



## **Titanium Dioxide - Advances and Applications**

*Edited by Hafiz Muhammad Ali*

Titanium Dioxide - Advances and Applications  
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# Preface

Titanium dioxide plays an important role in many scientific and engineering applications, thus it is important to further explore the fundamentals and mechanisms of this useful compound.

This book focuses on recent advances and applications of titanium dioxide. It examines the compound's applications in environmental remediation, photocatalytic materials, rechargeable lithium-ion batteries, thin films, energy storage, semiconductors, and much more. It is a useful resource for researchers, scientists, engineers, and students. Chapters include various illustrations and graphs to represent the information in a comprehensive manner.

I would like to extend my appreciation and thanks to the contributing authors. I would also like to express my gratitude and appreciation to my beloved wife Aysha Maryam Ali for her companionship, continuous support, and patience throughout the preparation of this book.

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# TiO<sub>2</sub>: A Semiconductor Photocatalyst

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## Abstract

Titanium dioxide (TiO<sub>2</sub>) is considered as an inert and safe material and has been used in many applications for decades. TiO<sub>2</sub> have been widely studied, due to its interesting general properties in a wide range of fields including catalysis, antibacterial agents, in civil as nano-paint (self-cleaning) and especially photocatalysis, and that affect the quality of life. Thus, the development of nanotechnologies TiO<sub>2</sub> nanoparticles, with numerous novel and useful properties, are increasingly manufactured and used. TiO<sub>2</sub> doped with noble metal are good candidates in the performance these applications. The fascinating physical and chemical features of TiO<sub>2</sub> depend on the crystal phase, size and shape of particles. For example, varying phases of crystalline TiO<sub>2</sub> have different band gaps that rutile TiO<sub>2</sub> of 3.0 eV and anatase TiO<sub>2</sub> of 3.2 eV, determine the photocatalytic performance of TiO<sub>2</sub>. This chapter explains basic information on TiO<sub>2</sub> and theoretical concepts of nanostructure of TiO<sub>2</sub> nanoparticles as a semiconductor photocatalyst.

**Keywords:** TiO<sub>2</sub>, nanoparticles, semiconductor, photocatalyst

## 1. Introduction

An ideal photocatalyst should possess following nature: biologically and chemically inert, inexpensive, nontoxic, stable towards photocorrosion and certainly highly active and suitable towards the visible/UV light photoreaction. An additional criterion for such ideal photocatalyst is better redox potential of the H<sub>2</sub>O/\*OH couple (EOP = 2.80 V) [1]. Among the available semiconductors, TiO<sub>2</sub>, ZnO, ZnS and WO<sub>3</sub> have gap energies sufficient for catalyzing a wide range of chemical reactions [2].

Binary metal sulphide semiconductors such as CdS, CdSe or PbS are regarded as unstable photocatalyst due to easy photoanodic corrosion property and their toxic nature [2, 3]. Furthermore, the iron oxide semiconductors are not suitable as they readily perform the photocathodic reaction and finally produced corrosive materials [1]. ZnO on the other hand is known to have the similar quality of band gap energy like TiO<sub>2</sub> (3.0 eV) but it is highly unstable towards pH condition, where precipitate of Zn (OH)<sub>2</sub> is formed on the particle surface and resulted in photocatalyst deactivation [2].

Of these, titanium dioxide (TiO<sub>2</sub>) is the most ideal semiconductor for photocatalytic destruction of organic compounds [2, 4]. It also provides an excellent conciliation between catalytic performance and stabilization in aqueous media [5].

## **2. Titanium dioxide (TiO<sub>2</sub>)**

The strong oxidation and reduction power of photoexcited TiO<sub>2</sub> was successfully discovered by Honda-Fujishima [6, 7]. Since its innovation, extensive efforts have been made in the development of TiO<sub>2</sub> photocatalyst owing to its potential application in the removal of various types of organics and in both aqueous and air phase [8, 9]. It resulted in flexible applicability either in the form of a suspension, or immobilization [10–13].

Regardless the high superior photocatalytic efficiency of TiO<sub>2</sub>, it is popular for its unique wider applications as electroceramic, antifogging agent and as therapy for cancer [14, 15]. Moreover, it also acts as an antibacterial agent due to its strong oxidation activity and superhydrophilicity and was employed for electrolysis of water (H<sub>2</sub>O) to produce H<sub>2</sub> and for harvesting solar energy as dye-sensitized solar cells [16]. Additionally, it possesses superior pigmentary property, UV light absorption capability and durability [17]. The high refractive indices of rutile and anatase phase of TiO<sub>2</sub> laid a pathway for its versatility nature.

In general, the crystallinity, impurities, specific surface area and density of the surface hydroxy groups are the physical parameters that have influence over the photocatalytic activity of TiO<sub>2</sub>. Apart from that the band gap energy, charge-carrier and recombination as well play a crucial role for photoactivation [18, 19]. Among the available phases the anatase is the most stable form with 8–12 kJ/mol. This describes the quantity of energy transferred during phase transformation. The rest phases such as brookite is very uncommon [14, 20–21].

In the terms of density, rutile is greater than anatase, i.e., 4.26 and 3.90 g/mL respectively. However, in terms of utilization and ability, anatase phase is more efficient due to its open structure [14]. The anatase phase remains as most active photocatalyst with greater stability after repeated catalytic cycles illuminated under a photon energy between 300 nm <  $\lambda$  < 390 nm [22]. In addition, it is also chemically stable in aqueous media and in broad range of pH (0 < pH < 14) [23]. Hence TiO<sub>2</sub> with its major anatase phase had more applicability as photocatalyst [24]. Furthermore, it also exhibits relatively high reactivity and chemical stability only under UV light ( $\lambda$  < 387 nm) [7]. Thus, its limited utilization of the activation energy becomes a major drawback. Therefore, to overcome this limitation of TiO<sub>2</sub>, an extensive research needs to be emphasized to develop a titania based photocatalyst that can exhibit high reactivity under visible light spectrum ( $\lambda$  > 400 nm) and can persuades the utilization of solar spectrum, even under poor luminance of interior lighting [7].

The reduction in energy gap between the CB and VB further leads to the easy recombination of the formed electrons and holes. This recombination further decreases the interfacial charge-carrier transfer. The size of the TiO<sub>2</sub> further causes difficulties in separating them from the waste's stream. All these issues stress the reseachers to find a suitable composition/dopant for its modification [25].

## **3. Modification of TiO<sub>2</sub> photocatalyst**

The photoactivity of TiO<sub>2</sub> is hindered by the narrow UV wavelength spectrum for photoactivation under ambient conditions. The generation of this UV requires additional power source, which shoots the activation cost of the photocatalyst. Therefore, the utilization of renewable energy source could be a better sustainable choice for the photoactivation [26].

Type	Remark	Reference
Chromium (Cr)	This is a study on the effect of the photoreduction of dinitrogen in a gas-solid regime using Cr as a dopant for phenol degradation. The prepared Cr-TiO <sub>2</sub> showed an enhanced activity and displacement of the Fermi level at the TiO <sub>2</sub> interface. But the presence of Cr as donor ion did not favor the charge separation with respect to pure TiO <sub>2</sub> .	[34]
Iron (Fe)	Fe <sup>3+</sup> ion has a unique half-filled electronic structure which resulted in enhanced activity compared to Fe <sup>4+</sup> and Fe <sup>2+</sup> . The stable Fe <sup>3+</sup> ion detrapped the electron and hole to adsorbed oxygen and surface hydroxyl group, thereby suppressed the electron-hole recombinations.	[35]
	This study is about the effect of Fe doping into TiO <sub>2</sub> for the degradation of chloroform (CHCl <sub>3</sub> ). It observed that Fe provided shallow trapping sites for the charge-carriers and increase the photocatalytic efficiency. It also found that Fe can act as trap for both electron and hole, at high dopant concentration.	[36]
	This study is about the Fe-doped with TiO <sub>2</sub> for the degradation of 1, 2-dichloroethane. The study proved that Fe <sup>3+</sup> alleviates the surface phenomenon and act as both electron-hole traps.	[37]
	The effect of Fe doping for the inactivation of <i>E. coli</i> was studied. The Fe-doped TiO <sub>2</sub> proved the enhancement in the inactivation of <i>E. coli</i> . Fe played as a source for the inhibition of the anatase crystal growth.	[38]
Molybdenum (Mo)	The Mo as a cationic dopant was studied in 2009. The dopant increased the surface acidity of TiO <sub>2</sub> and accelerated the interfacial charge transfer process. Thus, prepared doped photocatalyst degraded the cationic dyes, rhodamine B (RhB) and methylene blue (MB) and anionic dyes, methyl orange (MO) and Congo red (CR) at both alkaline and acidic pH.	[39]
Zinc (Zn)	Similarly, the same group studied the effect of Zn as a dopant in 2010 and applied the prepared photocatalyst for the degradation of CR. They produced a small crystallite size and stable filled electronic configuration of Zn <sup>2+</sup> -TiO <sub>2</sub> . Moreover, this photocatalyst provided a shallow trap for the charge carriers contributing to the overall activity.	[40]
Manganese (Mn)	They further extended their study with Mn as a potential cationic dopant in 2011. From the study, it showed that Mn <sup>2+</sup> intimate contact between the mixed anatase and rutile phase of TiO <sub>2</sub> . Moreover, this dopant has synergistic effects in the bicrystalline framework of anatase and rutile. The unique half-filled electronic structure of Mn <sup>2+</sup> served as a shallow trap for the charge-carrier contributed for the appreciable degradation of both Indigo carmine dye and 4-chlorophenol.	[41]
Cobalt (Co)	Amadelli et al., 2008 studied the effect of Co-doping. They prepared the photocatalyst by incipient impregnation method and cobalt acetate as a precursor. Co addition brings about conspicuous changes in the point of zero charge and in surface polarity. They found that Co-doped TiO <sub>2</sub> is more active compared to TiO <sub>2</sub> . The best photocatalytic result of the prepared photocatalyst is obtained for heat treatment at 400°C for 30 min.	[42]
Thorium (Th)	Thorium a naturally occurring radioactive element was studied as a dopant in 2009. It observed that Th can contributed well for the BET surface area of photocatalyst. Moreover, the presence of two absorption edges at 460 and 482 nm in visible region enabled the photocatalyst to utilize more visible light. They reported that the strong adsorption of the pollutant was due to the complex formation between the vacant f orbital of Th <sup>4+</sup> . Besides, they observed that Th induced the oxygen vacancies which served as shallow traps. But thorium could further contribute for radioactive pollution.	[39]

**Table 1.**  
 Literature review of selected metal dopant/hybrid on TiO<sub>2</sub>.



Type	Remark	Reference
Nitrogen (N)	In 2001, researchers prepared $\text{TiO}_{2-x}\text{N}_x$ by sputtering the $\text{TiO}_2$ selectively on $\text{N}_2$ (40%)/Ar gas mixture for degradation of MB and acetaldehyde ( $\text{CH}_3\text{CHO}$ ) under UV/visible light.	[43]
	Researchers prepared a narrow band gap N-TiO <sub>2</sub> with oxygen vacancies. Thus, prepared N-TiO <sub>2</sub> failed to oxidize formate and $\text{NH}_3\text{-OH}^+$ under visible light illumination. Their preparation method resulted the N as a weak anion donor.	[44]
	This is the study about the preparation of N-TiO <sub>2</sub> by thermal decomposition of the $\text{Ti}^{4+}$ -bipyridine complex and had moderately better removal of NO <sub>x</sub> under artificial visible light.	[45]
	The researchers prepared $\text{TiO}_{2-x}\text{N}_x$ by solvothermal route, produced violet color of particles and further calcination between 200 and 800°C produced weak violet, bright yellow, weak yellow and gray; no white color of particles. Their findings proved the thermal stability of Ti-N bonding with $\text{TiO}_{2-x}\text{N}_x$ .	[46]
	N-TiO <sub>2</sub> was prepared using Ti-melamine and Ti-salen complex as precursors and reported a higher photoactivity was obtained for the Ti-melamine by degrading MB degradation. The reasoned for the low photoactivity of the Ti-salen complex was due to the low amount of N doping and smaller Ti-N bonds.	[47]
	N-doped layered titanates ( $\text{Cs}_{0.66}\text{Ti}_{1.33}\text{O}_4\text{-N}_x$ and $\text{H}_{0.66}\text{Ti}_{1.33}\text{O}_6\text{-N}_x$ ) with leucoprocate structure was prepared and achieved better photoactivity by degrading RhB under visible light illumination. The unique layered structure of titanates and homogeneous distribution of N dopant contributed for the mobility of the charge-carrier further contributed for the faster and better photoactivity.	[48]
	Multitype N-TiO <sub>2</sub> containing both substitutional (N-Ti-O and Ti-N-O) and interstitial NO characteristics was prepared. The substitutional and interstitial technique resulted in intraband states at 0.14 and 0.73 eV above the VB resulting in faster visible light driven photodegradation of gas phase toluene.	[49]
Carbon (C)	A carbonaceous coke-like species embedded in the $\text{TiO}_2$ matrix was obtained by pyrolysis of alcohol in the sol-gel titania processing and calcined at 250 and 400°C. The resulted C-TiO <sub>2</sub> calcined at 250°C exhibited a maximum photoactivity for degradation of 4-chlorophenol (4-CP) under visible light than that the later. Further calcination to the higher temperature caused the elimination of carbon from the titania matrix. Thus, showed significance of the calcination temperature.	[50]
	A rutile $\text{TiO}_2$ doped with carbon was prepared by a pyrolysis method using combustion products namely $\text{CO}_2$ and $\text{H}_2\text{O}$ and used for water splitting reaction. Their doping method resulted in a band gap of 2.32 eV against 3.0 eV and showed the supremacy of the preparation method.	[51]
	$\text{TiO}_2\text{-G}$ (titania-glycine) was successfully prepared through solution combustion route by glycine (G), hexamethylene (H) and oxalyldihydrazide (O) as fuel. The precursors employed contributed for the carbide ion and thus reduced band gap energy and resulted in a superior photoactivity, crystallinity and surface area. The photoactivity was successfully experimented by degrading aqueous phase MB under solar irradiation.	[52]
	$\text{TiO}_2\text{-C}$ was prepared through a simple carbon sources, which is tetrabutylammonium hydroxide ( $\text{C}_{16}\text{H}_{37}\text{NO}$ ) and glucose ( $\text{C}_6\text{H}_{12}\text{O}_6$ ) as precursors. The adopted precursors contributed in two visible absorption edges in their band gap namely 2.78 and 1.45 eV, respectively.	[53]
	Park et al., 2009 showed the importance of optimum calcination temperature like Lettmann and coworkers in 2001.	[54]

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