surface can repel both water and organic liquids simultaneously is a challenging problem. Hereby, super-hydrophobicity and super-oleophobicity are the two distinct surface effects having the capability to repel water and oil, respectively. However, since the surface tension of water and oil are sufficiently different, it is possible to separate oil from water. Ju [15] invented a chemical and equipment-free strategy to separate oil from water through a facile one-step process of candle soot deposition.

Because of high thermal resistance and transmission properties, porous silica has a lot of practical applications in optical devices, solar cells, optical filters, etc [16-21]. Naturally, hydroxyl groups existing on the silica surfaces make this material intrinsically hydrophilic. Nevertheless, high porosity can trap a layer of air underneath the liquid droplet and thus promotes hydrophobicity. Moreover, Cassie-Baxter regime presents surface topography and has a profound impact on the wettability. When probe liquid fills nanoscale-roughened surface, Wenzel regime predicts that surface roughness can make a surface wettable ($\theta < 90^{\circ}$) or non-wettable ($\theta > 90^{\circ}$) depending on the surface chemical properties [2]. When a small drop sits on a surface, it forms an equilibrium shape to minimize the total surface energy [22]. Modifying silica surface with silane is widely used to promote hydrophobicity. Efficiency of silane on the silica surface was investigated at different pH values and showed that the maximum grafting of silica and silane occurs at isoelectric point of silane (4.7) and above isoelectric point of silica (5-6) [23]. It has been also reported [2] that the first development on these superoleophobic surfaces have been done by Tuteja et al. [24] through the design of re-entrant surface curvature together with chemical composition and the roughened texture surface. By this way, combination of surface roughness (micro-nano combined) and low surface free energy is a successful way to fabricate superamphiphobic or super-omniphobic surfaces [25-27].



Figure 1: Water droplet on a porous silica surface. θ is CA and γ_{SV} , γ_{LV} , γ_{SL} are solid–vapor, liquid–vapor, solid–liquid interfacial free energies, respectively.

Recently, investigating wetting behavior of coated layers with different liquid mixtures is widely used [28-34]. Wettability depends both on probe liquid and solid surface. Hereby, using different liquid surface tension is an easy way to study solid surface characteristics.

Coating of aerogels is a highly challenging problem since stable fluidized beds can be hardly obtained exceptionally for low density aerogels [35]. Amorphous carbon nanoparticles network, which sometimes referred to as carbonaceous films obtained from flame synthesis, is highly interesting templates for making super-repellent surfaces and oil-water separation. Carbon black soot counts as a material with high porosity and fractal dimension. Bayer et al. [36] reported several new techniques regarding fabrication of robust amorphous carbon nanoparticles network on soft micro-rough polymeric surfaces. Carbon black soot layer with high fractal structure and porosity is a good hydrophobic surface with CA nearly to 132° [37].

In this study, we designed highly porous hydrophobic silica coating and investigated synergism in its wetting properties with different liquid surface tensions which are prepared by diluting organic solvents (ethanol, acetone or ethylene glycol) with distilled water. Carbon black soot, another porous material, is also used to compare its wetting properties with silica aerogel layer. Then, synergism in wetting phenomena and solid surface tensions are explored using CA experimental data. The effect of plasma treatment on the layers is also presented. Finally, a thin layer of gold (Au \sim 10 nm) is deposited on the coated layers to reveal the impacts of surface chemical properties on the wettability.

2. Experimental

2.1. Fabrication of porous coatings

Silica aerogel (Aerulate, Nano Sav, mean pore diameter of 20 nm, 60 Kg/m³) was used for coating on glass substrate without further purification. In a typical procedure, the suspension was prepared by dissolving 0.15 g silica aerogel in 12 mL ethanol (96%) under stirring for 10 min and then sonication for 24 min at room temperature, 25 °C. A thin layer of porous silica was coated on glass substrate (1.5×3.0 cm) with dip coating technique (V~2.0 mm/s). Then, it was heated to 300 °C for 1 h. This sample is labeled as S1 and its wetting properties are studied using different liquid

surface tensions. In addition, a clean glass substrate was held in the flame of paraffin candle to deposit a few micrometer of carbon soot at room temperature and atmosphere, labeled as S2. This layer with high water repellency was also prepared to compare its wetting properties with the porous silica layer.

To explore the influencing factors on the surface wetting properties, droplets with different liquid surface tensions were put on the coated layers. In this way, organic solvents such as ethanol (96%, Sigma-Aldrich), acetone (≥99.5%, Sigma-Aldrich) or ethylene glycol (Merck, 99%) were diluted with distilled water to provide different liquid surface tensions. Then, static CA was measured for various liquids. The average of CA values for three tiny drops are reported. Moreover, to study the influence of chemical properties on the wettability, 10 nm Au layer was deposited on the S1 and S2 samples via sputtering method. Citric acid (≥99.5%, Sigma-Aldrich) was also used to examine the resistance of the coated layers against organic acids. To investigate the effects of plasma on the samples, they were treated with plasma (Edwards-vas) under pressure of 5×10^{-1} bar for 10 min. Current and voltage were 4 mA and 0.4 kV, respectively.

2.2. Apparatus

Au layer was coated via sputtering system (Nanostructured Coating Co.) under pressure of 10⁻³ mbar for 100 s. The coated layer structures were observed via JEOL JSM-6700F field-emission scanning electron microscope (FE-SEM) at 15 kV. The three-dimensional topologies were analyzed with atomic force microscopy (AFM, SPI3800N, Seiko Instruments Inc., Japan). Contact angle measurements were performed by using Dino-lite AM 2011 digital microscope. CA measurement was done in such a way that instrument's crosshair is tangent to cross-sectional curvature of the droplet at a point where all three media (solid, liquid, and vapor) meet. Plasma apparatus (Edwards-vas) was used under clean condition for thermal treatment.

3. Results and Discussion

Figure 2 shows the FE-SEM images of silica and black soot structures at different magnifications. Figures 2a, 2b and 2c belong to S1 sample, while 2d, 2e and 2f belong to S2 sample. It can be seen that particles are agglomerated to some extent and both of the coated layers are porous.



Figure 2: FE-SEM images of coated silica (S1-a, S1-b and S1-c) and black soot layers (S2-a, S2-b and S2-c) at different magnifications.

Particle size distribution was obtained using FE-SEM images of silica and carbon layers. In this way, Image J software is used to perform the image processing of particles. Figure 3 shows the particle size distribution histogram for silica (Figure 3a) and black soot (Figure 3b) layers. The total numbers of detected particles are 1596 and 2441 for silica and black soot, respectively. As can be seen in Figure 3a, there is more or less a random distribution for silica particles. However, the population of particles is larger for 11 and 17 nm in size. Figure 3b represents the histogram of black soot particles which is fitted by Gaussian function. The average particle size of black soot is approximately 17 nm. As can be seen, particles are agglomerated to some extent for both coatings. Aggregation of particles basically occurs when attractive van der Waals force dominates the repulsive electrostatic force. Particles with low surface charge density are easily aggregated since double layer repulsion is too weak to overcome van der Waals force. Hence, particles are aggregated to form dimer, trimer or larger clusters depending on the magnitude of the surface charge density [38].

Figure 4 shows AFM images of silica layer (S1 sample). AFM results reveal that average roughness (Ra) and root mean square roughness (Rq) are 103 and 137 nm, respectively. S2 sample was also investigated under the same conditions. However, no clear results were obtained mainly because of the surface softness and non-uniformity.

Liquid surface tensions were extracted from other literatures [39-41] and some of them are presented in Table 1. As can be seen from Table 1, raising the concentration of ethanol, acetone and ethylene glycol gradually affects the solution surface tension.



Figure 3: Particle size distribution histograms of (a) silica and (b) black soot particles coated on substrate.



Figure 4: Different views of AFM image for silica layer. Ra and Rg are 103 nm and 137 nm, respectively.

Liquids	Surface Tension in mN/m for water content (%)				
	100	80	60	40	20
Water	72	-	-	-	-
Ethanol	21.82	23.82	26.23	30.16	37.97
Acetone	23.1	27.6	29.4	33.0	40.5
Ethylene glycol	48.2	-	-	-	64.9

Table 1: Liquid surface tension data as a function of the mass percent of the non-aqueous component [39-41].

To examine surface wetting properties, we used 1 μ L droplets with different surface tensions on coated layers. Figure 5a represents CA changes versus water content for different solutions on S1 sample. Porous silica layer has the CA close to 145° for distilled water. CA for diluted ethanol and acetone falls steeply as their concentration increases (~70% water content). It is also clear that a droplet containing 20 wt% water (80 wt% ethanol or acetone) completely wets the porous silica layer. Nevertheless, this is not the case for ethylene glycol. Thus, there is a plateau for ethylene glycol in Figure 5a, and CA approximately keeps constant at about 120°. Since ethylene glycol has larger surface tension, liquid merely tends to spread on the surface

and hence ethylene glycol has larger CA compared to the two other solvents, i.e. acetone and ethanol. Surface roughness and liquid/solid surface tensions are the most important parameters highly impact the surface wetting properties. In this way, Young–Laplace equation reveals the relationship between contact angle and surface tensions. So, organic materials, whose surface tensions are $20 \leq \gamma_{LV} \leq 48$, will spread on a surface if solid-vapor surface tension (γ_{SV}) is larger than that of the solid-liquid (γ_{SL}). When organic solvents are diluted with distilled water, contact angle changes as a result of modifying the liquid surface tension. In this way, the critical turning point is the condition under which γ_{SV} dominates γ_{LV} .



Figure 5: Average CA vs. water content for three small drops located on the surface of (a) porous silica layer (S1 sample) and (b) black soot (S2 sample). Organic solvents have been diluted with distilled water to cover different liquid surface tensions. Solvents are ethanol, acetone and ethylene glycol whose densities are 0.8076, 0.79 and 1.11gr/cm³, respectively.

Zisman plot based on the Zisman theory is a graphical method to quantitatively measure the wettability and critical surface tension (γ_c). To determine the solid surface tension, an empirical expression is used for equation-of-state (EOS) [39, 40] Eq. 1:

$$\cos\theta = -1 + 2\sqrt{\frac{\gamma_s}{\gamma_L}}e^{-\beta(\gamma_L - \gamma_s)^2} \tag{1}$$

where β is a constant, θ is the surface contact angle, γ_L and γ_S are the liquid and solid surface tensions, respectively. Figures 6 and 7 show Zisman plots for silica and black soot, respectively. In this way, γ_S and β are estimated via fitting EOS equation to experimental data.



Figure 6: Zisman plot for silica layer and corresponding EOS fitting to experimental data. Output results: β = 0.01954 and γ_{s} = 25.82 mJ/m².



Figure 7: Zisman plot for black soot layer. β and γ s are determined based on EOS fitting to experimental results. Fitting results are: β = 0.0065 and γ_{S} = 29.9 mJ/m².

To study the dependency of morphology and surface roughness on CA changes, carbon black soot (S2 sample) was deposited as a highly porous material on glass substrate to compare the results. We investigated this well-known hydrophobic layer to reveal important parameters affecting its wettability. Water contact angle for S2 sample is close to 132°. We again used solutions with different liquid surface tensions to investigate the surface wetting behavior under the influence of different organic solvents. Figure 5b illustrates the wettability of different organic solutions on S2 surface. CA gradually decreased by increasing the concentration of ethanol, acetone and ethylene glycol. In this way, CA for drops containing mixture of water with ethanol or acetone decreases from approximately 130° with the critical water content of 70-80 wt% (~20-30 wt% ethanol or acetone). However, in the case of ethylene glycol, it happens at 50% for water content. There is a critical region in which transition suddenly occurs from hydrophobicity to hydrophilicity. This transition is originated from the fact that γ_{SL} dominates γ_{LV} at this critical point. Comparing the results indicates that steep falling of CA is available for both S1 and S2 samples. However, this situation is steeper for S2 sample. Hence, based on these results, two factors may affect the sudden transition from hydrophobic to hydrophilic, porosity and surface chemical composition.

To investigate the effect of surface chemical composition on wetting properties, S1 and S2 samples were coated with 10 nm gold layer via sputtering technique. Figure 8 shows AFM images of S1 sample after coating with Au. This amount is negligible compared with surface roughness and thus morphology does not relatively changed through this Au coating. In this way, the average roughness and root mean square roughness (Ra and Rq) are 106 nm and 139 nm, respectively. So, we changed the chemical properties of coated layers while keeping the roughness constant in order to study the impacts of chemical properties on the wettability.

In the next step, we examined CA for modified silica and black soot layers with different concentrations of diluted ethanol. In this case, water CA for modified silica layer is 124°. The results are represented in Figure 9. Comparing Figure 9a with Figure 5a indicates a significant shift. Coating of Au thin layer on porous silica layer makes the surface more hydrophilic. Surface topography results based on atomic force microscope reveals that surface roughness did not change in the presence of the Au layer. Thus, a steep falling which was inferred from surface roughness and liquid surface tensions are now shifted to the more diluted concentrations. This shift may be due to the chemical properties synergism into wettability. Figure 9b illustrates that changing the surface energy of the porous silica layer via Au deposition shifts the transition point into more diluted concentrations. It can be also concluded from comparing Figures 9 and 5 that although the surface composition affects both surface energy and CA, it does not change the sudden transition from hydrophobicity to hydrophilicity.

We have also coated the same thickness of Au layer (10 nm) on a bare glass substrate and measured its CA to investigate and compare its wetting properties with previous layers. Figure 10 represents CA for various concentrations of ethanol solvent on a bare glass containing Au thin layer. Water CA is close to 61° for

this layer. It can be seen that CA gradually decreases from 61° to zero. It is also interesting to report that there is not any steep falling or sudden transition in hydrophobicity/hydrophilicity. It would be also crucial to mention that a bare glass is initially flat without any morphologies.

Figure 11 illustrates the resistance of coated layers to the citric acid. It is clear that coated layers are resistant to the organic acid. Our findings show that surface chemical composition does not mainly affect sudden transition in wettability. The hydrophobicity/ hydrophilicity transition may be mostly due to the surface roughness and porosity.

Moreover, plasma treatment has been also used under the pressure of 5×10^{-1} bar, I = 4 mA, V = 0.4 kV for 10 min to study the effects of plasma on the silica and black soot layers. Figure 12 shows that silica is completely converted to a hydrophilic layer, while black soot layer remains unchanged.



Figure 8: Surface topography for S1 sample coated with 10 nm Au layer.



Figure 9: Wettability of modified silica and black soot with Au deposition. Probe liquid is diluted ethanol which has different liquid surface tension. (a) S1 and (b) S2 both modified with approximately 10 nm Au deposition.



Figure 10: CA of different concentrations of ethanol on bare glass coated with Au thin layer.



Figure 11: Resistance of layers to citric acid: (a) silica, (b) modified silica with Au layer, (c) black soot and (d) modified black soot with Au layer.



Figure 12: Effects of plasma on the (a) silica and (b) black soot layers.

Equipments required perform surface to measurement are usually specialized and expensive [44]. Hence, using probe liquid to study the surface wettability has attracted the attention of many researchers. Hansson et al. [45] studied the wetting and interaction forces of ethanol/water mixtures on silicon wafer surface with pore spacing of 1.4- 4.0 µm. Their results concerning sudden decrease for static CA is in compatible with ours. They also reported that surface pore depth does not influence the wetting while pore spacing has a minor effect. So, theoretical predications based on surface geometry and three-phase contact line such as Wenzel and Cassie-Baxter are compatible with experiments on water/ethanol mixtures on porous silica. Although both water and ethanol penetrate into pores, AFM measurements showed that air cavitation is formed upon locating a water drop on the pores. However, small increase in ethanol concentration results in weaker adhesion force; hence the mixture penetrates into pores and removes the air. Heng [46] also reported that liquid properties as well as surface roughness and chemical composition have important impacts on wettability. So, they manipulated surface wettability by controlling the concentration of ethanol/water on the thiol-modified copper meshes. Declining in CA (as a result of increasing the ethanol content) was faced by two sudden decreases located at 50 and 90% ethanol volume fraction in mixture. Finally, cumulative effect of ethanol concentration leads this surface lyophilic. However, more fraction of thiols promotes lyophobic effects and sudden downfalls happen at larger ethanol concentration. Dufour [47] suggested to use mixtures instead of liquids of various chemical natures since it enables

tuning of surface energy while minimizing variations in density, viscosity and liquid-solid interactions. They also stated that CA hysteresis remains unchanged for the water/ethanol mixtures up to about 40% water content but at lower water concentration, receding CA is saturated which actually attributed to surface structure. Synthesizing nanoparticles with functional groups is another way to alter the super-hydrophobicity or super-hydrophilicity [48]. It has been also reported that anionic surfactant is more efficient to reduce water CA on the porous hydrogel silica [49].

4. Conclusions

Sophisticatedly combination of appropriate surface roughness and low surface energy material are a wellknown way to design superhydrophobic layers. Although silica is naturally categorized as a hydrophile material due to its Si-OH surface groups, air trapping in porous silica leads sitting liquid drops to slip easily. In this way, we fabricated highly porous silica and black soot layers on glass substrates and then some thermal treatment has done to promote their mechanical robustness. Nevertheless, hydrophobicity of silica is removed under the influence of plasma. It has been also presented sufficient resistance of coated layers to organic acids.

Wettability of fabricated hydrophobic silica layers has been studied by measuring the contact angle for different liquid surface tensions, prepared with mixtures of distilled water with other solvents (ethanol, acetone or ethylene glycol). There are steep falling CA rates with transition from hydrophobic to hydrophilic is observed for ethanol and acetone. But, this situation is more or less kept constant for ethylene glycol. Moreover, Zisman plot was used to estimate the surface tension of coated silica. The results were also compared with black soot layer as a highly porous material.

To investigate the impacts of chemical properties on the wetting behavior, thin layer of Au (~10 nm), which was negligible compared to the surface roughness, was coated. Results indicated that a

5. References

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significant shift occurs as a consequence of gold chemical nature for the wetting and also steep falling CA rates are still available. Thus, we can conclude that these sudden transitions are mainly attributed to layers porosity and different affinities of the solvents to the liquid/air and liquid/solid interfaces. While, surface chemical properties seem to shift this critical point into more diluted concentrations.

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