Preparation of Superhydrophobic Polypropylene / SiO_2 Coating for Self-Cleaning Application

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Abstract

This study introduces a straightforward and cost-efficient dip-coating technique for the fabrication of hierarchical superhydrophobic nanocomposite coatings. Hydrophobic silica nanoparticles (NPs) were synthesized via the sol–gel method and subsequently integrated into a polypropylene matrix. The degree of superhydrophobicity was adjusted by varying the concentration of silica NPs within the polymer solution. Upon optimization, the coating achieved a water contact angle (WCA) of 165° and a sliding angle of 7°. Surface morphological analysis revealed a hierarchical rough structure on the coated glass substrate, which facilitated air entrapment and contributed to the observed water-repellent behavior. The coating demonstrated excellent wetting stability under water jet impact and exhibited notable self-cleaning characteristics. Mechanical durability was assessed through sandpaper abrasion and adhesive tape peeling tests, confirming the coating's robustness. These highly non-wettable and self-cleaning surfaces hold promise for a range of practical applications.

Keywords: Superhydrophobic, Self-cleaning, Nanocomposite, Coatings, Glass

Introduction

In recent years, the development of superhydrophobic surfaces has garnered significant attention from both scientific and industrial communities due to their remarkable water-repellent **[1]**, self-cleaning **[2]**, anti-fouling **[3]**, and anti-corrosive **[4]** properties. A surface is deemed superhydrophobic when its static WCA exceeds 150°, and the sliding angle (SA) is less than 10°, allowing water droplets to bead and roll off easily, thereby removing contaminants. This self-cleaning effect is inspired by natural surfaces such as lotus leaves and butterfly wings, which exhibit hierarchical surface roughness combined with low surface energy materials, resulting in minimal solid–liquid contact **[5]**. The application of this principle in engineered coatings has led to advancements in fields such as construction materials, textiles, solar panels, marine applications, and biomedical devices.

Among various polymeric materials, polypropylene (PP) has emerged as a promising matrix for superhydrophobic coatings due to its inherent hydrophobicity, excellent chemical resistance, low density, and thermal stability **[6]**. However, pristine polypropylene lacks the surface roughness and surface energy manipulation required to achieve true superhydrophobicity. Therefore, modifying polypropylene by incorporating nanostructured fillers such as silicon dioxide (SiO_2) and optimizing surface morphology becomes essential[6]. SiO_2 nanoparticles are particularly advantageous for this purpose due to their high surface area, tunable size, chemical stability, and ability to form micro/nano hierarchical textures on polymer surfaces. Several strategies have been reported for fabricating superhydrophobic polymer/inorganic composite coatings, including sol–gel synthesis **[7]**, spray coating **[8]**, dip coating **[9]**, and phase separation methods **[10]**. These approaches generally involve creating multi-scale roughness and reducing surface energy by employing fluorinated or silane-based functional groups. However, challenges persist in ensuring uniform dispersion of nanoparticles, maintaining mechanical durability, and achieving scalable, environmentally friendly fabrication processes. Moreover, the integration of low-cost polymers like polypropylene with functional inorganic components such as SiO_2 opens new avenues for industrially viable, high-performance self-cleaning surfaces **[11]**. The synergistic combination of polypropylene and SiO_2 nanoparticles can provide a dual-functional system: the hydrophobic polypropylene serves as the base matrix to enhance chemical stability and weather resistance, while the SiO_2 nanoparticles introduce hierarchical surface roughness essential for water droplet mobility.

By tailoring the dispersion of SiO_2 in the polymer matrix and optimizing the coating process, it is possible to fabricate robust, superhydrophobic surfaces suitable for real-world applications. This study aims to develop a superhydrophobic PP/SiO_2 composite coating via a facile spray-coating method for self-cleaning applications. The research focuses on optimizing the concentration of SiO_2 nanoparticles and evaluating their effect on surface wettability, morphology, and durability. Comprehensive characterization of the prepared coatings is performed using contact angle measurements. Ochir et al. **[12]** successfully engineered a durable superhydrophobic surface by applying a coating of hydrophobic silica nanoparticles (SNPs) onto polypropylene (PP) through a straightforward compression molding technique. The resultant coating exhibited a high WCA of approximately 170° and a low sliding angle of less than 5°, attributed to its nano/microscale roughness and low surface energy. The coating demonstrated remarkable mechanical durability, maintaining its superhydrophobic properties after enduring 25 tape peel cycles and 100 cm sandpaper scratch tests. Additionally, it exhibited good chemical stability in acidic and neutral environments, although it was less stable under alkaline conditions. This method presents a scalable and cost-effective strategy for fabricating robust, self-cleaning superhydrophobic surfaces, making it suitable for practical applications. Moradi et al. **[11]**developed a superhydrophobic and anti-corrosion polypropylene (PP) coating for mild steel surfaces by incorporating polypropylene grafted with maleic anhydride (PP-g-MAH) and graphene oxide (GO) into the PP matrix. These additives significantly enhanced the coating's compactness, corrosion resistance, and hydrophobicity. The optimal formulation, consisting of 1.5 g PP-g-MAH and 2 mg GO, achieved a contact angle of 166°, a surface roughness of 87 nm, and corrosion resistance 800 times greater than that of pure PP. Molecular dynamics (MD) simulations further confirmed an increase in coating density from 0.80 to 0.85 g/cm^3 due to the presence of GO. After immersion in NaCl solution for 2 hours, the coating with 2 mg GO maintained superior corrosion resistance, demonstrating its effectiveness in protecting construction metals such as mild steel.

In this study, hydrophobic silica NPs were synthesized using the sol–gel method with methyltrimethoxysilane (MTMS) serving as the precursor. A suspension containing the synthesized silica NPs and polypropylene was applied to glass substrates through a controlled dip-coating process, which involved regulated dipping and withdrawal speeds. The resultant coating demonstrated excellent self-cleaning properties, as evidenced by a high WCA of 165°, allowing water droplets to roll off the surface effortlessly.

Experimental section

Materials

Sigma-Aldrich (USA) provided Methyltrimethoxysilane (MTMS, 98%) and Polypropylene Changshu Hongsheng Fine Chemical Co., Ltd. (China) supplied Ethanol (99.9%). Ammonium hydroxide (MW 35.05) has been provided by Research Lab Pvt. Ltd. (Mumbai, India). Micro-glass slides (75 × 25 × 1.35 mm) were obtained from Blue star, Polar Industrial Corporation, India.

Synthesis of Hydrophobic Silica Nanoparticles

Hydrophobic silica nanoparticles were synthesized using the sol–gel method, employing methyltrimethoxysilane (MTMS) as the silica precursor. In a typical synthesis, 4 mL of MTMS was combined with 40 mL of methanol and 8 mL of deionized water. The resulting solution was stirred with a magnetic stirrer for 20 minutes at room temperature to initiate the hydrolysis process. Subsequently, ammonium hydroxide solution, serving as a catalyst, was added dropwise to the mixture under continuous stirring. The reaction was allowed to proceed for an additional 30 minutes to facilitate condensation and gelation. The resulting gel was aged at room temperature overnight to enhance the structural integrity of the silica network. Following aging, the gel was dried in an oven at 80 °C for 5 hours to remove residual solvents and moisture. The dried xerogel was then mechanically ground using a mortar and pestle to obtain fine, nanoscale silica particles suitable for further use.

Preparation of Polypropylene/Silica Nanocomposite Coating

To prepare the superhydrophobic coating, 10 mg of polypropylene (PP) granules were dissolved in 10 mL of o-xylene under magnetic stirring for 30 minutes to ensure complete dissolution of the polymer. Subsequently, 150 mg of the previously synthesized hydrophobic silica nanoparticles were added to the polymer solution, and the mixture was subjected to ultrasonication in an ultrasonic cleaner for 20 minutes to promote uniform nanoparticle dispersion. Glass substrates were cleaned and then dip-coated in the prepared PP/SiO_2 nanocomposite solution. Each substrate was immersed for 6 seconds per cycle, and the dip-coating process was repeated for a total of 10 cycles to achieve adequate coating thickness and surface uniformity. The coated substrates were subsequently dried in a hot air oven at 120 °C for 3–4 hours to ensure solvent evaporation and proper film formation.

Characterizations

WCA and WSA were determined using a contact angle goniometer (HO-IAD CAM-01, Holmarc Opto-Mechatronics Pvt. Ltd., India). To ascertain the roll-off angle, the coated glass substrate was affixed to the instrument’s sample holder, which was incrementally inclined using an adjustment screw until the droplet commenced rolling off. For the water jet impact test, a continuous stream of water was applied to the coating via a 15 mL syringe. Furthermore, water droplets were released from a height of approximately 10 cm onto the coating surface, inclined at an angle of 30°, to evaluate dynamic wettability. The mechanical durability of the coating was assessed through adhesive tape peeling, sandpaper abrasion, and sand gran impact tests. Self-cleaning performance was evaluated by dispersing artificial dust particles onto the coated surface, followed by observing the removal behavior under water flow.

Results and discussion

The synthesis of hydrophobic silica nanoparticles (SNPs) via the sol–gel method using methyltrimethoxysilane (MTMS) as the organosilane precursor is a well-established approach for producing functionalized nanomaterials with tunable surface properties. MTMS, a trifunctional silane, provides both the silica source and hydrophobic methyl groups, which are crucial for tailoring the surface energy of the resulting nanoparticles. The initial mixing of MTMS, methanol, and deionized water, followed by stirring at room temperature, facilitates the hydrolysis of methoxy groups on MTMS to form silanol groups (–Si–OH). This reaction is essential as it governs the extent and uniformity of network formation. Methanol acts as a solvent and homogenizing agent, while water serves as the reactant for hydrolysis. The dropwise addition of ammonium hydroxide introduces a basic environment that catalyzes the condensation of silanol groups, leading to the formation of a three-dimensional silica network through siloxane (Si–O–Si) bonds. The controlled addition and continuous stirring help in regulating nucleation and growth processes, resulting in uniform nanoparticle formation. The subsequent aging step, performed overnight at ambient conditions, is critical for network rearrangement and strengthening of the gel, contributing to improved porosity and mechanical integrity of the silica matrix.

Drying the gel at 80 °C removes volatile components, transitioning the gel into a xerogel. This step must be carefully controlled to avoid pore collapse, ensuring retention of the nanoscale porosity. Mechanical grinding of the dried xerogel breaks down the bulk structure into fine particles, yielding nanoscale hydrophobic silica with a large specific surface area and suitable surface chemistry for functional applications.

Wettability and self-cleaning ability

The wettability of a solid surface is largely determined by its chemical composition and surface morphology. In this study, coating performance was evaluated by varying the number of dipping cycles—specifically 8, 9, and 10 cycles—resulting in samples labeled P1, P2, and P3, respectively. The P1 sample exhibited a WCA of 145° with a low sliding angle (SA) of 9°, indicating hydrophobic behavior. The P2 sample demonstrated improved performance, with a WCA of 151° and a reduced SA of 7°, suggesting enhanced water repellency.

As shown in Figure 1a, the S3 sample displayed nearly spherical water droplets, corresponding to a high WCA of approximately 165° (Figure 1b). The slight difference of only 3° between the left and right contact angles indicates excellent surface uniformity. Water droplets rolled off the surface with ease, supported by the low SA of 7°, confirming the strong non-wetting, superhydrophobic character of the P3 coating. This superior performance is attributed to the rough and porous surface morphology of the coating. This confirms that the silica nanoparticles–polypropylene nanocomposite was uniformly deposited, forming a consistently rough and porous structure across the substrate.

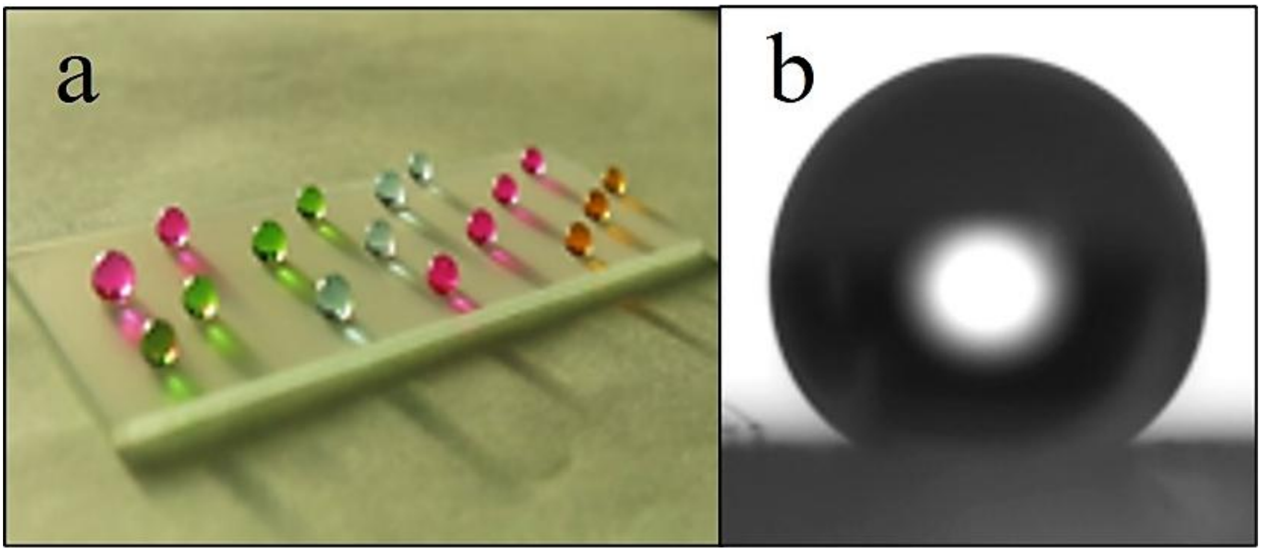

Figure 1. (a) The optical photographs of the colored water droplets on the P3 sample. **(b)** WCA image on the P3 sample.

The images shown demonstrate the self-cleaning behavior of a superhydrophobic coating applied to a glass substrate. In image (a), the surface is intentionally soiled with fine dust or soil particles. In image (b), a stream of water is gently poured onto the coated surface. As observed, the water droplet rolls down the inclined surface, carrying the soil particles away with it. This clearly indicates that the surface exhibits excellent self-cleaning properties, where the low surface energy and micro/nano-structured roughness prevent the adhesion of contaminants. Instead, the dirt is picked up and removed by rolling water droplets, confirming the effectiveness of the superhydrophobic coating in maintaining a clean surface with minimal effort.

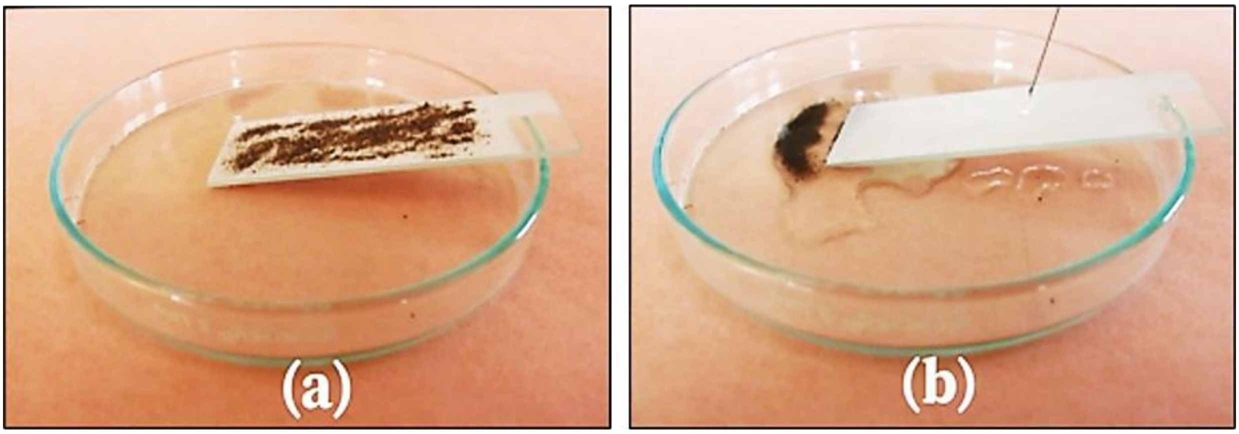

Figure 2. **(a)** The optical photographs of the soil particles spread on the P3 sample, **(b)** soil particles washed away easily by the rolling water droplets.

Mechanical durability of the coating

The mechanical durability of the superhydrophobic coating was inspected by adhesive tape, sandpaper abrasion test, and sand grain impact. The adhesive and mechanical durability of the superhydrophobic P3 coating was evaluated through adhesive tape peeling and sandpaper abrasion tests. In the tape peeling test, an adhesive tape was gently pressed onto the coated surface and slowly peeled off to assess the coating's adhesion strength. The coating retained its superhydrophobic nature for up to one peeling cycle. However, after the second cycle, the WCA dropped to 139°, indicating partial removal of the nanocomposite layer. With subsequent cycles, the WCA continued to decline, reflecting poor adhesion, and by the sixth cycle, the coating became hydrophilic with a WCA of 56° (Figure 3a).

The mechanical robustness was further assessed using the sandpaper abrasion test. Here, the P3-coated surface was placed face-down on 400-grit sandpaper under a 20 g load and moved across a 10 cm distance per cycle. The coating maintained superhydrophobicity up to two abrasion cycles. Beyond this point, the WCA declined sharply, signifying the progressive erosion of the nanocomposite layer caused by mechanical abrasion. After 50 abrasion cycles, the coating was completely removed, leaving a hydrophilic surface with a WCA of 68°, as illustrated in Figure 3b. These results highlight the coating’s limited mechanical durability and adhesion strength under repeated physical stress.

To evaluate the mechanical durability of the coating under simulated sandstorm conditions, a series of sand impact tests were conducted on the P3 marble sample. In each cycle, 10 g of sand was dropped from a height of 20 cm above the sample, totaling 30 g over three cycles. As depicted in Figure 3c, the WCA progressively decreased with increasing impact cycles. Following three sand impact cycles, the coating exhibited a significant reduction in WCA to approximately 46°, indicating moderate resistance to mechanical wear under simulated sandstorm conditions.

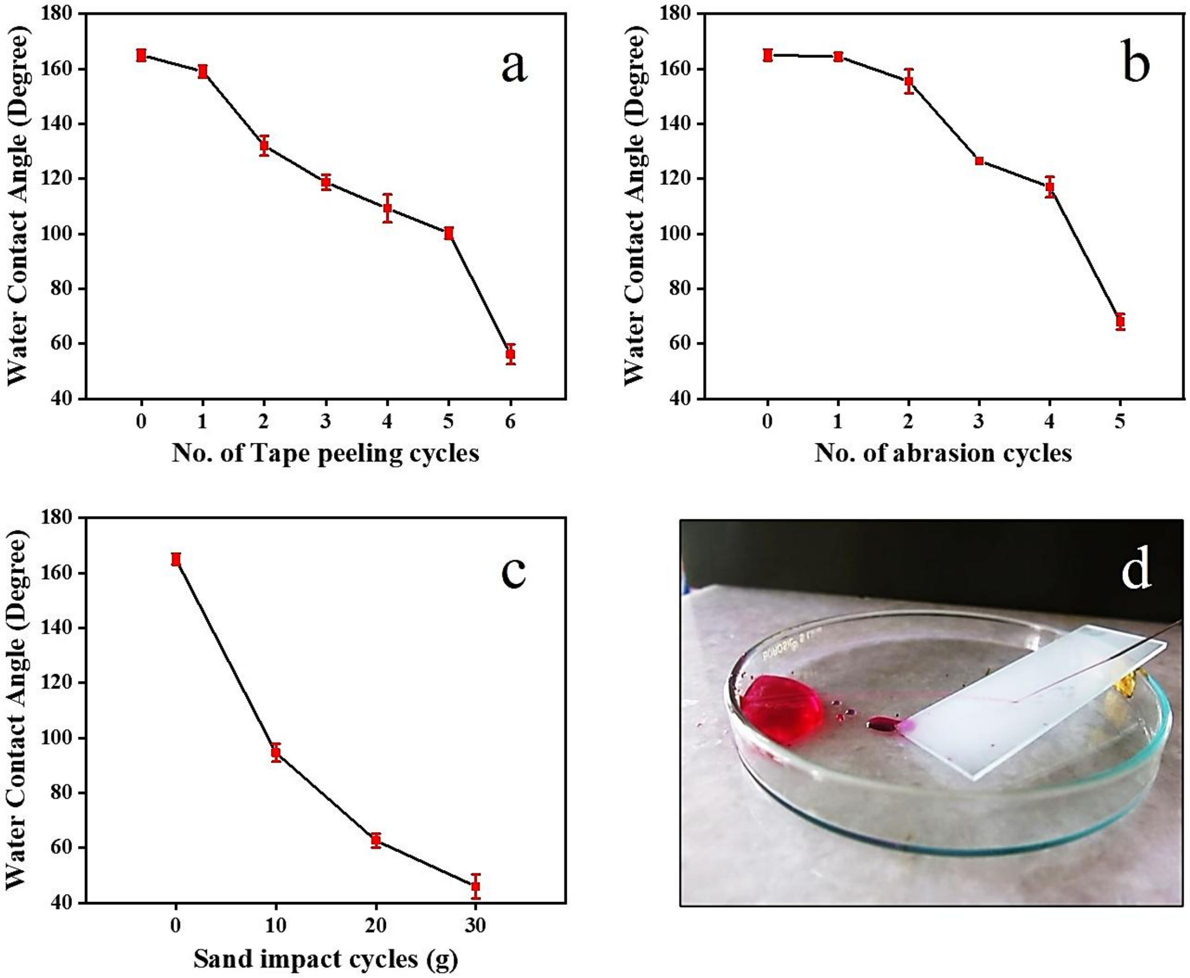

Figure 3. (a) Variation in WCA after each adhesive tape cycle, **(b)** variation in WCA after each sandpaper abrasion cycle, **(c)** variation in WCA after 10 g of sand gran impacting, **(d)** optical photograph of P3 sample repelling water jet.

A water jet generated from a syringe was directed onto the coated glass substrate. Upon striking the SHP P3 sample, the water jet bounced off the surface, indicating its strong water-repellent nature, as illustrated in Figure 3(d). This behavior can be attributed to the hierarchical surface structure, which traps air pockets and prevents water penetration into the coating. The water jet was continuously applied to a fixed point on the surface for over 2 minutes, and the persistent rebounding of the jet confirmed the excellent mechanical robustness and durability of the P3 coating.

Conclusions

In conclusion, a superhydrophobic coating was effectively developed on glass substrates by applying a layer of polypropylene and silica nanoparticles using a dip-coating method. The resultant coated surface demonstrated exceptional water-repellent characteristics, with a WCA of 165° and a water sliding angle (WSA) of 7°. In terms of durability, the coating exhibited significant mechanical resilience, maintaining its superhydrophobic properties after undergoing one cycle each of adhesive tape peeling and sandpaper abrasion, as well as after exposure to 10 g of sand impact and prolonged water jet testing.

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