A Facile Modifier-free Approach to Fabricate Antistatic Superhydrophobic Composite Coatings with Remarkable Thermal Stability and Corrosion Resistance

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Abstract

Research on antistatic superhydrophobic surfaces has attracted widespread attention in some fields. However, in the application of superhydrophobic materials, fabricating stable and practical superhydrophobic surfaces through facile and low-cost approaches still faces considerable challenges. Herein, a polyphenylene sulfide (PPS)-based antistatic superhydrophobic composite coating with a high water contact angle (166°) and a low sliding angle (2°) was fabricated on a Q345 steel surface through a simple spray-coating method without any modifier. Furthermore, the as-prepared superhydrophobic coating also displayed excellent superhydrophobicity for water droplets at different pH values, as well as self-cleaning, anti-fouling and anti-icing properties. Importantly, the superhydrophobic coating still exhibited superhydrophobicity after calcination at 350°C for 1 h, indicating its outstanding thermal stability. Excellent antistatic and anticorrosion properties were obtained on the prepared coating surface, which allows the coating to be applied under harsh conditions. Benefiting from the above characteristics, compared with the commercial coating, the as-obtained antistatic superhydrophobic coating may be applied more widely in related fields.

Keywords: no-modifier, superhydrophobicity, thermal stability, antistatic property, anticorrosion

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1 Introduction

Metal materials are widely used in various fields because of their excellent comprehensive properties^[1]. Nevertheless, poor corrosion resistance seriously affects their service life and limits their further applications^[2]. It is well known that a surface with superhydrophobicity can play a good protective role on the matrix. Superhydrophobic surfaces, inspired by plants and animals in nature, show significant importance in fundamental research and industrial applications such as corrosion-resistance^[3–5], self-cleaning^[6,7], anti-icing^[8–10], and drag reduction^[11,12]. They possess static contact angles of Water Contact Angles (WCAs) higher than 150° and Sliding Angles (SAs) lower than 10° ^[4–6]. Various methods, such as sol-gel^[13–15], template^[16,17],

self-assembly^[18,19] and etching^[20–22], have been adopted to fabricate superhydrophobic surfaces.

Coating is an important method for surface modification of materials^[23]. Furthermore, spraying, which is not limited by the shape and size of the sample surface, is a simple and efficient surface treatment method^[24]. In some special application fields, such as coal mining, aviation, defence and petrochemical applications, the coating should also possess excellent antistatic property and the surface resistance is usually less than $10^6 \Omega$ ^[25,26]. In recent years, with the development of science and technology, multifunctional superhydrophobic surfaces with outstanding antistatic performance have become a research focus in the superhydrophobic field^[27,28]. Owing to the excellent electrical properties, electrostatic charges cannot

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accumulate on the antistatic superhydrophobic surface, thus avoiding the hazards caused by electrostatic accumulation in special application fields^[29]. Furthermore, antistatic superhydrophobic coatings also play an important role in waterproof electronic devices and electromagnetic interference shielding^[30,31]. Carbon-based materials are often used to prepare conductive coatings because of their excellent conductivity. Currently, a series of carbon-based conductive superhydrophobic coatings have been obtained^[32–34]. For instance, Das *et al.*^[32] fabricated a highly conductive superhydrophobic composite coatings filled with carbon nanofiber/PTFE polymer, which is the first study to report data on the effectiveness of conductive superhydrophobic composite coatings as ElectroMagnetic Interference (EMI) shielding materials. By partially embedding fluorinated graphene into ThermoPlastic Polyurethane (TPU), Wang *et al.*^[33] proposed a dissolution and re-solidification method to manufacture a superhydrophobic composite with highly stretchable and excellent electromechanical properties. Yao *et al.*^[34] employed a simple spraying method to prepare superhydrophobic nanocomposite coatings, composed of spherical SiO₂ nanoparticles and fluorinated Multi-Walled Carbon Nanotubes (MWCNTs). The obtained coatings display fine transparency and conductivity. However, most of the reported surfaces were either modified with low surface energy modifiers or based on complex preparation processes, seriously hindering their further applications in practice. In addition, these surfaces display poor thermally stable superhydrophobicity (below 200 °C), limiting the engineering value of them. Thus, it is more feasible to develop conductive superhydrophobic surfaces with good thermal stability by economical and widely available methods.

As a result of excellent conductivity and chemical stability, CNFs are widely applied in industrial production^[35,36]. Rare earth oxides have been reported to show intrinsic hydrophobicity and still maintain stable hydrophobicity at 1000 °C^[37]. Cerium dioxide (CeO₂), a widely used rare earth oxide, has been attracted intensive interest owing to its excellent properties^[38]. Thus three-dimensional CeO₂ particulates can not only provide rough structural support for superhydrophobic

coatings, but also improve the thermally stable superhydrophobicity of the coatings. Additionally, polyphenylene sulfide (PPS), as a special engineering plastic, possesses excellent physical and chemical properties, including high flame retardancy, radiation resistance, good thermal stability and excellent electrical properties^[39]. As a coating, PPS has good bonding properties to the surface of carbon steel, cast iron, aluminum, glass and ceramics^[40]. With excellent chemical stability, corrosion resistance and low surface energy, polytetrafluoroethylene (PTFE) is an ideal candidate material for surface modification^[41]. Some studies related to PPS coatings by blending PTFE have been applied to prepare superhydrophobic surfaces^[42–44]. Up to date, to our best aware, there are few reports on antistatic PPS/PTFE superhydrophobic composite coatings. It is vital and challenging task to fabricate antistatic superhydrophobic coatings with good thermal stability in industrial applications.

Herein, a simple, modifier-free and efficient method is applied to prepare antistatic superhydrophobic coatings with good thermal stability on the surface of alloy steel. The technology involved combining mechanical mixing and spraying, which is potentially applicable to the large-scale fabrication of conductive superhydrophobic coatings. Using this simple method, CeO₂ particulates were fabricated by an oil bath method, and the surface roughness was achieved by blending CeO₂ particulates and modified CNFs. Afterwards, a certain amount of PPS, PTFE and PDMS were added to the above mixed particles to generate a grey suspension, which could be used for spraying on the surface of alloy steel. The prepared coating, with a high WCA (166°) and low SA (2°), exhibited excellent hydrophobic properties. Furthermore, the superhydrophobic coating also displayed great antistatic property, corrosion resistance, thermal stability, fouling resistance and freezing resistance. It is believed that the processes elaborated in this article possess practical certain application value and reference significance for superhydrophobic research.

2 Experiment

2.1 Materials

Q345 steel sheets (30 mm × 20 mm × 0.2 mm) were purchased from Shanghai Baoshan Iron & Steel Co., Ltd.

CNFs 100 nm in diameter and 20 μm – 200 μm in length were obtained from Sigma-Aldrich Co., Ltd. Cerous nitrate ($\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$) was supplied by Chengdu Kelong Chemical Reagent Factory. Urea ($\text{CO}(\text{NH}_2)_2$, 99.0%) was provided by Shantou Xilong Chemical Co., Ltd. Tetrapropylammonium bromide (TPAB) was obtained from Sinopharm Chemical Reagent Co., Ltd. Poly (1,4-phenylene sulfide) (PPS, average Mn: 1×10^4 , yellow powder) was purchased from Sigma-Aldrich Co., Ltd. PTFE powder (500 mesh – 600 mesh) was supplied by Shanghai Weiwei Plasticizing Co., Ltd. Polydimethylsiloxane (PDMS) was provided by Dow Silicones Corporation in the USA. The other reagents were used as received without further purification.

2.2 Preparation of 3D flowerlike CeO_2 particulates

The 3D flowerlike CeO_2 particulates were synthesized with a simple procedure based on previous literature^[45]. In short, 3.7 g $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, 4.5 g $\text{CO}(\text{NH}_2)_2$ and 10.0 g TPAB were added into a round flask containing 300 mL ethylene glycol. Afterwards, the above mixed solution was magnetically stirred at 180 $^\circ\text{C}$ for 1 h. After that, the CeO_2 precursor was centrifuged at a speed of 6000 rpm for 5 min and washed with ethanol three times. Finally, CeO_2 particulates were obtained by calcining the precursor at 500 $^\circ\text{C}$ for 2 h.

2.3 Preparation of sodium dodecylbenzene sulfonate modified CNFs

To enhance the dispersion of pristine CNFs in solution, sodium dodecylbenzene sulfonate (SDBS) was used to modify carbon nanofibers, denoted as SDBS-CNFs. Briefly, 0.3 g CNFs and 0.3 g SDBS were

added into 100 mL deionized water, adequately agitated and dispersed with ultrasonic, and then centrifuged at 6000 rpm for 5 min. After that, the centrifuged results were washed with deionized water several times and dried overnight in vacuum to obtain the SDBS-CNFs.

2.4 Preparation of antistatic superhydrophobic composite coating

Before spraying, the steel sheets were polished with 800 mesh sandpaper to remove the surface oxide film and then washed with acetone and ethanol for 20 min.

At room temperature, 0.02 g SDBS-CNFs and 0.07 g CeO_2 particulates were ultrasonically dispersed and stirred for 2 h in 25 mL absolute ethyl alcohol. Afterwards, 0.4 g PPS, 0.3 g PTFE and 0.1 g PDMS were successively added into the above homogeneous solution, ultrasonically dispersed and adequately agitated to form a well-dispersed suspension. Subsequently, the prepared dispersed suspension was sprayed onto the substrate surfaces using a N_2 gas spray gun. Finally, the antistatic superhydrophobic composite coating (ASC-coating) was obtained by heating the sprayed samples at 330 $^\circ\text{C}$ for 2 h. The schematic diagram of the preparation process of the ASC-coating is shown in Fig. 1.

2.5 Anti-icing tests

The anti-icing property was measured by the icing time of droplets on the samples. Deionized water (35 μL) was dripped on the surfaces of the specimens and placed into a refrigerator (-10 $^\circ\text{C}$). The relative humidity of the air was approximately 45%, and a digital camera was used

Fig. 1 Schematic diagram of the preparation process of the antistatic superhydrophobic composite coating.

to observe the changes in the droplet surfaces every 30 s.

2.6 Thermal stability tests

The ASC-coating samples were placed into a muffle furnace (KSL-1100X) and heated for 1 h at 100 °C, 150 °C, 200 °C, 250 °C, 300 °C, 350 °C, 400 °C and 450 °C. After that, the stability was evaluated by measuring the WCAs and SAs of each specimen.

2.7 Antistatic tests

A resistivity meter (MCP-T610) with a four-point probe was used to measure the electrical conductivity of samples with different SDBS-CNFs contents. The average values of surface resistance were obtained by measuring five different regions of the sample.

2.8 Corrosion resistance tests

The corrosion resistance of the specimens was measured in 3.5% sodium chloride (NaCl) aqueous solution using a three-electrode electrochemical workstation (CHI660D). The surface area of the working electrode sample is 10 mm × 10 mm. The platinum and saturated calomel electrodes were used as the auxiliary electrode and reference electrode, respectively.

2.9 Characterization

The surface morphologies of the particles and coated specimens sputtered with Au before testing were observed by Field Emission Scanning Electron Microscopy (FESEM, JSM-6701F, measured current of 10 μA, accelerating voltage of 5 kV). A Fourier Transform Infrared spectrometer (FTIR, Nexus 870) was employed to analyze the chemical compositions in the measuring range of 4000 cm⁻¹ – 400 cm⁻¹. X-ray diffraction (XRD, D8Discover25), with a measuring range of 2 θ from 5° to 90°, was used to identify the crystal structures of the samples. The WCAs and SAs were measured with 5 μL and 8 μL deionized water droplets, respectively, on a CA system (JC2000D1CA meter). The average values of WCAs and SAs were obtained by measuring five different regions of the sample. The element distribution and chemical composition were analyzed by energy-dispersive X-ray spectroscopy (EDS, JSM-5600LV) and X-ray

Photoelectron Spectroscopy (XPS, ESCALAB 250Xi), respectively. Field Emission Transmission Electron Microscopy (FETEM, FEI Tecnai G2 TF20 S-TWIN) was performed to examine the morphology and structure of the particles. A thermogravimetric analyzer (Netzsch STA 449F3) was employed to measure the thermal stability of the coating under a nitrogen atmosphere. The temperature rose from 20 °C to 800 °C.

3 Results and discussion

3.1 Microstructure characterization of the ASC-coating

It is well known that the surface microstructure of coatings is extremely vital for hydrophobic property^[46]. Fig. 2a displays the micromorphology of the as-obtained CeO₂ particulates, with diameters ranging from 2 μm to 3 μm, presenting a 3D flowerlike structure, which can provide rough structural support for the ASC-coating. The corresponding XRD pattern is shown in Fig. S1, which indicates the as-prepared CeO₂ particulates possess good crystal and cubic fluorite structures (JCPDS Card No. 34-0394). The variations in pristine CNFs before and after modification with SDBS are reflected by FTIR spectroscopy in Fig. S2. By comparison, there are no significant differences between the two curves, indicating that SDBS has no obvious effect on the structure of CNFs. Nevertheless, compared with the CNFs, a new characteristic peak at 1462 cm⁻¹ corresponds to the C = C stretching vibration in the benzene ring of SDBS and the absorption peaks at 1130 cm⁻¹ and 675 cm⁻¹ are assigned to the S = O stretching vibration of the benzenesulfonic acid group in SDBS^[47]. The results suggest that the surface of CNFs is successfully coated with SDBS, which is propitious to the uniform distribution of carbon fibers in the ASC-coating.

The SEM image of the CeO₂ particulates mixed with SDBS-CNFs (CeO₂-SDBS-CNFs) is exhibited in Fig. 2b. It can be clearly seen that CeO₂ particles and modified CNFs are evenly distributed in the corresponding magnified image (Fig. 2c). By contrast, in Fig. 2d, pristine CNFs agglomerate with CeO₂ particles. The above results show that SDBS can improve the dispersion of CNFs in solution, which is extremely vital to improving the hydrophobicity of the coating. Fig. 2e exhibits the micromorphology of the ASC-coating. From

Fig. 2 SEM images of (a) 3D flowerlike CeO_2 particulates, (b, c) CeO_2 -SDBS-CNFs, (d) CeO_2 -CNFs, (e, f) ASC-coating.

the magnified image (Fig. 2f), it is clearly visible that SDBS-CNFs, CeO_2 particulates and other polymers are evenly crosslinked to form the ASC-coating morphology, which generates a stable micro-nano rough structure for the superhydrophobic coating.

As seen from the Selective Area Electron Diffraction (SAED) pattern in Fig. 3a, the CeO_2 particulate exhibits a high polycrystalline structure and all crystal planes match well with the XRD pattern. The clear lattice fringes can be seen from the representative High-Resolution Transmission Electron Microscopy (HRTEM) image (Fig. 3b), which also illustrates the polycrystalline characteristics of the as-prepared particles. Moreover, the lattice fringe spacing is approximately 0.313 nm (Fig. 3b), which is aligned with those of the (111) planes of ceria particulates in the XRD pattern. The TEM analysis (Fig. 3c) illustrates that CNFs exhibit hollow architectures and 3D flowerlike structures are composed of petals. Furthermore, the magnified TEM image of SDBS-CNFs (Fig. 3d) indicates the SDBS adsorbs well on CNFs, which is consistent with the results in Fig. S2.

From the FTIR spectra of the ASC-coating (Fig. 4a), the peaks at 1468 cm^{-1} and 1008 cm^{-1} are assigned to the benzene ring of PPS. The bands at 1089 cm^{-1} and 478 cm^{-1} are attributed to the C-S bond and S-S stretching vibration in PPS. The absorption bands observed at

1076 cm^{-1} , 643 cm^{-1} and 553 cm^{-1} are associated with the C-C bond, $-\text{CF}_2^-$ wagging vibration and $-\text{CF}_2^-$ scissoring vibration for PTFE, respectively. The $-\text{CF}_2^-$ groups in PTFE provide the essential low surface energy for the ASC-coating. The bands at 1260 cm^{-1} and 796 cm^{-1} correspond to the asymmetric bonds of Si-O-C and Si-C in PDMS, reducing the surface energy of the ASC-coating. In addition, the strong peaks at 1636 cm^{-1} should be ascribed to the symmetric stretching vibration of R-SO₂-R and the stretching vibration of S = O^[39], indicating a thermal oxidation crosslinking reaction occurs in PPS. It can further improve the adhesion strength between the Q345 steel sheet and the ASC-coating and play an important role in the formation of the superhydrophobic coating. However, the FTIR characteristic peaks of CeO_2 are not observed, which might be explained by the fact that the peak strength is too weak and covered by the other characteristic peaks. According to the XRD pattern of the PPS powder in Fig. 4b, four diffraction peaks are found at 18.8° , 20.6° , 25.6° and 27.4° , which are indexed to the (110), (111/200), (112) and (211) planes of PPS, respectively. After annealing, only the (111/200) plane of PPS is observed in the XRD pattern of the ASC-coating, indicating PPS undergoes a thermal oxidation crosslinking reaction^[48]. In addition, the diffraction peaks at 18.0° and 28.4° are associated with

Fig. 3 (a) SAED pattern and (b) HRTEM image of the as-obtained CeO_2 , TEM images of (c) CeO_2 -SDBS-CNFs and (d) SDBS-CNFs.

(100) of PTFE and (111) of CeO_2 , respectively. As presented in Fig. 4c, the F1s at 688.5 eV and S2p at 164.1 eV indicate that PTFE and PPS are dispersed in the superhydrophobic coating. Furthermore, from the EDS spectrum in Fig. 4d, it can be easily seen that the ASC-coating is composed of C, O, F, Si, Ce and S. The F, Si, Ce and S peaks are attributed to the existence of PTFE, PDMS, F, Si, CeO_2 and PPS in the ASC-coating, respectively.

3.2 Wettability

The hydrophobicity of the coated and uncoated specimens is shown in Figs. 5a–5c. In Fig. 5c, it can be clearly seen that the coated surface (black part) displays excellent water-repellent property. It has a high WCA (166° , Fig. 5b) and low SA (2° , Fig. 5b). However, compared with the coated surface, the uncoated surface (grey part) in Fig. 5c shows hydrophilicity (65° , Fig. 5a), which indicates the hydrophobic property of the sample surface is obviously improved by the ASC-coating. In addition, the dynamic variations in the water droplet

adhesion on the ASC-coating surface are displayed in Fig. S3, which suggests the adhesion force, between the droplet and coating, is very small.

In practical applications, water droplets on the surface of objects tend to be acidic or alkaline^[49]. Therefore, it is significant to investigate the influence of different pH values on the surface wettability of samples. The variations in the WCAs and SAs on the surface of specimens at different pH values ranging from 1 to 14 are exhibited in Fig. 5d. It can be clearly observed from the figure that the WCAs vary steadily with changes in pH and that the ASC-coating possesses high WCAs (156° – 166°). With the changes in pH, the SAs fluctuate obviously, but all of them are still less than 10° (2° – 5.2°). The above analyses indicate that the ASC-coating still displays excellent superhydrophobicity for water droplets with different pH values.

3.3 Self-cleaning and anti-fouling properties

Metal components are easily contaminated in the working environment, which seriously reduces their

Fig. 4 (a) FTIR spectra of the ASC-coating, (b) XRD pattern of PPS powder and ASC-coating, (c) XPS spectra and (d) EDS spectrum of ASC-coating.

Fig. 5 (a) Contact angle of Q345, (b) contact and sliding angles of the ASC-coating, (c) photograph of the Q345 (grey part) and ASC-coating (black part), (d) contact and sliding angles of the ASC-coating under different pH conditions.

service life^[50,51]. The performances of self-cleaning and anti-fouling can prevent the sample surface from being contaminated in applications. The self-cleaning property of the ASC-coating was investigated using soil from the

park. As shown in Figs. 6a and 6b, both coated and uncoated steel sheets are evenly covered with soil. Then the water was slowly dripped onto the contaminated sample surfaces. In Fig. 6a, when water flowed through

the surface of the untreated steel sheet, a large amount of soil remained on the surface of the sample. In contrast, on the surface of the ASC-coating, water droplets easily rolled off the coating surface and removed the soil from the surface of the sample. As a result, in Fig. 6b, no soil particles were left on the surface of the coated sample, illustrating the excellent self-cleaning performance of the ASC-coating.

The anti-fouling performance of the ASC-coating was evaluated by a muddy water mixed solution, which was made up of water and soil mixed in a certain proportion. After being dripped on the surface of the untreated sample, the muddy water droplets flowed slowly through the surface. Afterwards, a large amount of wet soil adhered to the surface, as shown in Fig. 6c. However, in Fig. 5d, on the ASC-coating surface, the muddy water droplets displayed spherical shape and rapidly rolled down from it. No contaminated particles were left on the surface, which indicated the ASC-coating has outstanding anti-fouling property.

3.4 Anti-icing property

The freezing time of water droplets is an important index to evaluate the anti-icing property of the sample surface^[52]. The superhydrophobic surface possesses an excellent ability to delay ice formation^[9]. The variations in water droplets with condensation time are shown in Fig. 7. Water droplets were dripped on the coated (in Fig. 7a) and uncoated (in Fig. 7b) surfaces, respectively. With an increase in condensation time, the wetting state of droplets will change. In Fig. 7b, the water droplet on the uncoated surface began to condense into ice at 183 s, while the water droplet on the ASC-coating still showed transparency without ice crystal formation. As time increased, water droplets froze completely at 210 s. Nevertheless, for the coated surface, ice crystals did not appear until 802 s and white opaque ice droplet appeared at 856 s (Fig. 7a). The main reasons for the excellent anti-icing performance of the coating surface can be summarized as follows. On the one hand, a small amount of air existed between the droplet and the superhydrophobic surface, which prevented heat transfer and decreased the cooling rate of the water droplets, thereby prolonging the freezing time. On the other hand, the water droplet had a larger contact angle and smaller

Fig. 6 Self-cleaning property of the Q345 (a) and ASC-coating (b); anti-fouling property of the Q345 (c) and ASC-coating (d).

Fig. 7 Variations in droplet icing on the surfaces of (a) coated and (b) uncoated samples at different times.

contact area with the superhydrophobic surface, affecting the speed of heat transfer and slowing down droplet icing^[54].

3.5 Thermal stability

The thermal stability of superhydrophobic coatings, restricting their further application, is of great significance to their practical application^[55]. Furthermore, temperature is a significant factor affecting the superhydrophobic properties of coatings^[56]. The thermogravimetric (TG) and derivative thermogravimetric (DTG) curves of the ASC-coating are displayed in Fig. 8a, and the corresponding thermal parameters are listed in Table S1. For the coating, during thermal decomposition, only one thermogravimetric platform exists in the TG curve. Correspondingly, only one obvious characteristic peak is observed in the DTG curve. As seen from the table, the initial decomposition temperatures of the superhydrophobic coating at 5%

($T_d5\%$) and 10% ($T_d10\%$) weight loss are 496.6 and 519.8 °C, respectively. Moreover, the maximum weight loss rate appears at 553.9 °C (T_{max}) in the DTG curve. Evidently, the above results indicate that the ASC-coating possesses excellent thermal stability which can be explained by the fact that the heat accumulated in the ASC-coating can dissipate quickly due to the excellent thermal conductivity of CNFs^[57]. In addition, for the ASC-coating, no significant weight loss is observed below 250 °C. This is mainly attributed to the outstanding stability of the ASC-coating compositions at 250 °C.

Fig. 8b shows the superhydrophobic variations in the ASC-coating surface with heating temperature. With increasing heating temperature, the WCAs decrease and SAs increase, but the ASC-coating surface still displays superhydrophobicity below 350 °C with high WCAs (151° – 162°) and low SAs (3° – 8°), illustrating the great superhydrophobic thermal stability of the coating surface. However, the coating, with 141° WCA and 15° SA, loses its superhydrophobicity at 400 °C because of the thermal decomposition of PTFE and PDMS. When the heating temperature is further increased to 500 °C, the WCA of the ASC-coating surface is 0°, which is ascribed to the thermal decomposition of the coating at 496 °C.

3.6 Antistatic analysis

Surface Resistivity (SR) can be applied to measure the antistatic properties of coatings^[58]. Table S2 shows

the surface resistivity and logarithmic values of the superhydrophobic coating with different carbon contents (g and wt%). With the increase in SDBS-CNFs content (g), the variations in the logarithmic values and WCAs of the superhydrophobic coating are displayed in Fig. 9a. From Fig. 9b, it is found that there are not enough carbon fibers to form continuous conductive channels in the coating, which results in the SR ($3.96 \times 10^8 \Omega$) being very high at 0.005 g SDBS-CNFs. With the SDBS-CNFs content increasing to 0.010 g, the surface resistivity decreases sharply to $2.82 \times 10^5 \Omega$, achieving an antistatic standard, which is ascribed to the formation of continuous conductive paths (as shown in Fig. 9c). This result indicates the antistatic property of the ASC-coating increases with increasing SDBS-CNFs content. As the SDBS-CNFs content continuously increases, the SR of the ASC-coating varies slowly, which indicates the coating has an electrical percolation effect. Furthermore, the electrical percolation threshold (Φ) is approximately 1.12 wt%, corresponding to an SDBS-CNFs content of 0.010 g. Once the SDBS-CNFs content exceeds Φ , the antistatic property of the coating is greatly improved. As the SDBS-CNFs content increases from 0.015 g to 0.030 g, the corresponding SR, falling from $3.76 \times 10^4 \Omega$ to $3.47 \times 10^3 \Omega$, decreases increasingly slowly. Moreover, with further SDBS-CNFs addition, no obvious variations in the SR are found in the curve. Correspondingly, in Fig. 9d, the SDBS-CNFs distribute on the coating surface to form continuous conductive channels, which is not

Fig. 8 (a) TG and DTG curves of the ASC-coating, (b) variations in the contact and sliding angles of the ASC-coating at different temperatures.

Fig. 9 (a) Variations in the surface resistivity and contact angle of the ASC-coating at different CNFs contents; SEM images of the ASC-coating with (b) 0.005 g, (c) 0.01 g, (d) 0.03 g and (e) 0.04 g CNFs.

significantly different from the morphology shown in Fig. 9e. The above results can be explained by the growth process of the percolation group. The percolation ratio is the ratio of the content of SDBS-CNFs in the continuous conductive channels and total SDBS-CNFs^[59]. At the initial stage, a small addition in SDBS-CNFs content will significantly increase the percolation ratio, causing a rapid decrease in the SR. Furthermore, near Φ , the growth rate of the percolation group is extremely rapid. Nevertheless, when the SDBS-CNFs content exceeds the critical value Φ , the growth rate of the percolation group begins to decrease, and the corresponding SR of the ASC-coating decreases increasingly slowly with increasing SDBS-CNFs content. As the SDBS-CNFs content exceeds 0.030 g, the percolation ratio is close to 100%, resulting in the growth rate of the percolation group no longer varying significantly with increasing SDBS-CNFs content. As a result, the SR tends to be stable.

It can also be seen from Fig. 9a that the hydrophobicity of the coating is related to the SDBS-CNFs content. Compared with the SR, the WCA (146°) is less than 150° at low SDBS-CNFs contents (< 0.005 g), which is attributed to insufficient SDBS-CNFs to construct superhydrophobic rough structures. After that, as the SDBS-CNFs content increases, the hydrophobicity of the ASC-coating increases gradually due to the improvement of the surface hydrophobic rough structure. In particular, by increasing the SDBS-CNFs content to 0.020 g, the ASC-coating

possesses the best superhydrophobicity, with a WCA of 166° . The corresponding SR is $8.70 \times 10^3 \Omega$, illustrating the coating has outstanding antistatic property. However, with further SDBS-CNFs addition (> 0.020 g), the dispersion of SDBS-CNFs in the superhydrophobic coatings decreases, causing a decrease in hydrophobicity. As a result, the WCA of the coating decreases continuously. From the above analysis results, it can be seen that the optimum SDBS-CNFs content is 0.020 g, at which the ASC-coating possesses both optimal hydrophobicity and excellent antistatic property.

3.7 Corrosion resistance

Corrosion resistance is an important index to measure the service performance of iron and steel^[3,5]. Superhydrophobic coatings can improve the corrosion resistance of matrix samples. The Tafel plots of the uncoated and different coated samples in 3.5% NaCl aqueous solution are characterized in Fig. 10a. The corrosion resistance of the coating is evaluated through the corrosion potential (E_{corr}) and corrosion current (I_{corr}) obtained using Tafel extrapolation (as shown in Table S3). In general, higher E_{corr} and smaller I_{corr} indicate lower rates of thermodynamic corrosion and dynamic corrosion, respectively^[60]. Compared with the Tafel plot of the bare Q345 steel sheet ($E_{\text{corr}} = -0.603$ V, $I_{\text{corr}} = 3.166 \times 10^{-5}$ A), it can be clearly seen that the corrosion resistance performance was drastically improved on the ASC-coating surface as a result of the higher corrosion potential (-0.378 V) and lower corrosion current

Fig. 10 (a) Tafel plots of the bare and differently coated Q345 steel sheets in a 3.5% NaCl aqueous solution. The differently coated Q345 steel sheets include the samples of PTFE-free coating, CeO_2 -free coating, PDMS-free coating and ASC-coating. (b) Contact angle of the bare and differently coated Q345 steel sheets including the PTFE-free coating, CeO_2 -free coating, PDMS-free coating and ASC-coating.

(6.439×10^{-6} A). According to a previous study^[53], the improvement of corrosion resistance might be attributed to the existence of air bubbles in the micro-nano architectures of the coating. Owing to the low surface energy of the superhydrophobic coating, the corrosive solution molecules are prevented from penetrating into the micro-nano architectures of the coating, bringing about the better anticorrosion property.

However, in general, the barrier effect provided by coatings can also enhance the corrosion resistance of the matrix^[61]. Therefore, it is uncertain whether the good corrosion performance is determined by the shielding effect or the superhydrophobic ability. Based on the above indeterminacy, the anticorrosion properties of three other contrast coated samples, namely, the PTFE-free coating sample (PTFE-free), CeO_2 -free coating sample (CeO_2 -free) and PDMS-free coating sample (PDMS-free), were researched under the same conditions. As seen from the Tafel plots of three other coated samples, the corrosion potentials of PTFE-free, CeO_2 -free and PDMS-free samples are -0.576 V, -0.557 V and -0.546 V, respectively, and the corresponding corrosion currents are 2.581×10^{-5} A, 1.833×10^{-5} A and 1.502×10^{-5} A, illustrating the anticorrosion property of the substrate is enhanced by the barrier effect of the three coatings. Nevertheless, the anticorrosion performance of the ASC-coating is obviously better than that of the other three coatings,

which is closely related to the hydrophobic properties of several coatings. As presented in Fig. 10b, the WCAs (increasing from 129° to 166°) of the four coatings increase in the order of PTFE-free, CeO_2 -free, PDMS-free, and ASC-coating, which is consistent with the variation tendency of their anticorrosive properties. It can be explained that the contact area between the corrosive solution and coating surface with poor hydrophobicity increases, and the corrosive liquid molecules can easily penetrate into the micro-nano architectures of the coating surface. Specifically, for the superhydrophobic coating, the anticorrosion ability of the surface increases dramatically due to the air protection shield on the superhydrophobic surface^[62]. Consequently, it can be proven that the improvement of corrosion resistance on the ASC-coating surface depends mainly on superhydrophobic behavior rather than on the coating barrier.

4 Conclusion

In summary, a facile and modifier-free approach was applied to fabricate a PPS-based antistatic superhydrophobic coating. The as-obtained functional coating exhibits remarkable superhydrophobicity, a high water contact angle (166°) and a low sliding angle (2°). It has been shown that the coating, with outstanding water repellency stability under different pH conditions, possesses excellent self-cleaning, anti-fouling and

anti-icing properties. Due to the excellent thermal conductivity of CNFs, the coating still maintains its superhydrophobicity after calcination at 350 °C for 1 h. In addition, the surface resistivity and superhydrophobicity of the prepared coating are related to the SDBS-CNFs content. When the SDBS-CNFs content is 0.020 g, the antistatic superhydrophobic coating possesses both optimal hydrophobicity and excellent antistatic property. Furthermore, the superhydrophobic coating displays excellent corrosion resistance in a 3.5% NaCl aqueous solution. This approach with a facile and inexpensive preparation process may make possible true industrial applications. In addition to the common application areas, this kind of functional coating can be used in some special fields such as coal mining, aviation, defense and petrochemical applications.

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