



# Current Approach to Develop TiO<sub>2</sub> Thin Film as Photocatalysts for Low-Density Plastic Degradation

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## ABSTRACT

Low-density plastic bags waste disposal is a big issue in the current scenario which gives rise to grave threats to human beings and environmental health also. Amid the various approaches applied for dealing with the problem, photocatalytic biodegradation in visible light irradiation is an advanced prospect that has received attention nowadays. The present review paper is to provide an outline of the current progress on the synthesis of titania (TiO<sub>2</sub>) thin-film photocatalysts for solid waste removal. The Photocatalysis method contains the photoinduced redox reactions in the photocatalyst which facilitates the degrading of almost organic compounds like polyethylene into carbon dioxide (CO<sub>2</sub>), water, and other substance. One of the most excellent photocatalysts which has grabbed attention in an application is titania because of its high photocatalytic activity and chemical stability. The synthesis of the photocatalyst as a thin film is a result of the unfeasible application of conventional powder photocatalyst which may cause a certain environmental hazard. The photocatalyst-coated thin film along with some environmental applications have also been reviewed. Likewise, various approaches for modifying thin-film property, film deposition techniques, and deposition on various substrates are used for the enhanced photocatalytic activity of the TiO<sub>2</sub> thin film.

## INTRODUCTION

Plastic waste is a worldwide issue; still, there is variability from region to region. Fumes released from plastic burning are one of the sources of environmental air pollution. Furthermore, dumping plastic into the ocean leads to releasing toxic chemicals contained in it because of which a large amount of water is polluted. Plastic waste accounts for approximately 10% of all household waste disposed of in landfills (Barnes et al. 2009, Hopewell et al. 2009). However, 60 to 80 percent of waste is generated along coastlines, with the amount varying depending on the ocean (Barnes 2002, Derraik 2002). According to Central Pollution Control Board (CPCB) estimation, 8 million tons per annum of plastic is used in India, and annually about 5.7 million tons of plastic is transformed into waste (Rathi 2007). The rise in plastic consumption and production has resulted in increases in plastic waste (UNEP 2009). As a result, in 2007, higher than 250 million tons of plastic waste was generated. Plastic materials are not biodegradable and have a low density, making them unsuitable for landfill disposal. Since 1950, this figure has risen at an average annual pace of roughly 9%, reaching 245 million tonnes in 2008. Plastic bags and other plastics have been a focus since they have been linked to several other problems in India, including animal deaths, contaminated soils, and clogged sewers (Gawande et al. 2012).

Large amounts of plastic will result in an increase in waste and have an adverse impact on the environment. Due to the lack of proper waste management practices, plastic waste will increase, adding to the already existing plastic waste. There is no evidence that specifies a definite time for plastic degradation, however, it could be thousands of years (Kershaw et al. 2011).

Waste disposal in landfills results in the irrevocable loss of major crude materials and energy. The incomplete ignition of Polyethylene (PE), Polystyrene (PS), and Polypropylene (PP) at the time of thermal usage can lead to a high amount of carbon monoxide (CO) and emanations, whereas PVC produces carbon black, dioxins, and aromatics compounds such as pyrene and chrysene. Bromide and different color pigments containing heavy metals such as copper, chromium, selenium, cobalt, cadmium, and lead, among others, can be detected in a noxious discharge. Plastic wastes are always accumulated at open grounds, drifts, railroad tracks, and drains (Islam et al. 2011). Despite many efforts to manage waste, over 91% of MSW gathered is still landfilled or unloaded on open grounds (Verma et al. 2016).

Every year, around 140 million tonnes of plastic are produced, with the excess amount ending up in the environment as an industrial waste dump (Shimao 2001). Cosmetics, food packaging, synthetic compounds, pharmaceuticals,

and cleansers account for about 30% of all plastics used worldwide. Furthermore, this is still increasing at a rapid rate of 12 percent every year (Sangale et al. 2012). Plastics have supplanted paper and other cellulose-based items for packing as plastics have good elasticity, delicacy, protection from water, and microbial resistance. Usually applied plastics have mostly been used in polyethylene (Low Density, medium density, high density, and linear low density), polystyrene (PS), polyvinyl chloride (PVC), and polypropylene (PP) (Khanam & AlMaadeed 2015). Because of its hydrophobic backbone chain, low-density polyethylene is classified as a thermoplastic (Pramila & Ramesh 2015). It is considered non-degradable in nature (Misra et al. 2015). As a result, several countries have been forced to devise measures to counteract the threat.

The environmental impact of main and secondary sources of microplastics must be addressed immediately (Arthur et al. 2009). More research is needed to determine how many degrees of exposure are caused by plastic waste, as well as the components of plastic that may impact humans and animals. Those at the high degree of the food chain would be exposed to synthetic substances in larger quantities.

### TiO<sub>2</sub> as a Photocatalysts

Since ancient times, TiO<sub>2</sub> powders have been widely used in paints to get white shades (Hashimoto et al. 2005). Later, its photocatalytic activity was explored and considered the most effective photocatalyst (Fujishima & Honda 1972). TiO<sub>2</sub> is known for its different properties, including strong photoactivity, high stability, low cost, and low toxicity (Pant et al. 2014a, Pant et al. 2020). Over the last few years, TiO<sub>2</sub>-based nanomaterial has received a lot of attention in academia and has been used in a variety of applications, including the removal of natural toxins, sensors, photovoltaics, antimicrobials, and energy conservation (Bai & Zhou 2014, Hsu et al. 2019, Nakata & Fujishima 2012, Pant et al. 2014a, 2019b, 2020, 2016).

TiO<sub>2</sub> nanostructures with a bandgap of 3.0–3.2 eV can be activated in the presence of UV light irradiation. Because of this, the use of TiO<sub>2</sub> is limited because only 5% of the sunlight falls inside the UV spectrum (Daghrir et al. 2013, Pant et al. 2014b, 2019a). TiO<sub>2</sub> is found in three types of crystal structures i.e. anatase, rutile, and brookite (Allen et al. 2018). Although brookite is known as the most unstable structure and is not favored for photocatalytic activity, rutile and anatase type crystal forms are examined for photocatalysis (Di Paola et al. 2013, Zhang et al. 2008). The bandgaps of mass anatase and rutile are 3.2 and 3.0 eV respectively, which relate to the absorbance at 388 nm and 414 nm, individually (Dette et al. 2014). In comparison to rutile, the anatase phase

crystal structure is favored as the most photo catalytically active due to its ability to adsorb hydroxyl groups and water (Wang et al. 2018). Research has indicated that the synergistic impact among anatase and rutile is useful in upgrading the photocatalytic activity of the TiO<sub>2</sub> nano-structures (Zhang et al. 2008).

When a photon (with energy equal to or greater than the bandgap of TiO<sub>2</sub>) stimulated the TiO<sub>2</sub> thin film or nanoparticles, the electrons moved from the valance bandwidth to the conduction bandwidth and generated electron-hole pairs. The created charge particles travel to the surface area and quickly react with the adsorbed organic compounds, resulting in the organic pollutant and solid waste being degraded. Fig. 1 depicts the technique for highly excited oxidant production and breakdown of organic waste.

### Correlation of Thin-film Structure and Photocatalytic Activity

Titania (TiO<sub>2</sub>) has been documented as an efficient photocatalyst along with high chemical stability and potent photocatalytic activity. As the outcome of various research, it was found that Titania has better photocatalytic activity than Zirconium oxide (ZrO<sub>2</sub>), Tungsten trioxide (WO<sub>3</sub>), Tin (IV) oxide (SnO<sub>2</sub>), and Cadmium sulfide CdS. For instance, for the decomposition of phenol organic compound, TiO<sub>2</sub> was exhibited higher photocatalytic activity compared to CdS (Thiruvengkatachari et al. 2008). Titania is present in three different types of crystal structure; rutile, brookite, and anatase. Titania is primarily used in the anatase-type crystal structure as the photocatalyst since it has greater bandgap energy than rutile. Because of this, the anatase crystal structure can reduce electron-hole recombination, resulting in a higher degree of surface hydroxylation and the ability to create more hydroxyl radicals as the surface is illuminated. Certainly, this can lead to the efficiency of photocatalytic activity. On the other side, the crystal structure with a rutile type can absorb the beams that are somewhat nearer to the visible light range. It is accepted that the rutile type is more desirable as a photocatalyst for photocatalysis applications. Nonetheless, the rutile type energy structure is unable to provide the preferred photocatalytic execution since its conduction band is close to the redox potential of hydrogen, resulting in its reduced ability.

The oxygen ions present on the surface of the anatase crystal structure are displayed in a triangle shape, allowing better absorption of organic substances such as polyethylene (Thiruvengkatachari et al. 2008). Furthermore, the location of Ti ions provides a more favorable environment for reaction, as does the absorbed organic compound, which is not accessible in the rutile crystal structure (Thiruvengkatachari et

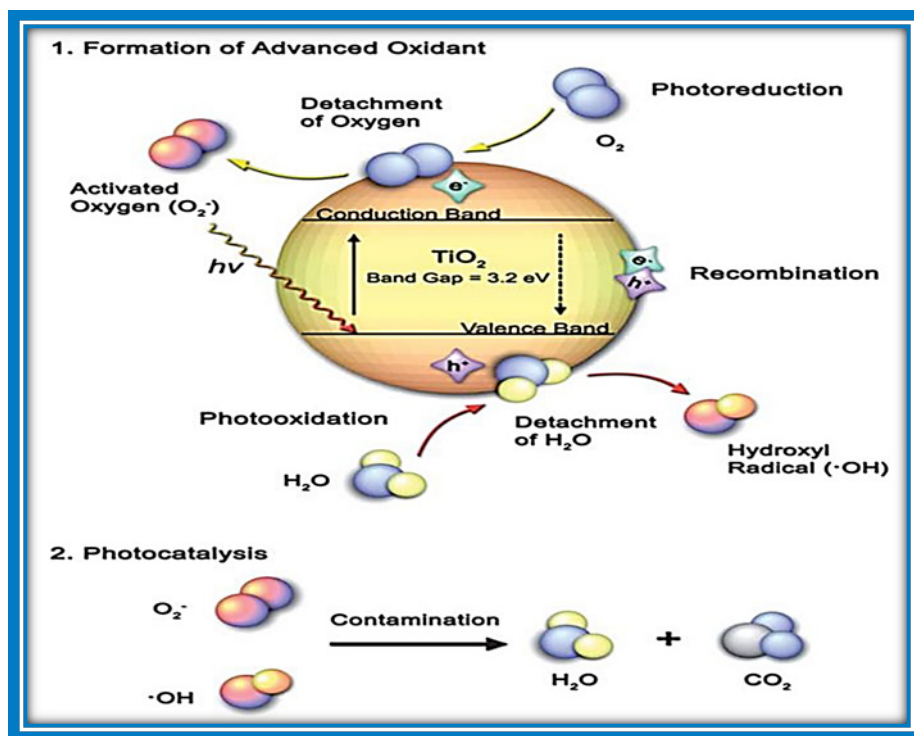


Fig. 1: A schematic diagram of photocatalysis by TiO<sub>2</sub> nanostructure (Tung & Daoud 2011).

al. 2008). This remarkable property of anatase would result in strong photocatalytic activity. Nonetheless, a few studies revealed that pure anatase titania would not result in significantly improved photocatalytic activity (Thiruvengkatachari et al. 2008). The presence of a mesoporosity type structure and more extensive pore dissemination in a small percentage of the rutile phase crystal structure could explain the increased photocatalytic action. One of the instances is the commercially accessible Degussa P25 TiO<sub>2</sub>, which consists of 70% anatase and 30% rutile type crystal structure.

The three-sided layout of oxygen particles on the uncovered precious stone surface of anatase allows for strong organic retention (Thiruvengkatachari et al. 2008). Furthermore, the presence of titanium particles provides a positive response state with assimilated organics that are inaccessible to the rutile structure (Thiruvengkatachari et al. 2008). This special character of anatase would prompt high photocatalytic movement. In any case, a few specialists expressed that unadulterated anatase TiO<sub>2</sub> would not lead to much better photocatalytic execution (Thiruvengkatachari et al. 2008). The inclusion of a few percent of the rutile stage results in a mesoporosity structure and more widespread pore dissemination, which could explain the increased photocatalytic activity. One of the models is the financially accessible Degussa P25

TiO<sub>2</sub>, which contains 70% anatase and 30% rutile.

The surface area of photocatalytic agents is also a key factor that influences photocatalytic activity. A small size particle has a larger surface area and thus increases the availability of active sites per square meter, which boosts pollutant absorption on the photocatalytic agent's surface. Photocatalytic activity is boosted as a result of this mechanism (Thiruvengkatachari et al. 2008).

Several investigations support that TiO<sub>2</sub> is a better photocatalytic agent for the degradation of organic compounds like polymer. Moreover, after degradation, no harmful by-products are released and this method has been confirmed as potent in eradicating trace concentrations of organic substance present in the liquid or gaseous phase. For instance; organic acid (Fallet et al. 2006, Sheng et al. 1999), alcohol (Choi et al. 2004, Xianyu et al. 2001), organic dyes (Bouras & Lianos 2005, Funakoshi & Nonami 2007, Ge et al. 2006, Gu et al. 2007, Jung & Imaishi 2001, Stoyanov et al. 2006, Yu et al. 2002b, Yuan et al. 2006, Zhou et al. 2006b), oxide gaseous (Takeuchi et al. 2000, Yu et al. 2006a, 2006b, Yusuf et al. 2002), and aldehyde gas (Chiba et al. 2005, Noguchi et al. 1998, Sopyan 2007, Sopyan et al. 1996, Watanabe et al. 2000) have been decomposed by TiO<sub>2</sub> photocatalytic activity. Various searches for biodegradation of organic

substances such as various plastics using microflora such as bacteria, fungi, and algae have also been documented (Ditta et al. 2008, Sunada et al. 1998, Sunada et al. 2003). To date, only fine powders type photocatalysts are applied for decomposing pollutants as it has more oxidation ability to make photo-generated holes on the outer surface photocatalysts. Nevertheless, the application of fine powder photocatalysts may cause some serious problems such as [a] Complications to separate suspension and the photocatalyst (Begum & Ahmed 2008, Cho et al. 2007, Ge & Xu 2007, Zhou et al. 2006a), [b] High amount of fine powder of photocatalysts may be aggregated with suspended particles (Cho et al. 2007), and [c] Photocatalyst powder can be easily separated from weak adhesion supports if it is covered with particular support (Cho et al. 2007). As a result, advanced approaches such as a thin coated layer on solid wafer material, which makes it a more potent photocatalyst, should be used for critical environmental applications. Numerous materials like polymer, cement, clay, silica and mild steel have been applied as the adherence of TiO<sub>2</sub> photocatalytic agents. In the current review, TiO<sub>2</sub> film as a photocatalyst and various methods for the improvement of photocatalytic characteristics, TiO<sub>2</sub> deposition methods, and its environmental application for degradation of polyethylene will be also discussed.

### Photocatalyst Thin Films

Nanocrystalline semiconductor thin films found a network in which electronic conduction can happen (Beydoun et al. 1999). A photocatalyst-coated thin film has unique efficiency properties that make it ideal for environmental purification. It is evidenced that titania thin films are extensively applied in various fields like adsorbent catalysts, as sensors (humidity and gas) functional materials, and mainly pollutant removal from wastewater and air (Machida et al. 1999, Mei et al. 2006). A photocatalyst-coated thin film can prevent inhaling particle problems caused by nano-photocatalyst powder, which can be hazardous to human health (Yu et al. 2006a). Furthermore, it is more dependable than the immobilization method, and as a result, photocatalyst-coated thin films may be applied to a number of substrates, such as thin film-coated stainless steel for a building, which has greater chemical corrosion resistance and mechanical strength. Besides this, the building material could be converted into an air-purifying and self-cleaning substance after being coated with a thin film of photocatalyst (Yu et al. 2006a). The penetration of UV or visible light is a key drawback for the photocatalysis process using slurry or powder of nanoparticles. However, these problems can be overcome by photocatalyst-coating on a solid substrate. Contrasting the nanoparticle, the photocatalyst-coated thin film has the ability to avoid light scattering and improve the light transmittance and ultimately increase

the efficiency of photoreaction (Ge et al. 2006, Kitano et al. 2007, Otsuka et al. 2008, Sheng et al. 1999, Yu et al. 2002a, 2002b). Furthermore, the advantage of the photocatalyst-coated thin film is connected with an external layer only.

### Deposition Method for the Development of Photocatalyst Thin Film

Various techniques have been applied for coating the photocatalyst thin film on different substrates such as reactive magnetron sputtering (Choi et al. 2004, Sheng et al. 1999), RF reactive ion plating (Chiba et al. 2005), solgel (Hu et al. 2008), liquid phase deposition (LPD) (Gu et al. 2007, Yu et al. 2006b, Zhou 2006b), vapor phase deposition (Chemical vapor deposition and metal-organic Chemical vapor deposition (Jung & Imaishi 2001), physical vapor deposition (PVD) (Aziz & Sopyan 2009), reactive evaporation (Kitano et al. 2007), atomic layer deposition (ALD) (Clouser et al. 2008), ionized cluster beam (ICB) (Zhou et al. 2006a), electrophoretic deposition (Chan et al. 2002), and pulsed laser deposition (PLD) (Kitano et al. 2007). Each technique has its specific pros and cons. For example, the solgel method required high-temperature calculations after treatment to achieve the required stabilities and mechanical strengths of thin coated films. (Zhou et al. 2006a). It could occur as a result of damage to porous substrates. The properties of TiO<sub>2</sub> thin films are largely dependent on the deposition process as well as the deposition parameter, according to several research findings (Hasan et al. 2008). All the above methods have been modified by researchers to obtain and develop nano-sized thin film along with required specific physical characteristics such as size, porosity, shape, crystalline structure, and surface area as per their application.

### Magnetron Sputtering Method

Some deposition methods have limitations for developing photocatalytic active thin films, such as spray coating, which requires a high temperature to decompose metallorganics during the heating process and is limited to non-refractory substrates (Fig. 2). Sheng et al. (1999) made an effort to develop photocatalytically active Pt-titania thin films along with the anatase crystalline structure at 200°C temperature on a flexible substrate i.e. polyamide to broaden the application of film as a photocatalyst. Fleetingly, they used the tripole magnetron sputtering system to develop thin films by applying two different methods of deposition. In the first method, interchangeably, sputtering co-axially placed titanium and platinum targets, and for platinum, the deposition time was 2 s, while for titanium it varied from 20 s to 140 s throughout each cycle to prepared different compositions. In the second method, the Pt-titania film was developed with



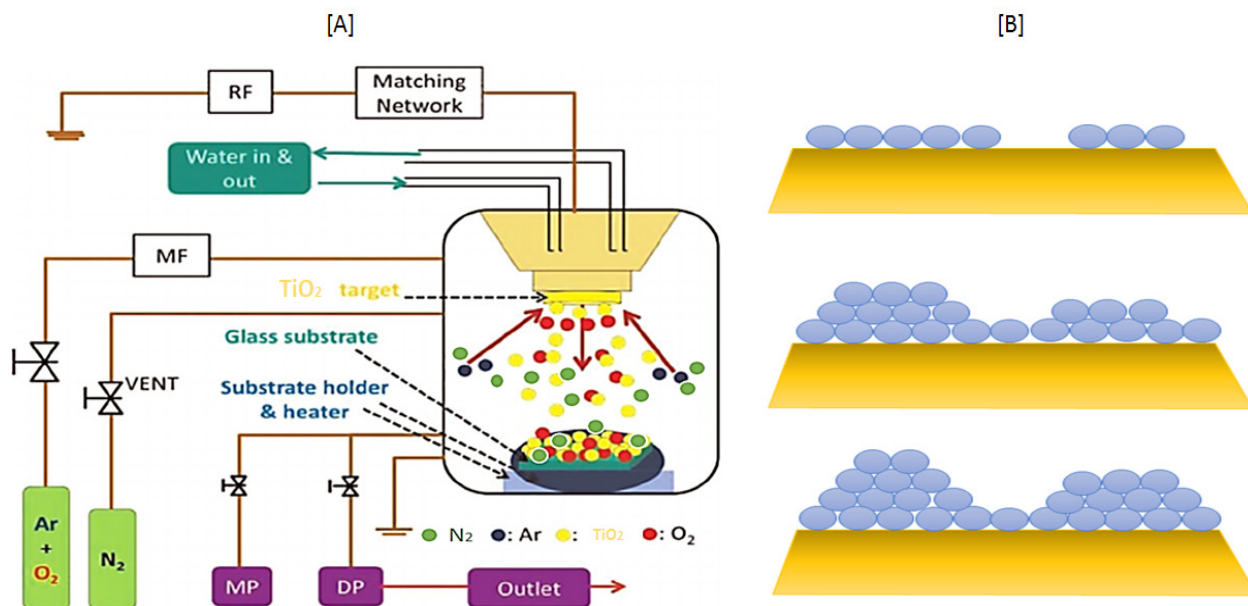


Fig.2: [A] A schematic diagram of RF magnetron sputtering machine [B] A diagram of thin-film growth mode.

pure titania film depositing first on a polyamide substrate, afterward coating a thin layer of platinum on the surface then again coated with titania thin film. They revealed that coating using the second method is to provide bonding strength between film and polyamide along with anatase crystalline structure at 200°C deposition temperature. Tomaszewski et al. (2007) used DC magnetron sputtering on ceramic targets to create transparent nanostructured TiO<sub>2</sub> thin films. Mei et al. (2012) used DC reactive magnetron sputtering to create transparent titania thin films on silica substrates at varied deposition temperatures (300-600°C). They discovered that at 500°C, TiO<sub>2</sub> thin films had the maximum photocatalytic activity, owing to the crystalline structure of TiO<sub>2</sub> thin films (Mei et al. 2012). Kavaliunas et al. (2020) studied the effect of dopants Mg, Cu, and Ni on photocatalytic activity of amorphous titania thin film which was developed by reactive magnetron sputtering. The results showed that dopant concentrations in titania films between 0.1 and 0.9 percent gave the best photocatalytic activity. SantAna et al. (2020) investigated of optical transmittance of photocatalytic active titania films which were deposited using radiofrequency magnetron sputtering (SantAna et al. 2020). They revealed the optimum deposition parameter such as 0.2 electric currents (A), 437 voltage (V), 87 RF power (W), 3600 deposition time (s),  $9.2 \times 10^{-3}$  background pressure (mbar). Moreover, the experimental results show that the TiO<sub>2</sub> films deposited on glass were associated with the anatase phase with a [0 0 4] preferred orientation. Ultraviolet near-infrared spectroscopy revealed the high transmittance of the films in visible light

and high absorption in the ultra-violet region under most deposition conditions.

## CONCLUSION

This review emphasized the necessity of the elimination of plastic waste contaminants from landfills. Various types of plastic waste pollutants have been listed and the constraint of initial titania photocatalysis on plastic degradation was discussed. Some deposition parameters which have significant impacts on the titania-based photocatalytic degradation, and depositing parameters like dopant, particle size, catalyst concentration, temperature, crystalline structure, and selection substrate that aids in the use of titania as a photocatalyst for further commercialization were reviewed. Hence, photocatalytic process design and technical deposition method optimization are critical to investigate using a key emphasis on low-density plastic deterioration.

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