

An all-water-based system for robust superhydrophobic surfaces

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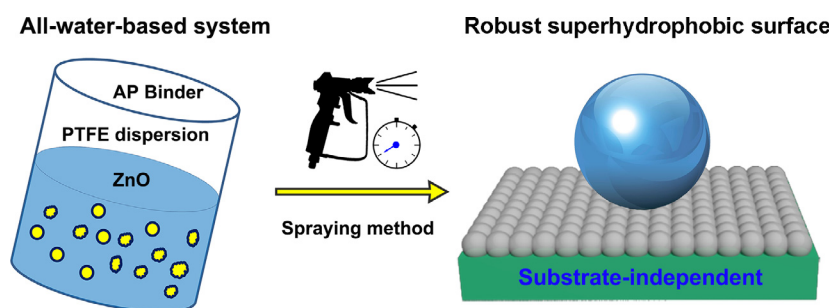
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GRAPHICAL ABSTRACT

An all-water-based system is presented to construct robust superhydrophobic surfaces. The substrate-independent coatings are durable against various harsh conditions and physical damages, and show promise for anti-icing and oil-water separation.



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ABSTRACT

Superhydrophobic surfaces with micro-/nanohierarchical structures are mechanically weak. Generally, organic solvents are used to dissolve or disperse organic adhesives and modifiers to enhance the mechanical strength of superhydrophobic surfaces. In this work, an all-water-based spraying solution is developed for the preparation of robust superhydrophobic surfaces, which contains ZnO nanoparticles, aluminum phosphate as an inorganic adhesive, and polytetrafluoroethylene with low surface energy. The all-water-based system is appreciated for low price and less pollution. Importantly, the prepared superhydrophobic surfaces are durable enough against various harsh conditions (such as UV irradiation for 12 h, pH values from 1 to 13, and temperatures from -10 to 300 °C for 12 h) and physical damages (including sandpaper abrasion and sand impact tests for 50 cycles). In addition, the obtained interfacial materials show promise for practical applications such as anti-icing and oil-water separation.

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1. Introduction

Lotus leaf-inspired superhydrophobic surfaces with extreme water repellency have drawn significant attention and research efforts [1–4]. They show superior application performance in self-cleaning, anti-icing, oil-water separation, anticorrosion, catalysis, and drag reduction [5–15]. According to already-established

wetting theory, the superhydrophobic surfaces require highly rough morphology and low-surface-energy chemical compositions [16–20]. Nevertheless, their widespread use in real-life application is seriously limited by poor mechanical robustness, because the required micro-/nanohierarchical surface structures are very vulnerable to physical damage. To solve the problem, Lu and co-workers employed organic adhesives to strengthen the mechanical durability of artificial superhydrophobic surfaces. Ethanol solution containing perfluorosilane and titanium dioxide nanoparticles was sprayed on the organic adhesive-modified substrates [21]. After that, superhydrophobic surfaces with enhanced mechanical strength have been widely designed and constructed with the help of organic adhesives [22–31]. For example, Xiong et al. prepared superhydrophobic precursor by mixing hydrophobic nanoparticles, UHMWPE as an organic adhesive, and decahydronaphthalene as a solvent [22]. Wang et al. dispersed perfluorosilane and perfluorosilane-modified silica nanoparticles in acetone and painted them on PS to fabricate robust superhydrophobic surfaces [30].

The dispersibility of the spraying solution is of importance for the formation of uniform and dense coatings. As such, the aforementioned wet-chemical coating systems for robust superhydrophobic surfaces broadly use organic solvents, which favor the dissolution or dispersion of organic modifiers and adhesives. The heavy use of organic solvents not only increases the preparation cost, but also causes safety hazard and poses a significant environmental risk, which limits the large-scale production and extensive applications. Therefore, it is highly desirable but challenging to develop an all-water-based coating system for the preparation of robust superhydrophobic surfaces [32,33].

There are two important criteria in the exploitation of the all-water-based superhydrophobic coating systems. First, a stable spraying aqueous solution should be prepared. Second, the binder force between the superhydrophobic coatings and substrates must be strong enough after water evaporation. In this work, commercial polytetrafluoroethylene (PTFE) aqueous solution was used as a low-surface-energy component. Water-soluble aluminum phosphate (AP) as an inorganic adhesive can enhance the mechanical strength of the superhydrophobic coatings. In addition, ZnO nanoparticles were employed to increase the surface roughness. The proposed all-water-based coating system was available for various substrates, such as glass, ceramic, fabric, nickel foam, and stainless steel mesh (SSM), forming robust superhydrophobic surfaces. These surfaces withstood various harsh conditions including high-intensity UV irradiation, acidic and alkaline solutions, and remained extreme water repellency after sandpaper abrasion and sand impact tests. Furthermore, the prepared superhydrophobic materials performed multiple functions including self-cleaning, anti-icing, and oil-water separation.

2. Experimental section

2.1. Materials

Glass, ceramic, fabric, nickel foam, and SSM were commercially available. All the chemical reagents were analytical grade and used without further purification.

2.2. Fabrication of superhydrophobic coatings on various substrates

AP binder was prepared by mixing $\text{Al}(\text{OH})_3$ with orthophosphoric acid (H_3PO_4 , 60%) in a molar ratio of 1:3 under stirring at 100 °C for 3 h. Afterward, ZnO nanoparticles (0.8 g, about 100-nm size) were dispersed into 15 mL deionized water containing AP binder (2.4 g). After stirring for 10 min, 5 mL PTFE aqueous solution was

dropwise added into the above-mentioned AP-ZnO aqueous solution under stirring for another 10 min. Meanwhile, all the substrates were ultrasonically rinsed with deionized water and anhydrous ethanol for several times. Subsequently, the AP-ZnO@PTFE aqueous solution was uniformly sprayed onto the cleaned substrates with a spray gun under 0.2 MPa N_2 gas. In order to realize the cross-linking and curing of the AP-ZnO@PTFE coating, the samples were continuously heat-treated at 120 °C for 2 h and then 240 °C for 1 h.

2.3. Mechanical stability tests

Two methods were utilized to evaluate the mechanical strength of the AP-ZnO@PTFE coating, such as sandpaper abrasion and sand impact tests. For sandpaper abrasion test, taking AP-ZnO@PTFE coated fabric as an example, the AP-ZnO@PTFE coating was faced down to the sandpaper (Grit No. 1000) and dragged to move for 10 cm along a ruler under a 200 g weight by external drawing force. After the sandpaper abrasion for ten times, water contact angles (CAs) and sliding angles (SAs) were measured. For sand impact test, 50 g sands in the funnel fall down from 50 cm height to impact the AP-ZnO@PTFE coated ceramic.

2.4. Oil-water separation

(I) Oil adsorption. The boat made by the AP-ZnO@PTFE coated nickel foam can move directionally under external magnetic force and adsorb light oils floating on the water. Similarly, the superhydrophobic nickel foam can adsorb heavy oils at the bottom of the beaker. (II) Oil filtration. The AP-ZnO@PTFE coated fabric was fixed between two glass tubes and placed vertically. Three different types of immiscible oil-water mixtures (40 mL, 50% v/v) were poured onto the surface. The oils include hexane, dichloroethane, and chloroform. The flux was calculated by measuring the time after collecting 20 mL of oils. The separation efficiency of a variety of oils was evaluated by the oil permeation coefficient ($R(\%)$) according to the following equation: $R(\%) = m_{\text{separation}}/m_{\text{initial}}$, where $m_{\text{separation}}$ represents the weight of oil after each separation and m_{initial} denotes the weight of oil in the initial oil-water mixtures.

2.5. Characterization

All optical photographs were taken by a digital camera (Sony, DSC-HX200). The chemical composition and crystal structure of samples were characterized by X-ray photoelectron spectroscopy (XPS, Thermo Scientific ESCALAB 250Xi), Fourier transform infrared spectra (FTIR, Thermo Scientific Nicolet iS10), and X-ray diffraction (XRD, X'PERT PRO), respectively. The surface morphology was observed on a field emission scanning electron microscope (SEM, JEOL JSM-6701F) with Au-sputtered specimens. The accelerating voltage and current were 5 kV and 10 μA , respectively. The element distribution was investigated by energy dispersive spectroscopy (EDS, KeveX). The three-dimensional roughness profile was imaged using an atomic force microscope (AFM, Agilent 5500) in the contact tapping mode. CAs were got on a JC20001 contact angle system (Zhongchen digital equipment Co., Ltd. Shanghai, China). The average CA value was obtained by measuring the sample at five different positions. The water adhesion force was measured using a high-sensitivity micro-electromechanical balance system (DCAT11, DataPhysics Deutschland) in air. First, a water droplet (10 μL) was suspended with a metal ring which was hung on the microbalance. The substrate which was placed on the balance table was moved upward at a constant speed of 0.005 mm s^{-1} until contacted with the water droplet. Then the substrate was moved down. Adhesion forces were obtained from the

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