Supplementary Information

Eco-Friendly Self-Clean Coatings: Fundamentals, Fabrication, Applications, and Sustainability

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S1. Different Approaches for Achieving Self-Cleaning Properties

Several approaches are employed to achieve self-cleaning properties in coatings. A few of them have been discussed in this section.

S1.1 Photocatalytic Self-Cleaning

Photocatalytic self-cleaning is an approach that makes use of photocatalytic materials, typically titanium dioxide (TiO_2). When these materials are exposed to ultraviolet (UV) light, they generate reactive oxygen species. These reactive species have the ability to break down organic contaminants that may be present on the surface, effectively rendering it self-cleaning [1]. This method is particularly practical for outdoor applications where exposure to sunlight or UV sources is readily available. The chemical reactions initiated by the photocatalytic materials help maintain a clean surface by disintegrating organic matter, making it a sustainable and low-maintenance option. Anderson et al. [2] demonstrated that when TiO_2 is paired with SiO_2, a synergistic enhancement occurs in the self-cleaning ability of the treated surface. The addition of SiO_2 not only increases the surface area around the TiO_2 but also elevates the surface acidity of the photo-catalyst. Furthermore, the presence of SiO_2 works as a protective role for the substrate and shielding the substrate during the photo-catalysis process. Pillai et al. [3] presented a method for fabricating photo-catalytic cotton surfaces using tin oxide (SnO_2) and TiO_2 nanoparticles mixture. The excessive use of TiO_2 coating can cause health concerns and researchers now focusing on the development of self-cleaning photocatalytic SnO_2 nanoparticle coating. The procedure involved coating the cotton fabric surface using an acrylic binder by pad-dry-cure technique. The resulting TiO_2-coated cotton surfaces exhibit greater self-cleaning activity than the SnO_2-coated cotton fabric and SnO_2 samples showed lower stain pick-up. The SnO_2-TiO_2 mix nanoparticles showed intermediate results between SnO_2 and TiO_2-coated cotton. This approach offers potential applications in various fields, including transparent films or optoelectronic devices and the elimination of pollutants from water and many other sources. Yi et al. [4] discussed a polymer-assisted approach of an aqueous chemical solution method for creating a photocatalytic anatase TiO_2 carbon nanotube thin film on a glass substrate for green intelligent building. The process involves the formation of a TiO_2 carbon nanotube thin film on the quartz surface by spin coating at 2500 rpm for 30s and then the film is calcinated at 400 °C for 2 h in air. The resultant TiO_2 carbon nanotube thin film showed nearly two-fold photocatalytic activity performance than pure TiO_2 because of effectively preventing electron-hole pair recombination and fast electron transport. In this line, Yang et al. [5] presented the fabrication of TiO_2 self-cleaning coating for glass substrate by hydrothermal reaction using different concentrations of tetrabutyl titanate, hydrochloric acid, and ethanol and then compounded with polyvinyl alcohol (PVA) and waterborne polyurethane as an adhesive. The mixed composite coating was applied on a glass substrate by a spray coating method. The resulting glass substrate exhibits transmittance of 86.95-88.89% and excellent self-cleaning performance and could degrade organic dyes efficiently and repeatedly. Fateh et al. [6] discussed the fabrication of transparent photocatalytic TiO_2–ZnO coating using the sol-gel technique. To ensure good adhesion of the SiO_2 interlayer on the polycarbonate sheet the researcher first modified the polycarbonate sheet with UV(C) light irradiation followed by coating with the dip and spin coating method. The best self-cleaning properties are exhibited at the molar ratio of 1:0.05 TiO_2 –ZnO with good mechanical and UV irradiation stability. Gao et al. [7] utilized a sono-chemical technique for the synthesis and deposition of TiO_2/SiO_2/GO nanocomposites onto polyester/cotton surfaces. The results of their work towards the development of TiO_2/SiO_2/GO nanocomposites achieved better self-cleaning performance in visible light as compared to pure TiO_2 alone. The addition of SiO_2/GO into TiO_2 nanocomposites gives a more uniform and smooth coating with little agglomeration, resulting in increased specific surface area and surface energy. Xu et al. [8] modified the cotton fabric with 3-chloro-2-hydroxypropyl trimethyl ammonium chloride (CHTAC) and 1,2,3,4-butanetetracarboxylic acid (BTCA) to increase the stability of TiO_2/SiO_2 coating on cotton fabric. The resulting modified cotton fabric showed the optimum molar ratio of TiO_2/SiO_2 sol of 50:1 for the degradation rate of methylene blue on various treated samples. Furthermore, the crystallinity and tensile strength of ionic cross-linked cotton decreased. The result also indicates that the 60 gpl of BTCA gives an adequate improvement increase recovery angle with no significant influence on the tensile strength of cotton. Wu et al. [9] fabricated a novel photocatalytic self-cleaning cotton fabric functionalized with Bismuth oxyiodide (p-BiOI)/n-TiO_2 heterojunction. The procedure involved a coating of TiO_2 film on cotton fabric by sol-gel method and loading of p-BiOI nanosheets on TiO_2 film by a chemical bath deposition method. This BiOI/TiO_2 coating exhibits improved photocatalytic activity against methyl orange, mainly due to BiOI possessing a narrow band gap that enables the utilization of visible light for excitation. The p-BiOI/n-TiO_2 heterojunction increases the transfer of photo-induced carriers while reducing the recombination of electron-hole pairs.

S1.2 Hydrophobic and Superhydrophobic Coatings

Hydrophobic and superhydrophobic coatings are designed to repel water, and they contribute significantly to self-cleaning surfaces [10,11]. These coatings work by preventing water from adhering to the surface and by minimizing the contact area between the surface and contaminants. As a result, when it rains or when other cleaning agents, such as water, come into contact with the surface, they effectively wash away dirt and other particles, leaving the surface clean and free from unwanted substances. These coatings are particularly useful for a variety of applications, from outdoor signage to household windows, due to their ability to maintain a clean appearance in wet conditions. Cheng et al. [12] utilized a method for fabricating a fully sustainable, fluorine-free, nanoparticle-free, and robust superhydrophobic cotton fabric surface through a combination of enzyme etching followed by coating with cured epoxidized soyabean oil (CESO) thermoset and low surface energy stearic acid modification. The CESO thermoset, stearic acid played a crucial role in achieving the superhydrophobic coating by forming a hydrophobic layer on the etched cotton surface. The procedure involved immersing the cotton fabric in a cellulase enzyme solution and then coated with CESO through dip-coating, followed by modification with stearic acid. The resulting surface exhibited excellent water repellence, with WCA reaching up to 157°. This approach is applied as an environmentally friendly substitute for non-renewable and harmful chemicals containing oil/water separation materials. In this line, Xu et al. [13] obtained a route for fabricating green and sustainable superhydrophobic cotton fabric surfaces through a combination of phytic acid (PA) etching followed by coating with CESO thermoset and low surface energy stearic acid modification. The procedure involved immersing the cotton fabric in a PA solution and then coated with CESO through immersion, followed by modification with stearic acid. The resulting surface exhibited excellent water repellence, with WCA reaching up to 156.3°. In addition to this, there were no changes in inherent properties of the cotton fabric like water vapor permeability, flexibility, and water absorption. Zhang et al. [14] fabricated a superhydrophobic surface from beeswax, lignin, and cotton with achieved WCA of up to 163°, making them highly water-repellent. The procedure involved the spraying of beeswax and lignin suspension directly on cotton fabric. This innovative strategy holds significant potential for renewable oil-water separation material from natural polymer. Ishizaki et al. [15] explored that corrosion-resistant superhydrophobic surfaces were formed on a magnesium alloy substrate by a facile one-step immersion process. Through a simple technique, they achieved a high WCA of over 150°. The resulting myristic acid-modified micro-/nanostructured surface coatings exhibited various large-scale industrial production of corrosion resistance Mg alloy having new applications like airplanes, trains, and automobiles. Wen et al. [16] presented a green and environmentally friendly method to fabricate superhydrophobic nano coating based on the use of recyclable eggshells, stearic acid, and ZnO. The fabrication process involved the use of eggshells and ZnO to obtain the micro- and nano-hierarchical structure, stearic acid to lower the surface energy, and carboxymethyl cellulose to enhance the interaction between substrate and coating. This energy-effective superhydrophobic nano coating performs WCA of 153° and a low sliding angle of 4° on a glass substrate. Furthermore, these superhydrophobic coatings tend to provide superior resistance to mechanical damage, UV resistance, and excellent anti-icing properties. Teli et al. [17] discussed a study of modified nano-silica coating for nylon knitted fabric. The silica nanoparticles coating on nylon fabric was modified by in situ deposition of ZnO and subsequent hydrophobic modification with sodium stearate. The resulting modified nylon fabric showed WCA of 151° and excellent ultraviolet protection with an ultraviolet protection factor (UPF) of 279.68. Forsman et al. [18] presented the hydrophobation of different cellulosic textiles using a thin, open coating based on carnauba wax particles. To optimize the hydrophobicity of the samples, the researchers experimented with various parameters, including curing temperature, cationic polymer, and fabric properties. The optimum curing temperature was found at 70 ℃ helped to obtain the highest WCA of 148.5°. The cationic starch was not more efficient compared to poly-L lysine, but still, good water repellency obtained with two bilayers and higher roughness fabric showed the highest WCA. Yu et al. [19] illustrate the approach to the use of ammonium polyphosphate@SiO_2-silicone oil to create flame retardant and superhydrophobic coating. Ammonium polyphosphate@SiO_2-silicone oil coating applied on cotton fabric showed WCA of 151.28°. The coated cotton fabric also exhibits good flame retardancy properties. This multifunctional cotton fabric provides a large number of commercial applications.

S1.3 Lotus-Effect Emulation

The concept of lotus-effect emulation draws inspiration from the unique properties of the lotus leaf. Some self-cleaning coatings incorporate micro- and nano-structured surfaces that mimic the lotus leaf's texture [20,21]. These micro- and nano-structures create a surface that minimizes the contact area between the surface and contaminants, making it challenging for dirt to adhere. When water droplets encounter the surface, they effectively pick up and carry away contaminants, ensuring the surface remains clean. This approach is particularly beneficial for applications where rainwater or other water sources are readily available to help with the cleaning process. Ebert et al. [22] fabricated the lotus effect with hierarchically structured surfaces using micro- and nanosized hydrophobic silica particles through a spray coating method. The resulting surface formed by using the micro- and nanoparticles exhibited WCA of 166° and CAH of 2° at the optimal pitch and the surface with micropatterns and nanoparticles exhibited nearly identical properties with WCA of 168° and CAH of 1°. In both cases, the surface met the requirements for superhydrophobicity (WCA>150°) and self-cleaning (with CAH<10°). Furthermore, coating surfaces demonstrated superior wear resistance compared to epoxy resin on the multiple-length scale in atomic force microscopy (AFM).

S1.4 Oleophobic Coatings

Oleophobic coatings are specifically designed to repel oils and grease. They are highly suitable for surfaces exposed to oily substances, such as kitchen countertops and electronic devices with touchscreens that may accumulate fingerprint oils [23]. Like their hydrophobic counterparts, oleophobic coatings work by preventing the adhesion of substances, making it easy to wipe away oils, grease, and other hydrophobic contaminants. These coatings are particularly practical for maintaining the cleanliness and appearance of surfaces where oily residues are common. Cao et al. [24] highlighted the use of waste corn straw as a natural ingredient to achieve underwater superoleophobic and under oil superhydrophobic waste corn straw powder (CSP) coated cotton fabric. The dully pre-wetted underwater superoleophobic waste CSP-coated fabric was fabricated by a spray coating method. The resulting coating exhibits stable underwater oleophobicity for all oils, including dichloroethane and chloroform with contact angles reaching greater than 150°, indicating strong oleophobicity. This strategy provides the environment-friendly low-cost, and simple approach for the application in the field of separation of light/water/heavy oil three-phase mixtures. Brown et al. [25] addressed the issue of the mechanical durability of superoleophobic coatings. To solve issue, a layer-by-layer approach was employed, enhancing flexibility, improving adhesion to the substrate, and providing a low surface tension coating at an air interface. The mechanically durable superoleophobic coating on glass substrate was developed through separate spray deposition of a polyelectrolyte polydiallyldimethylammonium chloride (PDDA), SiO_2 nanoparticle, and a fluorosurfactant. The PDDA was complexed with a layer of fluorosurfactant, which provides repellency to oil while being hydrophilic. The addition of SiO_2 nanoparticles was carried out to enhance oleophobic/superhydrophilic behavior, resulting in a superoleophobic coating with hexane contact angles exceeding 155° and a tilt angle of less than 4°.

S1.5 Antimicrobial Self-Cleaning

In certain applications, self-cleaning extends beyond removing physical contaminants to inhibiting microbial growth [26]. This is achieved by incorporating antimicrobial agents into coatings. These agents actively prevent the colonization of bacteria and other microorganisms on the surface. As a result, surfaces with antimicrobial self-cleaning coatings are especially valuable in healthcare sectors, food preparation areas, and other environments where maintaining a sterile and clean surface is of utmost importance [27]. These coatings contribute to the overall hygiene and safety of the environment in which they are applied. Bukit et al. [28] described the self-cleaning and antibacterial activity of fabrics coated with oil palm boiler ash (OPBA) nanocomposite, TiO_2, and chitosan showed promising results. The fabrication process involved a simple dip-coating method, followed by a UV curing process, and dried in the sun-light. The OPBA nanocomposite, TiO_2, and chitosan played a crucial role in achieving the antimicrobial self-cleaning coating. The resulting surface exhibited antimicrobial activity against Staphylococcus aureus and E. coli. The introduction of TiO_2 and SiO_2, particularly silica sourced from OPBA along with the incorporation of chitosan improves self-cleaning performance and hydroxyl groups play a crucial role in causing damage to bacterial cells. El-Bisi et al. [29] discussed beeswax/chitosan, nano chitosan for antimicrobial self-cleaning coating of cotton. The simple pad-dry-cure technique was employed to create nanostructures of inherently antibacterial beeswax/chitosan, nanochitosan on cotton. The resulting surface provides 154° and 151° of WCAs for cotton fabric treated with chitosan, nano chitosan/beeswax, and silica emulsions. This was achieved when applying 2% chitosan, 0.2% nano chitosan/beeswax, and 2% silica emulsions. Seth et al. [30] employed a simple solution technique to create superhydrophobic and superoleophobic coating with excellent antibacterial and photocatalytic properties on cotton fabric using in-situ generated zirconium zinc stearate. The optimized coated cotton fabric exhibits WCA of 163° even after undergoing multiple cycles of machine laundering and mechanical abrasion. The hierarchical morphology of zinc stearate enhances the antimicrobial and self-cleaning properties, while the presence of zirconium contributes to increased laundering durability and mechanical robustness. This coated fabric holds promise for practical applications such as biomedical clothing and military uniforms. Furthermore, it proves effective in separating oil/water mixture with an efficiency of around 99% and this suggests its potential application in treating oil-contaminated marine water and industrial wastewater. Shaban et al. [31] fabricated superhydrophobic and antimicrobial coated cotton fabric by using ZnO nanocatalyst. The procedure involved the preparation of ZnO by sol-gel method and then loading on cotton fabric by a spin coating method. The optimized parameters like ZnO precursor concentration, precursor solution pH, number of coating runs, and Mg doping percentage were addressed. The superhydrophobic coated cotton fiber achieved the WCA of 154° with the optimized condition of ZnO precursor concentration at 0.5M, pH value at 7, and the number of spin coating runs to 20. The optimized ZnO-coated cotton demonstrated excellent abrasion resistance and environmental durability under outdoor conditions and UV illumination. The antibacterial activity of fiber functionalized with ZnO was assessed against various bacteria, with notable efficiency against K. pneumonia. The technology presented offers advantages such as low fabrication cost, improved hydrophobicity, and antibacterial properties.

Table S1: Different fabrication methods of self-clean coatings reported in the literature.

Fabrication method	Observation
Sol-gel [32]	The process involves the transformation of a colloidal solution (sol) to an integrated network (gel) and subsequent solidification.
Etching [33]	Selectively remove the material from the substrate surface by using acids or enzymes to get the desired substrate surface
Electrospinning [34]	The extrusion of polymer solution through the fine hole and then solidification into ultrafine fibers under the influence of an electric field
Chemical vapor deposition [35]	The substrate is exposed to volatile precursors, leading to a chemical reaction in the vapor phase, and solid material gets deposited on the substrate surface
Electrochemical deposition [36]	The use of electric current to drive the redox potential and resulting in the deposition of oxide, metal, or compound onto a surface
Hydro-thermal [37]	The growth of thin film at very high temperatures and high pressure enables precise control over the growth of thin film
Phase separation [38]	Controlled extraction of different components within a solution during deposition to achieve pattern structure on a substrate

Templating [39]	It involves the use of the negative template for the deposition of thin film onto a substrate
Lithography [40]	Thin film coating involves precise pattering of thin film on the substrate for microfabrication applications
Imprinting [41]	It employs a replication process to create patterns on substrates for microfabrication purposes
Layer-by-layer deposition [42]	Deposition of alternating layers to construct thin film coating
Self-assembly [43]	It utilizes the spontaneous organization of the components to create order structure on the surfaces.

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