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Self-Cleaning Technology in Fabric: A Review

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Abstract. This article gives an overview on photocatalytic self-cleaning technology on fabric resulting from titanium dioxide (TiO_2) and zinc oxide (ZnO) as photocatalyst which decompose the organic stain into water and carbon dioxide (CO_2) in presence of UV light source. The self-cleaning concept is useful in various application including the textiles materials which are normally used in daily life. This technology also can be developed in other application for instance medical textiles, athletic wear, and military uniform and also outdoor fabrics. Additionally, it is beneficial as it effectively conserves water and improves the appearance of the environment and in long term it will reduce energy, laundry cost and time as well.

1.0 Introduction

Self-cleaning concept has attained remarkable interest because of their distinctive features and wide range of possible applications in various fields. As well as increasing the demand for sanitary, self-disinfecting and contamination free surfaces, interest in self-cleaning protective materials and surfaces has developed rapidly throughout the years since it has high potential as commercial product, which is able to meet the market demands globally. There are numerous materials that utilized the self-cleaning technology including interior applications such as fabrics, furnishing materials, window glasses, and outdoor construction materials such as roof tiles, car mirrors, and solar panels [1].

The self-cleaning theory was instigated from nature phenomenon which can be noticed on leaves of lotus plant, rice plant, butterfly wings, fish scales, etc. [2-3]. For lotus leaf, it is a type of plant which grows in mud without let the mud affect the purity of the plant. The waxy surfaces of the lotus leaves combined with the presence of microscopic structures result in an extremely hydrophobic surface [4]. In 1997, the investigation of the self-cleaning ability of the plant leaves surface was done by Barthlott and Neihuis [2]. The surface characteristic was analysed by a high-resolution scanning electron

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microscope (SEM) and the contact angle (CA) of leaves from 340 plant species. Majority of the investigated leaves have contact angle less than 110° without any prominent surface sculpturing especially epicuticular wax crystals. Contrary to water-repellent leaves, there are various different surface sculptured, mainly epicuticular wax crystals were exhibited in combination with papillose epidermal cells and their contact angle always exceeded 150°. They observed that on water-repellent surfaces, water contracted to form spherical droplets and came off the leaf very quickly, even at slight angles of inclination ($<5^{\circ}$), without leaving any residue.

Generally, all kinds of particles are always removed entirely from water-repellent leaves when exposed to natural or artificial rain, as long as the surface waxes are not destroyed. The dirt particles that deposited on the waxy surface of the leaves are generally larger than the microstructure of the surface of the leaf. Hence, deposited on the tips, which results in the minimizing of the interfacial area between both surfaces. In the case of a water droplet rolling over a particle, the surface area of the droplet exposed to air is reduced and energy through adsorption is gained. The particle is removed from the droplet's surface only if a stronger force overcomes the adhesion between particle and water droplets. Due to the very small interfacial area between particle and rough surface, adhesion is minimized. Therefore, the particle is 'captured' by the water droplet and removed from the surface. This phenomenon is called "Lotus effect".

There are two principal ways of self-cleaning materials, namely hydrophobicity and hydrophilicity. Both types of coating clean themselves with the action of water by rolling droplets for hydrophobic and sheeting water for hydrophilic that carries dirt away. Nevertheless, hydrophilic have an additional property, which can chemically break down the adsorbed dirt in sunlight through the help of photocatalyst which also known as hydrophilic photocatalytic coating [5].

2.0 Hydrophobic and Hydrophilic

Hydrophobic surface repels water with the properties of low wettability and contact angles more than 90°. The higher contact angle the lower the value of adhesion on the surface and increase the hydrophobicity. For contact angle more than 150° the surface is termed as superhydrophobic. Hydrophobicity also can be regulated by the roughness factor. With rougher surfaces, the contact angle of water on it will increase, and form bumps that trap air between water and the surface. The "Lotus Effect" was applied in this superhydrophobic mechanism [2]. The lotus plant cleans itself by having superhydrophobic leaf which consists of microscopic bumps all across the leaf's surface that play the important part to its water-repelling properties. A rough coating of nanoscopic wax crystals on these bumps further increases the effect. It allows the water droplets rolls across and removes dirt away. The water and the dirt have more affinity each other than the surface [2]. Hang Ji et al. [6] have created a superhydrophobic polymer structure by directly replicating the surface of a lotus leaf. Poly (dimethysiloxane) (PDMS) was used to replicate the lotus leaf structure. The leaf was used as a template to cast a complementary PDMS layer. An anti-stick layer was added to the PDMS, which was then used as negative template for a second PDMS casting step. The second PDMS layer was then a positive image of the lotus leaf. The complex lotus surface patterns are transferred with high fidelity. The artificial PDMS lotus leaf has the same water contact angles and very low water roll-off angle as the natural lotus.

Hydrophobic surfaces can be useful for other application such as anti-corrosive systems and anti-icing [7]. The superhydrophobic coating by using nanoparticle-polymer composite for anti-icing can prevent the ice formation by inhibiting the frost nucleation process. It was observed that the composites are able to prevent ice formation depends not only on their superhydrophobicity but also on the size particles exposed on the surface upon impact of supercooled water both in laboratory conditions and in naturally occurring environments. These results open up possibilities for rational design of anti-icing superhydrophobic surfaces by tuning surface textures in multiple length scales [7]. In addition, superhydrophobic coatings can enhance the fuel efficiency in maritime industry by reducing the skin friction drags occurring in ship hulls. Such a coating increases the ship speed and also acts as anti-corrosive systems, preventing any organic contaminant or marine microorganisms to come in contact with the ship hulls. In vehicles, superhydrophobic coatings are applied on the glasses to prevent rain droplets from clinging, thereby helping to clean the car itself.

Meanwhile, besides hydrophobic, researchers also seek more interest in hydrophilic surface which is advantageous in presence of water for self-cleaning application. The wettability is high and the contact angle is approximately 0°. Therefore, the water lies flat on the surface in sheet instead of droplets. It cleans the surface alongside the photocatalysis process, followed by sheeting of water which makes the surface superhydrophilic thus carry the dirt molecules away [5].

3.0 Photocatalytic Process

Photocatalytic process is the acceleration of photoreaction in the presence of catalyst. This process will decompose the dirt molecules by utilize the sunlight. By utilizing the photoreaction induced by photocatalyst, the organic contaminants will be degraded into air and water [8]. The mechanism of photocatalytic reaction begins when a photocatalyst is irradiated by light, usually ultraviolet light (UV). With the energy equal to or higher than its band gap, electrons on the photocatalyst surface are excited, and escaped from valence band to the conduction band, leading to the formation of electron-hole pairs in the surface-excited negative charged electrons (e^-) in the conduction band, and the positive charged holes (H^+) in the valence band [9-21]. The created pairs can recombine or get trapped and react with other material that absorbed on the photocatalyst. The pairs will cause redox reactions at the surface, the negative electrons (e^-) and oxygen will combine to form super oxide radical anions (O_2^-), whereas the positive electric holes and water will generate hydroxyl radicals (OH^-) [8]. Eventually, all the formed highly active oxygen species will oxidize organics compound to carbon dioxide (CO_2) and water (H_2O). Hence, photocatalyst can decompose common organic matters in the air such as odour molecules, bacteria and viruses [14].

4.0 Photocatalyst

The semiconductor photocatalytic process has shown a great potential as a low-cost, environmental friendly and sustainable treatment technology. Those semiconductor materials also have been used to functionalize onto the different textiles for self-cleaning properties. The functionalized fabrics have the ability to oxidize the coloring substances in the form of solutions and stains [22].

4.1 Titanium Dioxide (TiO_2)

Titanium dioxide (TiO_2) is a semiconductor material that acts as photocatalyst and has been proved to be an excellent catalyst in the photo degradation of colorants and other organic pollutants. It is widely used because of its various advantages, such as, non-toxicity, availability, cost effectiveness, chemical stability and favourable physical and chemical properties. Titanium dioxide exists in three crystalline phases; rutile, anatase and brookite. Among these three forms, rutile which is used in pigments as sun-blockers and paints is more sustainable than other two forms. The anatase has an open crystal structure, which makes it highly photocatalytic and always been used in semiconductor photochemistry, as this appears to be the most active and easiest to produce. Anatase and brookite, on the influence of heat, would change to rutile [1]. TiO_2 is used in paint and cosmetics as pigment and as a food-additive. It is also used in anti-pollution applications and for water purification. Currently, TiO_2 is used for self-cleaning surfaces and has now emerged into commercial products ranging from kitchen and bathroom ceramic tiles and fabrics, to indoor air filter and window glass section [1]. Pisitsak et al. [23] investigated the self-cleaning properties of cotton fabrics finished with nano-TiO_2 and nano-TiO_2 mixed with fumed silica. The self-cleaning effect was stronger for samples coated with higher TiO_2 concentrations. Mostly, the finished samples appeared clean after one wash whereas the untreated fabrics required repeated washing. However, with the addition of fumed silica, a reduction in self-cleaning ability was observed. Aside from photocatalytic ability, H. Yaghoubi et al. proved that polycarbonate substrate with a self-cleaning TiO_2 layer can improve its mechanical durability by exhibits better hardness and scratch resistance [24]. These results are good mechanical properties as the self-cleaning coatings are vital to make these coatings attractive in auto and construction industries.

4.2 Zinc Oxide (ZnO)

Another semiconductor material, which appears to be an alternative to TiO_2 is zinc oxide (ZnO) [11,15]. The application of ZnO as a degradation material for environment pollutants has also been extensively studied, because of its nontoxic nature, low cost and high photochemical reactivity [17]. ZnO semiconductor exhibits a better efficiency of photocatalytic reaction than anatase TiO_2 in photochemical degradation of reactive dyes [13]. ZnO in powder form has been extensively used for photocatalytic degradation of dyes present in effluent [9,13,16]. However, the ZnO powder has a recovery problem after degradation, because some of ZnO powder is lost while draining the solutions. To overcome the problem, ZnO particles can be attached to a substrate particularly textiles because they offer the highest surface area. These ZnO functionalized textiles can be used for effluent treatment as well as self-cleaning. A few attempts have also been made to functionalize textiles with ZnO to generate self-cleaning properties. Ashraf et al. [15] studied the photocatalytic solution discoloration and self-cleaning of functionalized polyester fabric with ZnO nanorods. The nanorods of ZnO were deposited on polyester fabric by hydrothermal method and degraded the stains and decolorize the solution of different types of dyes [15].

5.0 Self-Cleaning Fabric

Textile products play an important role in our basic need. They are commonly produced and used in clothing industry. Textiles or fabrics are also important in other industries such as agricultural industry, transportation, building material and also health industry. Therefore, the technological advances of textiles are improved in order to meet the need of various industries. The properties and performance of textiles fibres are essential to fabric manufacturing and utilization [25]. Textile finishing has always leaded to introduction of new technical properties, which is useful in diversified end uses. Technological diversification in finishing decides the performance domain of fabrics and rendered it special functional properties. There has been a continuous improvement and innovation in the area of chemistry related to textile finishing. Research interest in the use of nanotechnology in the textile industry has increased rapidly in recent times. It was demonstrated that nanotechnology can be used to enhance textile attributes, such as fabric softness, durability, breathability, water repellence, fire retardancy and anti-microbial property in fibers, yarns and fabrics [25]. Textile finishing involving the application of inorganic colloidal nanosuspension (nanosols) can give rise to new fabrics in which properties of inorganic nanoparticles are transferred to the textile's surface [26]. Ever since the introduction of nanoparticles in textiles, efforts were made to produce finished fabrics with multiple performances. This is mainly due to the fact that textiles fabrics constitute one of the best substrates for the application of nanotechnology because those consisting of cotton fibre which is normally known possess large surface area [27]. The tight binding of nanoparticles to the textile surfaces is a significant part in application of nanotechnology in the textile industry. This is to ensure the durability of desired properties is increased and also minimum released of nanoparticles into the environment. In order to apply the nanoparticles, the maximum chemical compatibility between nanoparticles and textile surface should be achieved. There is commonly used method for this purpose which is a method that uses covalent linking agents.

The dip-pad-dry-cure process is often used to create bonds between nanoparticles and a fabric. Chaudari et al. [28] examined the effect of nano TiO_2 pre-treatment on functional properties of cotton fabric. The cotton fabric was treated with nano TiO_2 colloid by pad-dry-cure method. The TiO_2 concentrations in the padding liquor were varied and the specimens were immersed in a fresh colloidal bath for 10 minutes and squeezed using a laboratory padder at constant pressure. The samples were dried for 30 minutes and cured at 140°C for 3 min. Similarly, Ortelli et al [29] also applied the TiO_2 nanosol directly on textiles by pad-dry-cure method. The fabric was dipped in TiO_2 nanosol and left for 3 min, then passed through a two-roller laboratory padder, oven dried and cured for 10 min at 130°C. SiO_2 was used as a binder since it prevents textile from degradation during the photocatalytic activity of TiO_2. Pakdel et al. [20] functionalized wool fabrics using TiO_2 and TiO_2/SiO_2 nanocomposite through a low temperature sol-gel method. The prepared TiO_2/SiO_2 composite were applied to the surface of wool fabrics using dip-pad-dry-cure method. In order to make the TiO_2-SiO_2 layer, Yuranova et al. [28] deposited it on the textile by soaking cotton in a 1:1 solution of colloidal TiO_2 and SiO_2 Ludox SM-30. The sample was then dried in air at 100°C for an hour. The results of high-resolution transmission electron microscopy (HRTEM) and infrared spectroscopy show that the corrosion of the upper layers of cotton would occur due to the effect of daylight on the TiO_2 particles, if there were no SiO_2 as a binder.

Besides SiO_2, researchers also mostly used acrylic binder in the deposition of nanoparticle to textile surface. Sivakumar et al [29] immersed the cotton fabric for 1 min in aqueous solution with 0.5% acrylic binder in padding mangle. The nanoparticle was uniformly coated using laboratory padding mangle at speed of 15 m/min with pressure of 15 kg/cm^2. After padding, the fabric was dried at 70°C for 5 min and cured at 120°C in curing chamber for 3 min. The SEM image showed that the nanoparticles were well dispersed on the fiber surfaces and were finely dispersed and embedded. Moreover, Kumar et al. [8] also used acrylic binder (1%) and added 0.5 % Lissapol-N as capping agent. Through the SEM analysis, TiO_2 nanoparticles were well dispersed and embedded on fiber surface. Apart from that, previous reports indicated that carboxylic acid groups can acts as surface modifier to anchor TiO_2 nanoparticle into fabrics. Therefore, Wijesena et al. [30] investigated a method to modify the cotton fiber surface with carboxylic acid groups through slight carbomethylation of cellulose using sodium monochloroacetate (MCAA) and NaOH. Then, the fabric was grafted with TiO_2 nanoparticles through a process similar to dyeing. Once complete the grafting process, the fabric sample was sonicated to remove unattached particles and dried at 60°C. The catalyst loading was determined by measuring the weight gained after the TiO_2 nanoparticles grafting. The results showed the TiO_2 nanoparticles loading was increased with increase of MCAA concentration used to treat the cotton fabric.

The self-cleaning fabrics are function by implementing the photocatalytic reaction on the fabric with photocatalyst such as titanium dioxide and zinc oxide. As reported by Samal et al. [14] the fabric is coated with a thin layer of titanium dioxide particles then when this semiconductor exposed to light, photons with energy equal to or greater than the band gap of the photocatalyst excite electrons up to the conduction band. The excited electrons within the crystal structure react with oxygen atoms in the air, creating free-radical oxygen. These oxygen atoms are powerful oxidizing agents, which can break down most carbon-based compounds through oxidation-reduction reactions. In these reactions the organic compounds (i.e. dirt, pollutants, and microorganisms) are broken down into substance such as carbon dioxide and water. The TiO_2-SiO_2 coated cotton textiles showed a high photocatalytic activity

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superior to TiO_2 coated cotton alone due to the high dispersion and the structural effects of the amorphous silica present [28]. Meilert et al [31] reported that it is possible to bind TiO_2 to cotton textiles through chemical spacers. The coating procedure was straightforward and used non-toxic reagents. The TiO_2 loaded cotton textiles presented stable self-cleaning properties and allowed to eliminate partially the chromophore(s) of the red wine under daylight irradiation presenting long-term stable performance. In addition, Mirjalili et al [1] claimed that degree of self-cleaning of cured fabrics with cross-link method is higher than cured fabrics with non cross-link method. The reason for this higher self-cleaning is due to the higher percentage and more uniform distribution of titania particles over surface of cotton fabrics in cross-link method than in non cross link approach.

Bedford et al. [32] concluded that the photocatalytic self-cleaning textile fibres fabricated by coaxial electrospinning are compatible to nonelectrospun photocatalytic self-cleaning textile fibers that use better photocatalytic materials and sub-band gap wavelength light sources. This illustrates the important role played by the high surface to volume ratio of the material fabricated by coaxial electrospinning. Sivakumar et al. [29] assessed the ultra-violet (UV) protection and self-cleaning action of cotton fabric coated with nano ZnO and nano TiO_2 with acrylic binder. In the case of self-cleaning action, the smaller nanoparticle size *in-situ* coating of ZnO derived using wet chemical technique showed better self-cleaning activity as compared to larger nanoparticles of TiO_2 treated fabric. They also found that the ZnO and TiO_2 nanoparticles finishing of cotton fabric not only imparts the UV protection and self-cleaning characteristics to the treated fabric but also added two more functional properties like antimicrobial activity and soil release action. Chaudhari et al [28] also reported that the self-cleaning action of the pre-treatment of cotton fabric with nano TiO_2 is higher at the higher concentration level. In the meantime, Cakir et al. [10] also investigated the self-cleaning properties of textile coated with ZnO nanoparticles which were synthesized within PS-b-PAA reverse micelles cores and the result showed that the ZnO nanoparticles coated fabric at ratio 40:1 exhibited fairly high photocatalytic activity on the degradation of methylene blue. Meanwhile, self-cleaning function and hydrophilicity of wool fabrics were successfully improved through the integration of silica in the TiO_2/SiO_2 nano composite [20]. By increasing the concentration of silica, the TiO_2/SiO_2 nanocomposite showed more capability in decomposing the stains. Oda et al. [19] used cotton fabric as carrier and stabilizer of ZnO/Ag^+ to test for the photocatalytic activity by using methylene blue dye and artificial UV light. The result reveals that ZnO and ZnO-Ag which stabilized on cotton fibre were efficient in the decolorization of dye.

Later, Ashraf et al. [15] studied the self-cleaning and solution discoloration by the immobilized ZnO nanorods which grown on polyester (PET) fabric by hydrothermal method. They found that the functionalised fabric degraded the stains and decolorised the solution due to photocatalytic effect of ZnO. The fabric sustained the repetitive usage for solution discoloration but its stain degradability decreases after each degradation. The functionalized fabric has excellent washing fastness but fairly good rubbing durability. Oda et al. [18] continued their research involves synthesis of a composite of Ag/ZnO with cotton texture by photodeposition method. Silver doped zinc oxide showed a higher activity in dye removal in comparison with un-doped oxide. Additionally, the composite of Ag/ZnO/texture showed a higher activity with respect to Ag/ZnO. The composite Ag/ZnO/texture showed an ability towards self-cleaning of dye under irradiation. It was found that the activity of zinc oxide was increased in pH range that are close to its point zero charge (PZC). According to Kumar et al. [8] who studied the self-cleaning action of TiO_2 nanoparticles coated cotton, the coated textile structure demonstrates the significant self-cleaning activity when exposing under UV light spectrum. He also confirmed that the increased of percentage of TiO_2 nanoparticle caused increased in self-cleaning activity and the high duration of exposing under UV light also accelerated the self-cleaning action. These functionalized fabrics are not only gained self-cleaning properties but also improved in water repellence, soil resistance, wrinkle resistance, anti-bacteria, anti-static and UV protection [14].

4.0 Self-cleaning Testing

Photocatalytic self-cleaning fabrics are able to clean themselves when exposed to light, but they also possess antibacterial and UV blocking functions. Therefore, several testing methods can be used for the evaluation of the photocatalytic activity of functionalized fabric. Photocatalytic efficiency of functionalized fabrics is typically assessed by degradation of organic pollutants such as natural colorants stain or synthetic dye which commonly used as model pollutant. The photodegradation of the colorant was determined by two type of colorant decomposition activities such as solution discoloration and stain degradation. As for solution discoloration, pieces of the functionalized fabrics were placed dye solution and exposed under UV light. The dye solutions were collected periodically after certain time and its concentration is measured by using UV-Vis spectrophotometer [12, 15–16, 19, 29–30].

Meanwhile, for stain degradation, the functionalized fabric is stained with dye, and irradiated to UV light source for certain time. The stained sample is assessed for color strength (K/S) values on color spectrometer. The self-cleaning action can quantify by comparing K/S values of the exposed and unexposed portions of the same stain as shown below [8, 24–25, 27]:

$$\% \text{ Decrease in K/S value} = \frac{(K/S)\text{ unexposed} - (K/S)\text{ exposed}}{(K/S)\text{ unexposed}} \times 100$$

Where K is the absorption; and S, the scattering

The K/S represents the color strength on a surface and is directly proportional to the amount of dye present on it. The decrease in K/S values shows that the stains are disappearing [15]. Chaudhari et al. [27], Sivakumar et al. [24] and Kumar [8] evaluated the nano TiO_2 coated textile fabric by exposed the coffee stained samples to sunlight irradiation for 12-48 h and the percentage of decrease in K/S value was calculated. Ashraf et al. [15] stained the functionalized fabric samples with different types of dyes and exposed to UV light source. The K/S values as function of time were measured with color spectrometer.

Besides color strength value, the properties of self-cleaning fabric also can be determined based on highly oxidative intermediates generated at the stained cotton textiles surface, which can be measured by the gas chromatography. As reported by Meilert et al. [31], the released of carbon dioxide (CO_2) due to wine, coffee, make-up and perspiration stain were deposited on the treated textiles. These samples were irradiated for 24 h with Suntest solar light simulator with a light intensity of 50 mW/cm^2 and the release of CO_2 was followed by gas chromatography during 24 h. The higher amount of CO_2 measured, the better the self-cleaning properties.

The UV blocking function of textiles was usually evaluated by the determination of the ultraviolet protection factor values (UPF). The UPF is the ratio of UV radiation measured without the protection of the fabric compared to that with protection. UPF tests are conducted using spectrophotometer or a spectroradiometer and serve as a measure of the UV protection provided by clothing fabrics. A textile material must have a minimum UPF of 15 to be rated as UV protective [33]. Excellent UV-blocking ability of the ZnO-coated fabric, particularly in the region of the UVB (280–315 nm), was confirmed by Shateri-Khalilabad et al. [34]. Sivakumar et al. [29] assessed the ultra-violet (UV) protection of cotton fabric coated with nano ZnO and nano TiO_2 with acrylic binder. They found that the UV protection function of the fabrics treated with larger sized TiO_2 nanoparticles have better UV protection factor (UPF) value than the fabric treated with smaller sized ZnO nanoparticles.

Conclusion

Self-cleaning concept contributes a lot of benefits in various industries. Especially, self-cleaning fabric which has massive potential for improvement of products not only in clothing industry but also in health industry due to time, material, energy reduction and consequently cost-efficiency during production. Moreover, this technology embraces environmental friendly properties as it effectively decreases cleaning efforts and conserve a considerable amount of water and energy as well as saving time and laundering cost.

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