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### Review Article

# **Functionalized Activated Carbon Derived from Biomass for Photocatalysis Applications Perspective**

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This review highlighted the developments of safe, effective, economic, and environmental friendly catalytic technologies to transform lignocellulosic biomass into the activated carbon (AC). In the photocatalysis applications, this AC can further be used as a support material. The limits of AC productions raised by energy assumption and product selectivity have been uplifted to develop sustainable carbon of the synthesis process, where catalytic conversion is accounted. The catalytic treatment corresponding to mild condition provided a bulk, mesoporous, and nanostructure AC materials. These characteristics of AC materials are necessary for the low energy and efficient photocatalytic system. Due to the excellent oxidizing characteristics, cheapness, and long-term stability, semiconductor materials have been used immensely in photocatalytic reactors. However, in practical, such conductors lead to problems with the separation steps and loss of photocatalytic activity. Therefore, proper attention has been given to develop supported semiconductor catalysts and certain matrixes of carbon materials such as carbon nanotubes, carbon microspheres, carbon nanofibers, carbon black, and activated carbons have been recently considered and reported. AC has been reported as a potential support in photocatalytic systems because it improves the transfer rate of the interface charge and lowers the recombination rate of holes and electrons.

### 1. Introduction

Increasing environmental problems and the need for competitive and cost-effective products are becoming two major principles in modern material research [1-4]. Former developed routes to get a periodic porous carbon network were successful, but they did not take into account any criteria of sustainability [5-7]. For the last 20 years, countless laboratories of research institution have done some indepth researches on the conversion of biomass to carbon based materials without using the catalysts [5, 8, 9]. Studies of such researches have covered many model carbon material compounds, for example, methanol, lignin, glucose, cellulose, and some real biomass compounds [10–12]. As prosperous demonstrations were accumulated, kinetics, thermodynamics, and detailed reaction mechanism have created a solid base for subsequent researches [13-15]. However, in order to enhance the selectivity of carbon material manufactures, immense activation energy is necessary for the reaction without the use of catalyst. The excessive cost of tools and

operations has undoubtedly become the biggest hindrance to the formulation of this technology [15].

Hence, the main problem of carbon synthesis under sustainable conditions was currently revisited and executed by several terms, where catalytic treatment of biomass through either heterogeneous or homogeneous catalytic approach corresponding in mild condition provided a bulk, mesoporous, and nanostructure carbon materials [16-18]. Catalytic effects of homogeneous catalyst, especially ionic liquid on the biomass conversion, have been established by many of the open literature. The main characteristics of this catalytic technique are to have a conversion system with minimal energy to confirm the high yield of carbon materials [18-20]. If we compare homogeneous catalysts with heterogeneous catalysts, heterogeneous catalysts have the advantages of being highly selective, recyclable, and environmentally friendly [19, 21]. Heterogeneous catalysts have the advantages of being highly selective; therefore, heterogeneous catalyst with the ample range of solid acid, ion exchange resin, metal oxide, and zeolite has become a research hotspot in this

field [21]. Both techniques have been explored substantially, with the need for creating cheap and sustainable ways to obtain chemicals and carbon from raw materials other than natural gas or crude oil that could lead to a reexploration of this area. In addition, the implementation of low-priced pathway to recycle the products of farmed biomass would furthermore represent a way to sequester particular amount of  $CO_2$  creating a material advantaged at the same time [22, 23].

This paper will not debate on the preparation of the catalyst, but instead it would focus primarily on the use of the catalyst to bring out carbon based materials through the conversion process of biomass, which can be additionally used as a photocatalyst.

Various published papers and examined articles have indicated the theory and environmental supplication of heterogeneous photocatalysis by the employment of semiconductors [24, 25]. One of the biggest drawbacks of using these semiconductors is the power necessity because of the utilization of ultraviolet light [26]. However, upgrading the photocatalytic reactors may upgrade reaction rate and hence downgrade the time of residence and minimize consumption of energy per unit of volume being treated. It is accepted that this might be accomplished by depositing the photocatalyst on a high surface product that will particularly absorb the polluted molecules and will fixate them around the photocatalyst [26-28]. Lamentably, active absorption of pollutant lowers the diffusion rate into semiconductor powder, which may thus hinder the activeness of the photocatalyst. Such drawback has assisted various researchers to find any worthy initiative of semiconductor for the operation of some particular pollutants or try to formulate the latest techniques of deposition [29, 30]. The pH of the solution, the support, and the kind of pollutant play a significant role in the accomplishment of photocatalytic

Various efforts have announced utilization of activated carbon (AC) as a platform for the semiconductor but it has been cautioned that effective absorption of pollutants into the absorbent area may hinder diffusion into the catalyst and thus may affect the entire process [31, 32]. AC acts as a brace for the titanium oxide (TiO2) which could give tremendous results over the other mediums [32]. These consist its potential to swiftly absorb pollutants and also its high absorption ability because of its surface area and porosity [31]. As indicated, medium pores AC will make easier the diffusion of pollutants and product discharging from the surface [32, 33]. Furthermore, high ability to absorb fluid of AC may reduce the penetration of ultraviolet lights into small areas, and it may cause confining of pollutants within the pores without getting able to diffuse into the outer surface for further reaction with the OH radical of pollutants [34-36]. Additionally, some types of pollutants, for example, phenol, may go through polymerization on the AC's carbon surface, which causes unchangeable absorption [37, 38]. The general processes involved in semiconductor particles upon bandgap excitation are illustrated in Figure 1.

### 2. Activated Carbon

2.1. Properties of Carbon Materials. Carbon materials technology has made extraordinary progress in current years because of its diversity of physicochemical properties, such as tunable porosity, lightweight, exciting electronic properties, electrical conductivity, chemical and thermal balancing, and the potential to acquire an immense range of morphologies [40–43]. Hence, carbon materials have found a large number of applications in different domains, varying from environmental science [44], absorbent [45], drug delivery [46], catalyst [44], electrode materials [47], stationary phase in the chromatography system [48], energy storage [49], and many others according to its structure, morphology, and chemical properties (Figure 2).

However, for some particular applications, functionalization is essential at controllable size and shape [48, 49]. Nevertheless, the production of such materials usually requires very harsh conditions and has several limits such as extreme temperature of the carbonization process in the first steps up to >800°C and followed by chemical or physical activation to transform carbon materials into activated carbon [43–49]. Furthermore, it is significantly important here to explore economical and sustainable ways to get carbon materials from raw materials other than crude oil or natural gas which leads to a reexploration of this field (Table 1; Figure 3).

2.2. Conventional Conversion of Biomass to Carbon. The use of biomass extracted products or biomass is becoming vitally significant for the enhancement of effective and environmentally friendly technology and together it solves the issues of agricultural and forestry waste use [5–9]. Different synthetic methods, such as carbonization [56], high-voltage-arc electricity [57], laser ablation [58], or hydrothermal carbonization [56], have been disclosed for the preparation of carbonaceous, amorphous, crystalline carbon materials or porous with different sizes, chemical composition, and shape of the biomass feedstocks [56–59].

Moreover, the application of a low-priced pathway to recycle by-products of biomass formed would furthermore represent a way to separate significant amount of  $\mathrm{CO}_2$  and at the same time a material advantage would also be created.

Biomass conversion is very significant, yet, working with this complex biomass feedstock is challenging, and approaches based on the creation of simpler and more six balanced intermediate derivatives, known as platform molecules, have been shown to be active for efficient biomass conversion into chemicals and fuels [55], catalytic routes consisting deoxygenation, and reaction combined with -C-C- coupling processes [57]. Biomass conversion to carbon materials through catalytic approach normally begins with hydrolysis of dehydration and cellulose chains [57, 59] and is divided into monomer's soluble products that come from the hydrolysis of cellulose [58], condensation or polymerization of the soluble products, aromatization of the polymers hence formed, and growth of the nuclei so created by linkage and diffusion of species from the solution to the nucleus surface

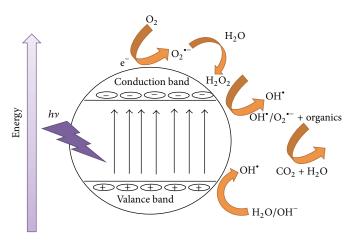


Figure 1: Processes involved in semiconductor particles upon bandgap excitation [39].

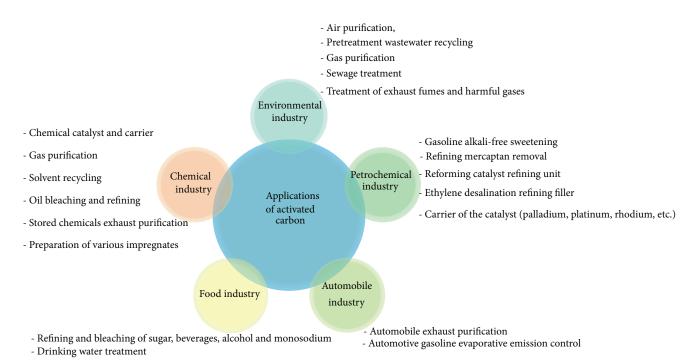


FIGURE 2: General prospective on application of AC.

and finally appearance of short burst of nucleation [56–59] (Figure 4).

- 2.3. Catalytic Conversion of Biomass to Carbon. Green chemistry, now days, is becoming more and more connected with the catalytic process on biomass conversion to carbon materials. Table 2 shows the list of catalysts corresponding to their category and advantages towards the conversion process of cellulose to carbon based materials.
- 2.3.1. Homogeneous Catalyst. Convention of sulfuric acid solution and catalyst cellulose hydrolysis into glucose is a time-consuming and well-formulated process [56–58, 60–65]. Many large scale segments have been developed, but

there are rigid conditions including the treatment and the recycling of the waste sulfuric solutions of acids, which also suffer from the complex separation of products from the solution, the lack of glucose selectivity, toxicity, and high prices which take this process away from the original approach of sustainability [56–58, 60–63].

Thus, the solvents of catalyst as ionic liquids have received enough attention because of their low vapor pressure, stability, and recyclability [62, 62–73]. The kinds of novel green solvent are ionic liquids with relatively less melting point and appear as a crystal in general conditions [64]. Cellulose is balanced via inter- and intramolecular bonds of hydrogen, so that rigid bundles could be created, which makes it difficult to solubilise with common organic solvents and water [16–18, 74–79]. It is important to make solvents for cellulose so as

TABLE 1: Classification of AC materials.

Criteria	Particle size	Properties	Applications	References
Powdered AC	<1.0 mm Diameter: 0.15–0.25 mm	Higher surface area, extraordinary volume per gram, and greater purity	As an additive in vessel, waste water treatment, classifiers, and gravity filter	[50]
Granule AC	0.42 to 0.84 mm	Suitable for many organic chemicals, able to improve taste/odor, and removes chlorine	Vapor and liquid adsorption, water treatment, deodorization, separation of components	[51]
Extruded AC	0.8 to 130 mm	Low pressure drop, high mechanical strength, and low dust content	Gas phase applications	[52]
Beads AC	0.35 to 0.8 mm	Low pressure drop, high mechanical strength, and low dust content	Adsorbent for waste water	[53]
Impregnated AC 0.8 to 200 mm		Porous carbon impregnated with inorganic materials (iodine, silver, cations) and antimicrobial antiseptic	Pollution control and purification of domestic water	[54, 55]

 ${\it Table 2: Catalytic approach on conversion of cellulose to carbon based materials.}$ 

Categories	Types	Advantages	
Homogeneous	Ionic liquid (e.g.: [BMIM]Cl; [EMIM]Cl and [EMIM]BF $_4$ )	<ul> <li>(i) Low melting point</li> <li>(ii) Appears as crystal in normal condition</li> <li>(iii) Acts as template on production of porous carbon structure</li> <li>(iv) Solvent reaction media</li> <li>(v) High yield of carbon production</li> </ul>	
Heterogeneous	Solid acid catalyst (i) Bronsted/Lewis solid acid (e.g.: ZrP; SiO <sub>2</sub> -Al <sub>2</sub> O <sub>3</sub> , WOX/ZrO <sub>2</sub> , c-Al <sub>2</sub> O <sub>3</sub> ,) (ii) HPA (e.g.: H <sub>3</sub> PW <sub>12</sub> O <sub>46</sub> ; H <sub>4</sub> SiW <sub>12</sub> O <sub>40</sub> ; Cs <sub>2.5</sub> H <sub>0.5</sub> PW; Cs <sub>2.5</sub> H <sub>0.5</sub> PW <sub>2</sub> O <sub>40</sub> )	(i) High catalyst selectivity (ii) Good separation process (iii) High promotion on depolymerization of cellulose (iv) Low formation of soluble oligomer (v) Low cellulose self-hydrolysis (vi) Favors direct formation of lactic acid (vii) High stability	
Heterogeneous	Ion exchange resin (e.g.,: Amberlyst; McM-41; HnbM <sub>0</sub> O <sub>6</sub> ; mixed oxides; niobic acid; silica-niobic; niobium phosphate)	(i) High accessibility of saccharides (ii) Satisfactory reaction rate	
Heterogeneous	Zeolite (e.g.: ZSM-5; Beta; Mordenite; Ferrient; FCC; Al-MCM-42; SBA-15; Al-MSU-F; MOR)	<ul><li>(i) High selectivity to adsorb molecules</li><li>(ii) Good separation process</li><li>(iii) Good thermal and hydrothermal stabilities</li><li>(iv) Production of porous carbon</li></ul>	
Heterogeneous	Metal ions (e.g.: Cr <sup>3+</sup> ; Mn <sup>2+</sup> ; Fe <sup>3+</sup> ; Fe <sup>2+</sup> ; Co <sup>2+</sup> )	(i) Good catalyst for ring opening and hydrogenation cellulose (ii) Require short reaction time (iii) High yield of glucose (iv) High turnover amount of catalyst (v) High activity and selectivity (vi) Good recyclability (vii) Easily separable	
Heterogeneous	Metal oxide (e.g.: $HnbM_0O_6$ ; $Al_2O_3$ )	<ul><li>(i) Green process</li><li>(ii) Nonvolatility</li><li>(iii) Highly stable</li><li>(iv) Nontoxic</li><li>(v) Reusability</li><li>(vi) Low cost</li></ul>	



FIGURE 3: Classification of AC materials.

to initiate a system of minimal efficiency to confirm greater yield of carbon materials in the conversion of biomass [17–20]. Ionic liquids have significant roles; it acts both as a soft template to formulate the characterized pore structuring for the development of a hierarchical porous carbon structure and as a catalyst which results in enhanced ionothermal carbon yields [64–66, 71–79]. Thus, it has been demonstrated that in the presence of an acid catalyst, the utilization of ionic liquids, can embrace the efficiency of the hydrolysis of glucose cellulose [66, 67, 80–84].

2.3.2. Heterogeneous Catalyst. The utilization of heterogeneous catalysis along with an immense range of designs is less expensive and extraordinarily stable at high temperature [85–88]. They are believed to upgrade catalyst characteristics and process conditions to get high yields of hydrocarbons while minimizing coke development in the wide range of reaction conditions [86-88]. This method has the benefit of being very economical and mild if we follow some rules of green chemistry since it does not add organic solvent [85, 86] with resulting carbon, which is spherically shaped and the surface is decorated with oxygenated functional groups [86]. This method also involves simple reaction mechanisms for the creation of carbon, which involves the dehydration of carbohydrate into a furan like molecules, mainly 5-(hydroxymethyl)-2-furaldehyde as an initial step and further polymerization and carbonization as the next step [89-92]. This reaction possesses high potential for the catalytic improvement biomass since -C-C- coupling takes place with consistent oxygen removal (the reaction involves the dispatchment of CO<sub>2</sub> and H<sub>2</sub>O) from carboxylic acids [89, 90], and the latter of which are mutual intermediates in the process of biomass conversion [91, 92]. If we compare homogeneous catalysis, carbon materials obtained through a heterogeneous catalytic process consist of an aromatic core containing polyfuran-type units which is surrounded by oxygen rich polar functional groups such as COOH, -OH, -C-, and -O-, which makes the materials more hydroscopic, hydrophilic, and have a lesser degree of graphitisations [93, 94]. These functional surface groups could act as a premier binder and depositor to promote and stabilize the carbon to form nanocables [95-98], a novel carbon-encapsulated core-shell composite, and hybrids. Moreover, heterogeneous catalysts are easy to recover and reuse [96, 97] (Figure 5).

Nevertheless, the effective exploitation of cellulose is a main challenge in heterogeneous catalysis application, since cellulose itself has a tough, mainly crystalline, chemically stable, and water insoluble characteristics, which are induced from the intra/intermolecular hydrogen bonds [99–101]. The most commonly used heterogeneous catalysts for the conversion of biomass are alkali salts [76, 102, 103], metals (Ni, Rh, PT, Ru, Pd, Ir, and Ag) [104–106], metal oxides (CeO<sub>2</sub>, ZrO<sub>3</sub> TiO<sub>2</sub>, and Al<sub>2</sub>O<sub>3</sub>) [107, 108] usually on supports, and metal oxide catalysts at medium temperatures (300 to 425°C) which involves deoxygenation reaction combined with –C–C– coupling processes [109].

(1) Solid Acid Catalyst. Transformation of cellulose to water soluble sugars via solid acid catalysts has received much attention in these years as a solution to remove mineral acids in the formulation of furfural which is easily recovered from the reaction mixture, reused without losing the activity, and minimized the posttreatment cost [75, 110–113]. Solid acid catalysts are basically a Bronsted or Lewis acid [113, 114] and it is of various types such as Vanadyl phosphate, ZrO<sub>2</sub>, zeolites, inorganic oxides, and ion exchange resins.

(1.1) Bronsted or Lewis Solid Acid Catalyst. These series of catalysts include WO<sub>x</sub>/ZrO<sub>2</sub>, C-Al<sub>2</sub>O<sub>3</sub>, Zr-P, SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>, and HY Zeolite which are used for mainly aqueous phase dehydration of xylose [115-119]. Lewis acid sites lower furfural selectivity through catalyzing a side reaction. In the hot water (190°C), solid Lewis acids promoted the cellulose depolymerization and lowered the creation of soluble oligomers and polymers as in connection to the cellulose selfhydrolysis which occurred in the familiar conditions [117-124]. By contrast, in normal conditions, strong Bronsted solid acid has not improved the extent of the cellulose depolymerization but has affected the product distribution [98, 122, 123]. By comparison, Lewis solid acids were not only potential to upgrade the extent of cellulose depolymerization but also favorable for direct formation of lactic acid, which gives high yield, approximately 30% [96, 125]. This comparison represents highly potential ways to optimize the conversion of cellulose and stabilize Bronsted acid catalyst which helps to understand the reaction pathways [111].

This also brought an idea for various researchers to study the effect of Bronsted acidities of water soluble heteropoly acid catalyst, known as HPA [112]. This heterogeneous HPA catalyst which is micellar, clean, economical, facile, and environmentally friendly could be recycled via centrifuge [112, 126]. The exhibited activity for the hydrolysis of polysaccharides comprising disaccharides, starch, and cellulose, is known to be HPA [121, 126]. Tungstophosphoric acids  $(H_3PW_{12}O_{40})$  [119] and tungstosilicious acids  $(H_4SiW_{12}O_{40})$ [109] are the vigorous acids used in the series of HPA catalysts in reaction of hydrocarbons and have been employed, for example, in alkylation, acylation, esterification, dehydration, and isomerization of the ethanol process [127-129]. Salt of acidic cesium H<sub>3</sub>PW<sub>12</sub>O<sub>40</sub> and Cs<sub>2.5</sub>H<sub>0.5</sub>PW<sub>12</sub>O<sub>40</sub> (Cs<sub>2.5</sub>H<sub>0.5</sub>PW) with intense acidity is insoluble in organic solvents and water and has meso- and micropores with relatively high surface area (130 m<sup>2</sup>gk) [119]. This Cs<sub>2.5</sub>H<sub>0.5</sub>PW

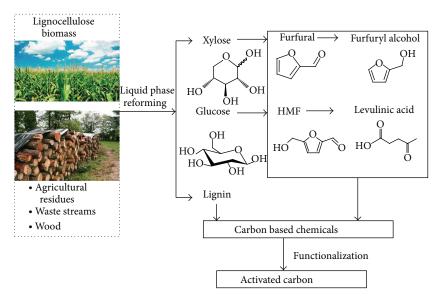


FIGURE 4: The route of AC derived from biomass.

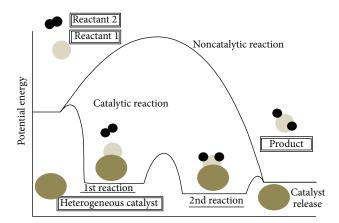


FIGURE 5: Heterogeneous catalytic routes in chemical reaction.

has been described as a solid acid catalyst with prudent and environment friendly specifications and is recommendable in industrial process like hydration of olefins or ester and liquid phase dehydrated alcohol [130, 131]. Till now, cellulose hydrolysis into saccharides employing a range of HPA catalyst has been researched for capable applications, in contrast to conventional acid-catalyzed reaction, whereas the yield of glucose is less [130-132]. It is because of the insolubility of cellulose in any of the solvents and problems of solid to solid mass transport [77, 133]. In a few cases, the acidity of mineral acid is lower than that of HPA. For example,  $(H_0)$  of H<sub>5</sub>BW<sub>12</sub>O<sub>40</sub> (0.7 M at 100°C), the Hammet acidity function, is -2.1 lower than that of HCl (0.7 M) and H<sub>2</sub>SO<sub>4</sub>. Hence, HPAs have been anticipated to be reusable and active catalyst for the hydrolysis. Along with that theory, amorphous carbon bearing SO<sub>3</sub>H, COOH, and OH groups had been described to show hydrolysis activity (10% of glucose yield) for 3 hours at 100°C. Meantime, Yabushita et al. demonstrated that the cellobiose hydrolysis and cellulose in water assisted by HPAs

such as Sn<sub>0.75</sub>PW<sub>12</sub>O<sub>40</sub>, H<sub>3</sub>PW<sub>12</sub>O<sub>40</sub>, and H<sub>4</sub>SiW<sub>12</sub>O<sub>40</sub> record total yields of decreasing sugar and it was around 40% at 200°C for 16 hours of reaction [134]. It has been found by Hara that  $H_5BW_{12}O_{40}$  exhibited a good performance for the transformation of crystalline cellulose to give glucose 77% yield and various types of HPAs like H<sub>3</sub>PW<sub>12</sub>O<sub>40</sub> (glucose yields 8%) and H<sub>4</sub>SiW<sub>12</sub>O<sub>40</sub> (37%) which are less active than H<sub>5</sub>BW<sub>12</sub>O<sub>40</sub> [135]. Other research groups also administered the screening of counter cations of  $\mathrm{PW}_{12}\mathrm{O}_{40}$  for the cellobiose hydrolysis and they found that there was a volcano-type correlation between Lewis acidities [136-138] and TOFs for glucose formation. They also observed that the acidity and the role of decreasing CrI were the significant factors for the catalytic activity of HPAs [139]. In reality, the H<sub>0</sub> role was  $H_3PW_{12}O_{40} < H_4SiW_{12}O_{40} < H_5BW_{12}O_{40}$  in a similar order of catalytic activity. The H<sub>0</sub> roles were corresponding to the concentration of acids and the number of anions. Hence, HPAs involving highly negatively charged ions are desirable. The anions were dissociated from hydrogen bonding between cellulose molecules to lower the CrI. Moreover, HPAs protons have also exhausted the bonds of hydrogen from cellulose and greater concentration of protons was effective in this role [135, 140]. Therefore, a strong catalyst for the cellulose hydrolysis is  $H_5BW_{12}O_{40}$ , which was recovered through the extraction and it was recycled for around 10 times [141].

(2) Ion Exchange Resins. Sulfonic acid functionalities of ion exchange resin with solid acid catalyst and sulfonic acid functioned materials resulted in high yield at 63% and 76% in pure dimethyl sulfoxide (DMSO) solvent, correspondingly [142, 143]. Solid acids such as amorphous carbon materials consisting of  $SO_3H$  groups layered transition (HNbMoO<sub>6</sub>) metal oxide and resin sulfated have been tested for the cellulose hydrolysis, but the yield of glucose is still comparatively less [144, 145]. It has been hypothesized that side reactions were abolished employing aprotic or organic solvents [146]. Vigorous acidic resins (Amberlite IR-200 and IR-120), niobic

acid, mixed oxides (silica-zirconia and silica-alumina), silica-niobia, and niobium phosphate created a strong acidity which was protonic and accessibility of simple saccharides to the most effective sites on the surface catalyst allows satisfactory reaction rates to be obtained [147–150].

Sulfonic resins which are acidic are represented in different literatures of active catalyst system for the hydrolysis of starch, cellulose, and disaccharides [151-153]. Generally, the rigid conditions in terms of temperature (>120°C temperature in the water and in critical condition) and high concentration of saccharide (>100 g) were employed to push the catalytic actions of solid acid towards the achievement of high transformations [154–156]. Sulfonated activated-carbon could transform cellulose of amorphous into glucose with 41% of yield for 24 hours at a temperature of 200°C [157]. Such things have been founded by Sun and Zhang demonstrated that p-toluenesulfonic acid can catalyze cellulose hydrolysis in ionic [EMIM][Cl] system, giving THF yields of 28% and 13% and a yield of mono- and disaccharides 10% and 3%, respectively [158]. Natural bamboo can be converted by a sulfonated biomass char with cotton and starch around 20% yield of glucose under microwave assistance [159-161]. The process of hydrolysis consisting of starch and cellulose was obtained via layered transition metal oxide, despite the fact that the yield from glucose was less than cellulose. Sulfonated carbon with a mesoporous like structure was used by the groups of Vyver for cellulose hydrolysis getting the yield of glucose around 75%, which is considered to be the highest recorded yield via solid-acid catalyst [162, 163]. Similarly, some interest has been shown in cellulose depolymerization in water because solid supported acid catalysis was used [164-166].

Current reports describe the cellulose hydrolysis by solid catalysts such as layered transition metal oxides, Amberlyst resin acid modified amorphous carbon, and sulfonated silica or carbon nanocomposites [167-169]. Also, the depolymerization of cellulose was also considered under catalysis of both FeCl<sub>3</sub> and Nafion supported on amorphous silica to be tested on a continuous flow reactor, given the residual that unreacted cellulose can be easily eliminated from the system [75, 170]. For the conversion of glucose, these surface species which are acidic were quite active [169]. Certainly, we are aware that the hydrolysis rate of cellulose depends on the acid strength. As an expansion on the previous reports on the usage of Nafion as a solid assisted acid catalyst for the transformation of cellulose into glucose and levulinic acid, many researchers have incorporated the reaction with alkali metal salts to embrace the reaction's yield [171, 172]. Namchot et al. and Klamrassamee et al. have recently formulated carbonbased solid acid with immense density of Bronsted acid sites (SO<sub>3</sub>H and COOH) to pyrolytically carbonize sugar, such as cellulose, sucrose, or glucose, and subsequently sulfonate the prepared carbons [173, 174]. Interestingly, these sulfonated carbon materials are very strong for the microcrystalline cellulose hydrolysis to produce water soluble saccharides with low reaction temperature (100°C) with the conventional and strong Bronsted acid catalyst such as H-mordenite, niobic acid, and others. The particular surface area of the sulfonated carbon was around  $2 \,\mathrm{m}^2 \,\mathrm{g}^{-1}$  but the soluble saccharides

yield reached nearly 70%. Catalytic performance of soluble saccharides was applied to its intrinsic ability to adsorb  $\beta$ -1,4-glucan, which is not absorbed on the other solid acids. Thus, it can be concluded that heterogeneous catalysis are more active and environmentally benign, mainly because of a hasty product separation and also catalysts recovery. Jule and Schoonover described that acid resins with considerably big pores could actively depolymerize cellulose in ionic liquid, but the main products were cellooligomers which failed to be dissolved in water [175].

(3) Zeolite Catalyst. The workhorse of the petroleum industry is zeolite catalyst that efficiently converts petroleum based feedstocks into the targeted chemicals and fuels and chemicals [176–178]. Crystalline microporous solids are an important part of zeolites because of their widespread application in absorption, separation, and catalysis [178]. Its importance stems from its unique structures of pores, which makes it highly particular to absorb molecules for separation reasons or towards product molecules in catalysis [175, 179, 180]. Furthermore, during heterogenous catalytic reaction, zeolite shows good hydrothermal and thermal stabilities. However, from the last twenty years, there have been many studies that focused on the catalytic transformation of biomass and its derived feedstocks with a variety of zeolite catalysts, including Ferrient, FCC, Al-MCM-41, ZSM-5, beta zeolite, Y zeolite, SSZ-20, IM-5, TNU-9, MOR mordenite, SBA-15, Al-MSU-F FER, ZSM-23, MCM-22, and MFI [181-186]. ZSM-11 and ZSM-5 among these series had the lowest amount of coke and the highest yield of aromatic because of its pore space and steric effects [181]. Manufacturization of ordered porous carbon material has been obtained previously through replication of ordered zeolite inorganic and nanocasting [187, 188]. Inside zeolite many reactions have occurred which include decarbonylation, dehydration, isomerisation, and decarboxylation and with that removing oxygen as carbon dioxide, water, and carbon monoxide and conversion of carbon and hydrogen into aromatics and olefins [181-188]. In these catalysts of zeolite, ZSM-5 has exhibited the highest olefin and aromatic yields from biomass of lignocellulosic. With a pore size of around 5.5 to 5.6 A, ZSM-5 has a threedimensional pore system. This small size of pore, internal volume, and internal structure has made it problematic for greater aromatic coke antecedents to form inside the pores [189]. It has been demonstrated by Zapata et al. that tin consisting zeolite is a highly efficient catalyst for the isomerisation of glucose in water [190]. Nevertheless, other studies have mentioned that, at low conversions in aqueous environments, faujasite and mordenite both resulted in great furfural selectivities up to 80% and 90% in 200°C as the selectivity lowers with high conversion and the final yield was relative [191, 192].

(4) Metal Ion Catalyst. Furfural and HMF are the two main and significant intermediates which are derived from biomass. They were directly manufactured from the microcrystalline cellulose hydrolysis with metal ions in ionic liquids such as Fe<sup>3+</sup>, Fe<sup>2+</sup>, Cr<sup>3+</sup>, Co<sup>2+</sup>, and Mn<sup>2+</sup> as a catalyst under mild conditions [193–195]. Metal ions as an acidic support are

a nice catalyst for hydrogenation and ring opening of cellulose polymers.

Many reports are there on carbon production through cellulose hydrolysis at a moderate temperature up to 250 and 300°C metal catalyst in a very less reaction time to control the deep exploitation of the formed glucose [196]. Some researchers even used Ru that is a ruthenium catalyst to enhance the transformation of oligosaccharides and increased glucose yield (almost 30%) with the TON that is the turnover number of the catalyst was immense (145 based on bulk Ru) in contrast to those of the sulfonated catalysts of carbon [197, 198]. Wang et al. found that the glucose yield was increased around 31% by upgrading the Ru loading to 10 wt% along with the recyclable number of catalyst up to five times without losing the activity or Ru leaching [199]. Various reports on the efficiency of the hydrolyzate of cellulose formulated by Ru catalysis as a source of carbon for the bacterial PHA production found that under the aqueous solution the reaction which occurs is desirable for delivering microbes which thereupon make it easily disconnected from yielded sugar via facilitating the race of the catalyst and filtration [200-203]. Such reports have accepted that Ru species were in fact in an oxidized state and acted as the real and effective site for the oligosaccharides hydrolysis. Prior, Ru/Co<sub>3</sub>O<sub>4</sub> catalyst has showed considerate selectivity and activity and good recyclability in the biomass conversion to carbon [204].

Researchers have even exclaimed that iron oxide nanoparticles and iron ions could adequately catalyze the hydrothermal carbonization of rice grains and starch beneath mild conditions ( $<200^{\circ}$ C) and had a powerful influence on the creation of nanomaterials of carbon with different shapes [205, 206]. Catalysts of magnetite with sulfonic groups like a mesoporous silica composite, Fe<sub>3</sub>O<sub>4</sub>, SBA-15 treated by sulfonated CoFe<sub>2</sub>O<sub>4</sub> embedded silica, and sulfuric acid were also employed for the cellulose hydrolysis [207, 208]. After the catalytic reactions they were easily separable via magnet. Fe<sub>3</sub>O<sub>4</sub>-SBA-SO<sub>3</sub>H catalyst managed glucose in 26% yield from microcrystalline cellulose at a temperature of 200°C for 3 hours, though levulinic acid becomes an important product giving 42% yield by delaying the reaction time up to 12 hours [209].

(5) Metal Oxide Catalyst. In many catalytic processes metal oxides play an important role [210, 211]. For example, metal oxide nanostructures are important components commercially available for synthesis of methanol [212-215]. There are many more benefits of these catalysts which make them efficient candidates for green processes. The characteristics of such catalysts are stable, nontoxic, low cost, availability, nonvolatility, and reusability [214]. Several studies have observed the processes of decomposition connected with formic acid on nanometal oxide surface, methanol, and formaldehyde [210-215]. It is strongly expected that metal oxide nanostructures would have a better catalytic activity in developing the conversion of cellulose to the value added products in hydrothermal media with an efficient separation from the reaction matrix in regard to the increased surface area of the nanomaterials [214]. Layered transition metal

oxides containing niobium were found to be specifically active in the hydrolysis of disaccharide, suggesting the importance to investigate niobium containing catalyst as an energy inefficient factor for the conversion of biomass [213–215].

The employment of transition metal oxide like  $\rm HNbMoO_6$  was also reported as an efficient solid catalyst to generate glucose from cellulose. Similarly, in the presence of noble metal consisting catalyst, for example,  $\rm Pt/Al_2O_3$ , Felica et al. have found the creation of sugar alcohols in yield up to 31% from cellulose in hydrothermal conditions. On the other hand, the researchers proclaimed that Pt free catalysts have generated only poor glucose hexitols amount [216].

### 3. Activated Carbon in Photocatalyst System

Activated carbon (AC), a carbonaceous material structured on plant-based material, is a porous, amorphous solid carbon [31, 32]. Well-developed porous surface, high pore volume, and extended surface area make AC the most commonly used technique for controlling pollution [35]. Well-developed pores over the surface are one of the main uses of AC as the photocatalyst. Nontoxic, chemical stability and being economical are the main reasons that in the past decades the heterogeneous technology has attracted the attention of many researchers [217, 218]. As the organic pollutants can be mineralized into neutral by-products such as H<sub>2</sub>O, CO<sub>2</sub>, and mineral acids as one of the main properties of heterogeneous AC, photocatalysis methods include the destruction of the wider category of organic compounds. Promoting solar radiation and working on the low temperature, eventually saving a lot of energy, make it very economical [35-39]. One of the advantages of the AC includes the regeneration of spending absorbent and demolishing of absorbed organic material on the site converting the loss of absorbents to burn them concluding that thermal regeneration is efficient [218, 219]. AC being the strong light absorbing compound has been successfully used as photoactive species [38]. Determining the band gap of the AC (band gap less than 4 eV) resulted in a semiconductor and therefore, a photoelectric material in the presence of ultraviolet radiations [218]. Recent reports suggest the abnormal reaction towards the aqueous environment by directing ultraviolet irradiation of the sample in the presence of the AC provided that no other photoactive materials are present [220] showing that the AC improved the photooxidation of phenol, beyond the degradation of photolysis in comparison to bare or unmoved TiO<sub>2</sub> [220, 221]. To find the difference whether this reaction is only shown by the AC or also by other carbon compounds, some researchers have worked on different porous AC materials obtained from different sites, procedures, and reactions and examined their behavior to the exposed ultraviolet radiations [222, 223]. The final solution we want to reach is that to remove any vagueness in the aqueous medium, ultraviolet radiations and absence of semiconductor AC are able to demolish the organic materials in the respective conditions [220]. Regardless of the type, AC acts as a catalyst during the removal of diatrizoate. Gamma radiations based AC is more efficient in a way that it has a higher proportion of C atoms

and contains sp<sup>2</sup> hybridized [224]. With more than 53% of synergistic effect in diatrizoate in the first minute of reaction, commercial carbon is produced from the ultraviolet/AC system [222, 223]. Providing more oxygen, the synergistic act of AC is boosted up. The reutilized AC is quite similar to the original C; presence of O in the sample increases the rate of removing diatrizoate by the ultraviolet/AC system, but the ultraviolet inclusion of system results in some modifications in AC chemically. There is a very vague relationship between the textual properties and the synergistic contribution of the AC. Gamma radiation involvement with AC reduces the band gap which results in a more efficient removal of organic compounds [225, 226]. One of the actions proposed include that AC is the photocatalyst which will promote the electrons in the valence band to the conducting band, resulting in enhanced generation of OH free compounds present in the polluting medium.

3.1. Activated Carbon Surface Properties in Photocatalyst System. For commercial use, textures and surface properties can alter, depending on the crude material used, activation conditions and carbonization procedure which may potentially result in well-defined photocatalytic performance [39]. Many researchers and authors reported many surface and textual features of AC [227, 228]. It is clear that basic AC in the presence of low oxygen in this medium has much more potential than the phenol adsorption [225]. An increase in the physical absorption and surface polymerization of phenols can be done by ensuring the deprivation of the acidic categories on the surface of the AC. Irreversible absorption and catalyzing the oxidative coupling of phenolic compounds can be enhanced by the O<sub>2</sub> containing basic categories over the face of AC [221]. Boosting up the interactions between  $\pi$ - $\pi$  electrons in AC and phenols is due to the graphene layers of an activated carbon which increases  $\pi$  electron density [222]. In carbon of basic nature, phenol is considered to be higher regardless of textual propertie and absorption capacity [228]. Retention of phenol is supposed to become less in the acidic carbon as compared to the basic carbon; in fact there is no clear relationship between the two.

On of AC limitations is that the waste organic materials are not really destroyed but are transformed from one phase to another and in result the used AC is transformed into a dangerous product [226]. So, regenerating AC becomes necessary for its reusability, which makes an economical process. The AC is carrying some limitations like its adsorption capacity, which is a function of inert concentration that results in low quality products [229]. Already used AC is to be disposed of as it is hazardous material or is regenerated to be used again. Thermal regeneration contains many disadvantages because of its off-site regeneration that hardly converts the pollutant from one phase to another. This may result due to depletion of carbon and may cause damage to the structure of activated carbon.

The process, which could produce high yield efficiencies, could be a chemical regeneration of spent AC but it has some drawbacks regarding chemical consumption nondestructive pollutant elimination and creation of unimportant steams

of waste [230, 231]. However, techniques like ultrasonic regeneration, microwave regeneration, and electrochemical regeneration are also being proposed as alternative techniques for the process of chemical regeneration of spent AC [230]. Chemical consumption, having increased footprints of carbon or having expensive facility requirement, is through the bench scale studies in which it has been proved to be effective but in industrial applications it has a limited appeal. Due to the limitations of the present technique, there is a need to develop another technique, which is more economical and environmentally friendly. Thus, to make one hybrid system, there is a need to merge semiconductor with AC [230, 231]. Organic pollutants, issues of destruction, and other hazardous problems are expected to arise from the oxidation semiconductor element. The reason lies in the generation of radical species like O' and OH' from the catalyst particles of the semiconductor, which causes oxidation of such species [230]. Another technique that has been studied for AC is the combination of heterogeneous photocatalysis and the Fenton reaction with the catalytic process, which is an oxidation based process.

Studies on this spent AC carried out previously were mostly about granular AC. As an example, it was demonstrated that there was an improved efficiency in herbicide removal from the water when granular AC adsorption photocatalyst hybrid system was used in comparison with a photocatalysis system [227, 228]. Similarly, it was also reported that the combination of photocatalytic and rotating adsorbent showed better efficiency in removing formaldehyde in comparison with adsorption [232].

3.2. Transition Metal Oxide Hybrids: AC in Photocatalysis System. In future, it anticipated that the coming photocatalyst generation would have improved internal efficiency regarding separation and also would be placed in contact with molecules of external pollutants [222–224]. So to improve the photocatalytic efficiency and separating catalyst from aqueous solution, the hybrid photocatalyst was designed by not moving the metal oxides having a large surface area, to condense pollutants which are diluted [233, 234]. In environmental purification field, heterogeneous photocatalysis with metal oxides of semiconductor has been applied as an efficient method [235].

By impregnation and adsorption along with various methods, metal oxides are expected to be impregnated into the surface of carbon; the same is applied for complex experimental procedures and process, which operate at high temperature. Considering ideal conditions, a photocatalyst should be inexpensive, highly proactive, nontoxic, and stable [233]. One more criterion that plays a role in the degradation of organic compounds is the potential of the redox residing in the band gap of the semiconductor. There are many semiconductors with the band gap energies ample to catalyze several chemical reactions, which include WO<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, SrTiO<sub>3</sub>, ZnS, TiO<sub>2</sub>, and ZnO [233–236]. The metal sulfate group with insufficient stability in the process of catalysis is kids, PbS, or CdS. This compound undergoes photoanodic corrosion readily and is toxic. For example, Fe<sub>3</sub>O<sub>4</sub> undergoes

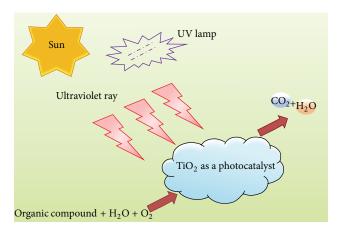


FIGURE 6: Basic principle on TiO<sub>2</sub> photocatalyst.

photoanodic corrosion readily so they are not suitable [233]. ZnO (3.2 eV) is unstable with Zn  $(OH)_2$  in water; also it has band gap similar to anatase, so it results in the deactivation of the catalyst [228].

#### 3.2.1. TiO<sub>2</sub>: AC Photocatalyst System

(1) TiO<sub>2</sub> Photocatalyst System. The most promising semiconductor for photocatalytic destruction organic pollutants is titanium dioxide (TiO<sub>2</sub>) [229]. It provides the most excellent agreement in aqueous media between stability and catalytic performance. Since it is nontoxic, cheap, biocompatible, and stable in sunlight, so it is of immense importance and that is why it is also considered usable in cleaning environmental operations [230, 231]. Consequently, the electron pairs and positive holes are created at the surface of TiO2. Once it has been irradiated with the UV light of the wavelength of 380 nm, TiO<sub>2</sub> would form reactive oxidants, such as OH radicals, hydrogen peroxide, superoxide anions, and other reactive species of oxygen and reactions that are reductive, to contribute in the organic compound decomposes which are adsorbed on the surface of TiO<sub>2</sub> [235–237] (Figure 6). The highest photocatalytic detoxification of TiO<sub>2</sub> is anatase phase. Deep studies have shown that photodegeneration of components like herbicides, phenols, dyes, pesticides, surfactants, and organic components (e.g. salicylic acid and sulfosalicylic acid) has been possessed by TiO<sub>2</sub> that is present in water wastes [238, 239].

TiO<sub>2</sub> powders contributed to some drawbacks in separating phase in photocatalysis, with the purpose of its emission in the atmosphere because of their small particle size and recovery, the loss of photocatalyst if the separation is not promising, the need of fluidization of the powder in gaseous phase with cost and energy, and the scaling difficulties involved [237, 240]. Also, since radiation from the light compromises 47% of visible light, 48% infrared radiation, and 5% UV light, so TiO<sub>2</sub> acts as a benchmark of UV photocatalysis that it goes deactivated under visible light because of its wide band gap [241]. Moving on, the holes and photogenerated electrons present in an excited state play a

vital role in the degeneration of pollutant and are unstable and without any effort they can recombine, they lead to low order efficiency which results in photocatalysis activities [231]. It is clear that the use of high potential solar photocatalysis cannot be made by  ${\rm TiO}_2$ .

(2) Activated Carbon Supported TiO<sub>2</sub> in Photocatalyst System. For easy manipulation in a process of total photocatalytic operation and quick decomposition of organic pollutants, it might speed up the process to load photocatalysis to suitable adsorbents to increase the strength of pollutants around the photocatalysis system [237, 238]. Therefore, researchers had made attempts to support TiO<sub>2</sub> on different matrixes as silica gel, clay, carbon materials, alumina, and zeolites which can be nanotubes, carbon microspheres, carbon black, carbon nanofibres, and AC [242, 243]. TiO<sub>2</sub> particles are hydrophilic when exposed to direct UV light whereas organic pollutants are hydrophobic [229]. The use of AC as a reference will provide help to molecules of pollutant closer to the active site of TiO<sub>2</sub> for a quick and effective photodegeneration process (Figure 7). The AC in comparison to organic pollutants that are approaching, in which secondary degradation takes place intermediately in situ, can generate new adsorption centers.

In gas and water remediation for support purposes, AC is used widely because of its high porosity, good adsorption, supported  $\mathrm{TiO}_2$ , and low cost that has marked the effects on disappearance of pollutants kinetics, with each pollutant being more quickly degenerated [244]. For example, the  $\mathrm{TiO}_2$  surface becomes static over glass surface; it has the benefits of high photodegradation productivity. The major limitation is the adhesion force in  $\mathrm{TiO}_2$  membrane and glass is poor, so  $\mathrm{TiO}_2$  is easy to decrease, which causes the decrease of the photodegradation productivity [231].

Hybrid of TiO<sub>2</sub> with AC support, as a sensitizer which is able to absorb light, was proven to be the best approach to developed photo-responding photocatalyst with great activity. The formation of heterojunction between TiO2 with a small band gap and negatively charged AC may result in the inoculation of conducting band electrons from AC to TiO<sub>2</sub> and it is very useful for electrons and hole division [245, 246]. At the same time, the immobilization of TiO<sub>2</sub> onto the AC support can compromise for the loss of photocatalytic ability of TiO<sub>2</sub> because of the difficulty to effectively disperse in water for complete interaction with pollutants. So, many researchers reported that there is an optimum used amount of TiO<sub>2</sub> and AC pore formation for attaining the higher photocatalytic productivity than TiO<sub>2</sub> [245-247]. The 3D relation between the particles of AC, TiO<sub>2</sub> photocatalyst small particles, and the molecules of organic toxic is reported representationally in the absence of light and in the presence of ultraviolet enlightenment [245, 248]. The organic pollutants are supposed to be small to be adsorbed in microspores. In most of the AC amounts, a large group of micropores exists over the broad surface of the substantial pores, mesopores, and macropores; a large amount of toxic particles is well balanced due to physical adsorption [247-250]. Instead, only a little number of pollutant particles are adsorbed on the surface of TiO<sub>2</sub>. By depositing TiO<sub>2</sub> molecules onto AC particles, some mesopores and micropores become closer to

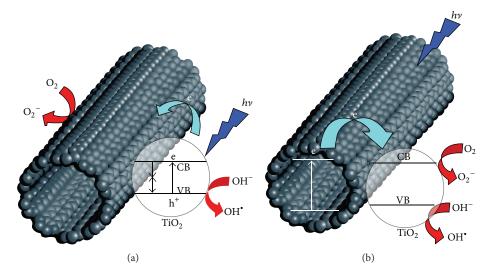


FIGURE 7: General mechanism of adsorption and photodegradation of TiO<sub>2</sub> supported CNTs photocatalyst [243].

their openings and this causes a marked lessening in the uncovered surface area [245]. The ultraviolet enlightenment over such  ${\rm TiO_2}$  used AC molecules; oxidative OH free radicals take birth on the  ${\rm TiO_2}$  and can destroy toxic molecules by oxidation (Figure 8).

Though these radicals will take birth over the surface of  ${\rm TiO_2}$  molecules and they are in the access of ultraviolet radiations and do not locate on the surface in the same radicals, they cannot diffuse through long distances and are limited to an area close to the active centers in  ${\rm TiO_2}$  [231–235, 251] (Figure 9). Adsorbed pollutant particles are photocatalytically demoted; they have to move along the surface of AC and  ${\rm TiO_2}$  molecules with the surface are not necessarily interacted to ultraviolet radiations [249].

The moving force of migration is actually the saturated gradient between organic toxic particles over the enlightenment of  $\mathrm{TiO}_2$  interface and on the other side some are over the surface of changing pore sizes of the AC molecules. The molecules diffused inside micropores of AC migrate with greater retaliation towards  $\mathrm{TiO}_2$  molecules resided on the interface of the AC particles. Thus, highly microporous AC particles are not usually advantageous for the  $\mathrm{TiO}_2$ : AC to have preferred the photocatalytic response [247, 248]. The effect of the substrate pore skeleton has been observed using AC surface area (770–1150  $\mathrm{m}^2\,\mathrm{g}^{-1}$ ) and a dip-hydrothermal process of photocatalyst preparation. Improved photocatalytic demoting of methyl orange (MO) has been attained with  $\mathrm{TiO}_2$ : AC than with a simple mixture of  $\mathrm{TiO}_2$  and AC [255] (Figure 10) and (Figure 11).

This got into the design of highly effective  ${\rm TiO}_2$ : AC hybrid heterojunction photocatalyst; also the demand of commonsensible crosscheck capacity of band potentials among hybrid modules, the spatially and flat accessible transmission of holes and electron at the exposed surface, and the hole and electron movement of the hybrid system are important to enhance the photocatalytic action. It has been reported that the surface chemistry and map of AC revealed major effect on the collection of  ${\rm TiO}_2$  particles

and photocatalytic deprivation of 4-chlorophenol [256, 257]. Adding to this, some researchers have constructed  ${\rm TiO}_2$  with AC microspheres to both maintain spreading and speeding up separation due to the AC microsphere that can be balanced with airy bubbles and it can be speedily settled in the reactor base with the help of some gravity due to the air bubbles [241]. In addition, some authors have fabricated  ${\rm TiO}_2$  with AC microspheres to both sustain spreading and acceleration division, because AC microsphere can be balanced with air bubbles and be able to speedily settle on top of the reactor base by gravity due to air bubbly. To overcome this shortcoming, the use of cobinder upholds the expansion of another method to obtain a fresh form of the  ${\rm TiO}_2$ : AC photocatalyst for such a high action and better division performance.

Jamil et al. found that TiO<sub>2</sub> supported with AC sample heated at almost 500°C, which mainly consisted of rutile phase, showed the greatest photoactivity for deprivation and elimination of methyl orange from aqueous medium [255]. Therefore, most samples which were cooked at higher temperature were very detrimental to photoactivity. Also, using different types of AC revealed the connected effects between TiO<sub>2</sub> and AC during the 4-chlorophenol photodegradation and found a clear enhancement of photoactivity due to an increase in electron density of the AC support [258, 259]. It is researched that attendance of AC in interaction by TiO<sub>2</sub> is beneficial due to its burly adsorption capability. In the same way it advances the relocation rate of the interfacial change and lessens the rejoining rate of the holes and electrons [260, 261]. This synergistic effect of the interaction of AC and TiO<sub>2</sub> has been previously been stated for deprivation of some organic compounds in the photocatalytic process. It has been credited to a common contact between the different solid phases, in which AC acts as an efficient adsorption agent for the organic pollutants [257] (Figure 12).

The organic more efficiently moved to the  ${\rm TiO_2}$  surface, where it is immediately photocatalytically degraded by a mass movement of the photoactivated  ${\rm TiO_2}$  [258]. Thus, the organic burning rate observed on  ${\rm TiO_2}$ : AC is like heading

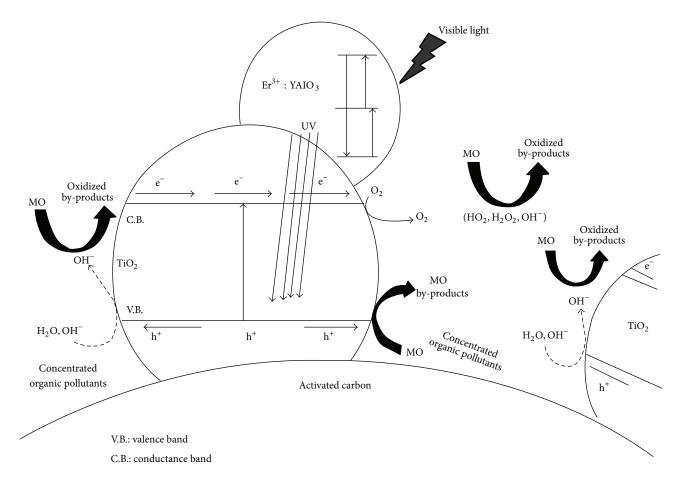


FIGURE 8: Mechanism on photodegradation of MO with TiO<sub>2</sub>/AC under visible light irradiation [245].

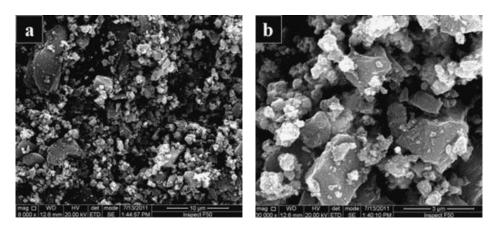


FIGURE 9: SEM images of  $TiO_2/AC$  powder. (a) 10  $\mu$ m and (b) 3  $\mu$ m [250].

both with surface diffusion toxin particles with the photocatalytic process rate; because adsorption occurs gradually, the variation in relative pollutant proportion with irradiation time depends on both adsorption and photodecomposition, mainly at the start of ultraviolet treatment. It is stated that the variance of the proportion of phenol (as model of organic pollutants) remaining in the solution by ultraviolet treatment time is compared for TiO<sub>2</sub>: AC, which were prepared by

hydrolysis of tetraisopropyl orthotitanate and heat treatment at 650 to 900°C. For example, we noted that, in the duration of first 1 hour, adsorption of pH occurs in native AC and after 3 hours, in the presence of UV irradiations saturation achieved [261]. Adsorption as well as the photodecomposition of pH takes place simultaneously, but on the other side the former is supposed to become the dominate method in the beginning, being similarly latter in the next stage [221, 222]. The noted

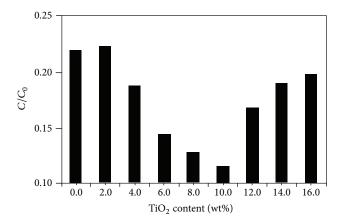


FIGURE 10: Effect on  $TiO_2$ /AC photocatalyst at different contents of  $TiO_2$  on MO degradation [250].

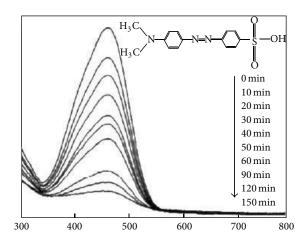


FIGURE 11: UV-Vis spectra of MO degradation with  ${\rm TiO_2/AC}$  under visible light [245].

trend was based on two linear processes; the change of one process to the other takes place approximately in 1 hour of irradiation.

(3) Synthesis of TiO<sub>2</sub>: AC Photocatalyst. A number of methods are available for the composition of TiO<sub>2</sub>: AC catalyst, such as precipitation, chemical vapor deposition (CVD), hydrothermal, aerosol pyrolysis, hydrolysis, dip coating, and sol-gel [235, 262, 263]. However, selection process which is used for the selection of a suitable impregnation method depends only on the support used in it and the pollutant which is degraded [262]. It is clear that those physicochemical properties of TiO<sub>2</sub>: AC catalyst have a heavy impact on the structure of the supported catalyst and they depend basically on the preparation method used, for example, thermal treatments. The main advantages of using physical methods are simple, lowcost and the use of commercially is present for photocatalyst with the wanted functionalities. By using the common wet methods, TiO<sub>2</sub>: AC hybrid was also being prepared, the mismatches in the level of lattice among two hybrid components lessen the required efficiency of separation and transmission

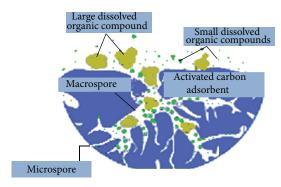


FIGURE 12: Adsorption mechanism of AC towards organic compounds.

of photogenerated carriers (electron and hole) [264, 265]. Adherence of AC surface to TiO<sub>2</sub> particles appears significant for increment of photocatalytic action and as well for useful applications of hybrid system. To increase the anchorage of TiO<sub>2</sub> on AC, the wet process of synthesis is warranted [264]. This is only because the physical stable TiO<sub>2</sub>: AC hybrid is in disagreement with hydrodynamic shearing method, surface chemistry plus AC pore structure can have a sufficient impact on spreading of TiO<sub>2</sub> over the synthesis; it results in the different photocatalytic presentations of  $TiO_2$ : AC. Nevertheless, TiO<sub>2</sub> photocatalyst usually has low precise surface region. Due to the crystalline particle development to happens in heat operation like that. To prevent TiO<sub>2</sub> sintering, or else to make it precise surface region better, AC can be examined like a better help for photocatalysis material [266]. They found that the TiO<sub>2</sub> particles calculated at 450°C can collect and go through the great pores of the activated carbon substrate, including a very burly contact among carbon matrix and TiO2. The contact between them leads to visible synergy to increase photocatalytic capacity for the degradation of the chromotrope 2R [267]. The prepared nanocrystal anatase TiO<sub>2</sub> particles are installed on activated carbon at a fewer temperature with the hydrolysis of the titanium but oxide in the acidic aqueous solution [268]. It is noticed that phenol toxin was absorbed by AC and after that drifted constantly over TiO<sub>2</sub>, which consequently accelerate photocatalytic oxidation. On the other side, for the chemical vapor, deposition method is used for nanosized TiO<sub>2</sub> particles which were exposed to stick to activated carbon and tetra-butyl titanate and to offer large activities on the behalf of the photodecomposition of methylene orange in the water. Adding of water in titanium tetraisopropoxide vapors was described to make possible CVD method at a higher deposition rate and lower temperature [269]. Introduction of H<sub>2</sub>O vapors for the duration of CVD method and adsorption on the AC in prior were announced to be critical to get hold of anatase type TiO2 nanoparticles at AC surface [270]. Investigations have also made it clear that the HNO<sub>3</sub> treatment results in more orderly TiO<sub>2</sub> loading by CVD, in comparison with other oxidation treatment. TiO<sub>2</sub>: AC ratio has been formed as a result of using dropping the support in solution produce with the alkoxide hydrolysis as well [269, 270] and subjected to heat operation at 300 to 500°C. In an alternative process, it can be produced by adding TiCl<sub>4</sub> drop by drop in aqueous suspensions of AC, come after by heat operation at  $500^{\circ}$ C in  $N_2$  atmosphere [271]. Load the TiO<sub>2</sub> powder exactly over AC has been achieved as a result of combining TiO<sub>2</sub> in AC aqueous suspension with stirring. On the other side, loading over AC filter was produced as a result of gluing granular AC over the glass cloth and it was formed by the water suspension of five mass % TiO<sub>2</sub> and the conclusion TiO<sub>2</sub>: AC particles inside CCl<sub>4</sub> solution of pitch, come after by heat operation at 750°C [263]. In resultant hybrid system, TiO<sub>2</sub> particles over AC were expected to be layered with carbon, composed of the pitch at the stage of the heat operation; it may function as a mean repair TiO<sub>2</sub> particle over the AC surfaces. TiO<sub>2</sub> particles also were able to load over AC by spray-desiccation procedure, with a little modification in the pore structure of the AC [272, 273]. In the other study, loading of TiO<sub>2</sub> over the AC surface was taking place via dipping the AC particles in a peroxotitanate solution; after that heating at 180°C in a Teflon lined stainless-steel vessel came after in calcinations at 300 to 800°C [274, 275]. With the help of the AC particles of 0.16 to 0.26 millimeter, the disjunction of the particles from the solution was not too much harder, and photocatalytic action on behalf of the decomposition of MO almost remains identical for 5 cycles. TiO<sub>2</sub> has also been achieved by plugging the pore of AC by paraffin [275]. After loading TiO<sub>2</sub>, by removing the paraffin at 250°C in the air, the high surface region of pristine AC, it could be recuperated and the high photocatalytic action was procured basically for the decay of methylene blue (MB) [276]. TiO<sub>2</sub>: AC has also been composed by mixing TiO<sub>2</sub> particles with some liquid or solid state carbon precursor. By hydrolysis of tetraisopropyl orthotitanate, TiO2 was caused in the exterior region of the poly vinyl butyral (PVB) and TiO2 overloaded PVB was carbonized at a high temperature in the flow of CO<sub>2</sub>.  $TiO_2$  loaded carbon microspheres with 25  $\mu$ m diameter have been prepared from the TiO<sub>2</sub> loaded cellulose microspheres, composed with one step stage division by using the sodium polyacrylate aqueous solutions and cellulose xanthate with the isolated TiO<sub>2</sub> powder [277–279].

(4) Performance of TiO<sub>2</sub>: AC Photocatalyst. Although it is a hard to understand how light could it penetrate the carbon particles to reach the inoperative photocatalyst, TiO<sub>2</sub>: AC composites have quite clear high efficiencies for the photodegradation of a variety of pollutants [238]. In such a case, the presence of the AC seems to change the photocatalytic activity of TiO<sub>2</sub> towards the abasement of organic pollutants beyond the so-called "synergistic" effect [227]. The harmonious aftereffect of the adsorption with AC and photocatalytic disintegration by TiO<sub>2</sub> has been noticed during the deprivation of many kinds of organic toxin.

The basic principle of photocatalysis over illuminated  $TiO_2$ : AC system. But model of a  $TiO_2$ : AC photocatalytic process can be more of a complex issue, it starts the photointensity to the classical aspects of the heterogeneous catalytic system; for example, temporal variations in concentration of iminoctadine triacetate (IT) that is frequently used in excess

plus orchard fields like an insecticide and in the water path of the fields are exposed for pristine AC and three  $TiO_2$ : AC [280, 281]. The hybrid systems were kept without any light on them for 200 h to saturate IT adsorption and after that were exposed to ultraviolet irradiation. 800 h behind this [280], the sample was separated from given test solution and spread once more in the virgin  $1.87 \times 10^{-4}$  mol/L solution and again kept without any light in it for approximately 200 h and then exposed to ultraviolet radiations. As far as pristine AC is concerned, concentration of pollutants was initiated to survive approximately steady without any light and to spread to some extent beneath ultraviolet irradiation. The bottom line is that the photocatalyst system of TiO<sub>2</sub>: AC could have similar photocatalytic results without any light and under ultraviolet radiations [282]. Before mentioned data was supported by the one who stated that the enhancement of photocatalytic activity for the organic burning of pollutants via TiO<sub>2</sub>: AC enzyme. Stated that is, the concentration of 4-chlorophenol solution demotes rapidly for the TiO<sub>2</sub>: AC than TiO<sub>2</sub>, saying that it enhances photoorganic burning approximate by a pseudo-first-order equation with a linear relationship between time and concentration change. The same harmonious result was noticed in the organic burning of pH and 2,4-dichlorophenoxyacetic acid applying similar AC and TiO<sub>2</sub> sample [283]. Furthermore, the pH disintegration is found to be dependent on the mass ratio of TiO<sub>2</sub> to AC (5/10 to 75/10) [231, 284, 285]. The harmonious result is thought to be attributable to the fast movement of pH molecules at the start adsorbed on the AC on the outer surface of TiO<sub>2</sub>; the motivating force of that movement is most probably the differentiation in the surface concentration of pH between AC and TiO<sub>2</sub>. Defined proportion of pH was found to remain at AC, even after the proportion in sample becomes negligibly small.

3.2.2. ZnO: AC Photocatalyst System. After TiO<sub>2</sub>, ZnO supported AC finds broader attraction of use, due to some of the excellent behaviors of ZnO, such as wider availability, stability, and suitable band gap of energy [228]. Problems associated with the usage of ZnO alone as photocatalyst are as well partitioned, especially the complexity in unraveling the powder with the sample after the reaction is over; gathering of particles in delay, particularly at high loading and complexity in purpose to the consistent flow of the domain system has been approved by its surface properties [37, 235] (Figure 13).

The problem is AC has been declared good as for the support of the ZnO photocatalysis system. Replying on using of dioxygen, photo-, and entirely mineralized organic as well as inorganic substances and particularly biorecalcitrant, make the technique is environmentally friendly for toxic waste reduction schemes [225]. Spherical AC particles having ZnS and ZnO were formed from a caption-exchange resin (polystyrene with sulfonate groups and cross-linked by divinylbenzene) and ZnCl<sub>2</sub> aqueous solution, followed by carbonization at almost 500 to 900°C [286]. It has been reported that ZnO is an appropriate substitute to TiO<sub>2</sub> for the photodegradation of Acid Red 14, an azo dye, because it is

photodegradation process that has been proved to be similar to  $TiO_2$  [37, 287] (Figure 14).

Recently it was reported that the instantaneous destruction of inorganic toxin like Cr(VI) and organic toxin, like 4-CP, can be recognized in ZnO: AC photocatalytic reaction system [286–289]. Hence, this technique can be functional over the broader level for aqueous waste reduction.

## 4. Advance Activated Carbon Photocatalytic System

4.1. Granular and Spherical Activated Carbon Photocatalytic Systems. In all kinds of AC, the spherical AC has benefits, such as its frictionless surface, high quality fluidity, and good strength on the powdered and rough AC. Carrying this forward, many proposals have lately examined the sustain of  ${\rm TiO_2}$  to spherical AC [290, 291]. But there are some statements on the granular AC that supported  ${\rm TiO_2}$  photocatalyst which could enhance the demolishing efficiency of many organic compounds, regarding many environmentally related conditions [252, 292] (Figure 15).

Granule AC supports TiO<sub>2</sub> powdered by adding more pollutants and alternatives around the TiO<sub>2</sub>. The pollutants and alternatives can diffuse to the surface of TiO<sub>2</sub>. Granular AC also reduces TiO<sub>2</sub> jelling, which lessens its surface, thereby reducing its enzymatic activities [292]. This was brought by the theory that the absorptivity of AC depending strongly on the molecular morphology and size of the pollutant particles to the TiO<sub>2</sub> surface happens straightforwardly from the solution, and not through the AC surface [293, 294]. The TiO<sub>2</sub> can destroy the pollutants, leading again to generation of granular AC in this situation [294]. Most of the porous AC still in granules, and the problem of separation and recovering of the photocatalyst from the reaction environment is already present.

4.2. ACF Photocatalytic Systems. AC is a freshly developed type of photocatalyst supporting materials consisting of nanographites known as AC fiber ACF. In comparison to the granular AC, ACF has a larger surface area, having greater pore volume, more uniform micropores size distribution, a greater rate of adsorption and desorption, and a rapid attainment of adsorption equilibrium with ACF in the form of fleet are preferable to the handling than granular supports [245, 253] (Figure 16).

The surface-area characteristics of the ACF are identified to depend powerfully on the creative processes, affecting the load of  ${\rm TiO_2}$  and eventually the adsorption of pollutant particles [255]. Nevertheless, the ACF supported  ${\rm TiO_2}$  photocatalyst has sometimes been used for the removal of gas phase pollutants in the environment (Figure 17).

4.3.  $TiO_2$ : Graphene Photocatalyst System. Meanwhile, the beginning of graphene supply is an idea to resolve the limitation brought by the  $TiO_2$ : AC photocatalyst. Recently, functionalized grapheme based semiconductor photocatalyst has attracted attention because of its larger definite surface

area, higher electron conductivity, and adsorption [248] (Figure 18).

A lot of the hard work is used for the combination of  ${\rm TiO_2}$ -graphene photocatalysis hybrid system [295, 296]. Considering advantages due to a higher specified surface region, graphene appeared like a better help to prepare the overloaded nanoparticle metal oxides to attain an identical division not including aggregation. Betterment of the photocatalytic actions of  ${\rm TiO_2}$ -graphene hybrid is associated with huge two-dimensional planar graphene structure supporting the dye adsorption plus squeezing electron hole rejoining because of higher electrical conduction property as has been indicated by the morphological analysis of  ${\rm TiO_2}$ /grapheme photocatalyst [226, 254] (Figure 19).

Additionally, absorption of additional catalyst particles into a particularly graphene leaf at individual places can supply better dynamically in achieving the choosy catalytic otherwise logical procedures and adjusts the composition also the morphology of photocatalysts to enhance their photocatalytic results [297].

4.4. TiO<sub>2</sub>: CNT Photocatalyst System. Barring graphene, carbon nanotube (CNT) has been regarded as a more attractive catalytic support than activated carbon because of combination of electronic, adsorption, and specific semiconducting characteristics [298]. Studies of TiO<sub>2</sub>: CNTs reveal a considerable synergy effect with the metal oxides and carbon phases [298, 299]. Going further, researchers have shown that CNTs can enhance the adsorption and photocatalytic activity of TiO<sub>2</sub> in the presence of ultraviolet [300]. Single walled CNTs execute enhanced and selective photocatalytic oxidation of pH [299]. Heterostructure CNT consists of TiO<sub>2</sub>-xNx and C prepared by carbonization of electron-spun polyacrylonitrile nanofibers containing stabilized titanium oxoacetate [301]. So, CNTs can be used as a reliable material for environmental pollution clearing and can be used to improve the photocatalytic efficiency of TiO<sub>2</sub>.

4.5. TiO<sub>2</sub>: AC Semiconductor Doped Photocatalyst System. The term "doping" means an additional semiconductor having unusual parallel valence bands and conduction of energy levels with the TiO<sub>2</sub> over the outer surface of TiO<sub>2</sub> has been authorized the outcome to progress the photocatalytic results. The method of using coupled semiconductors is the enhancement of the photocatalytic with good organization by raising the charge division and increasing the photoresponding domain. Some researches upon doping an additional semiconductor over outer surface of TiO<sub>2</sub>: AC includes SnO<sub>2</sub>, V<sub>2</sub>O<sub>5</sub>, ZnO, ZrO<sub>2</sub>, and CsS [239, 260, 302, 303]. The entire of it confirms the superior photodegradation efficiency as compared to the TiO<sub>2</sub>: AC. Recent researches have reported the modification of TiO<sub>2</sub>: AC by semi-conductor doping that has led to the improvement of removal of specific containments in the gas resulting in the changes of the physical and chemical properties of the carbon materials. Iron (Fe<sub>3</sub>O<sub>4</sub>) dopant TiO<sub>2</sub>: AC has been found to be a promoter to photocatalysis with a good performance on degradation of Congo red and methyl orange [32]. Some researchers have

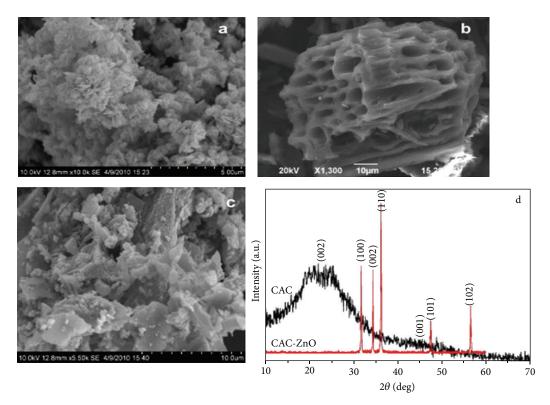


FIGURE 13: Surface morphology of the (a) pure ZnO, (b) pure AC, and (c) AC–ZnO mixture. XRD spectrum of pure AC and AC– $TiO_2$  mixture [37].

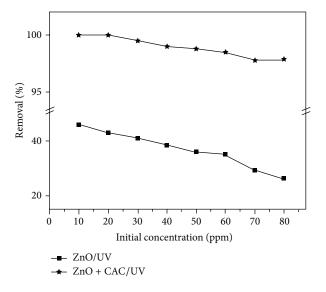


FIGURE 14: Effect of initial concentration of dye in the presence and absence of AC in ZnO/AC photocatalyst system (contact time: 90 min; dose of the catalyst: 200 mg; dose of the CAC: 40 mg) [37].

combined the  $\mathrm{Fe_3O_4}$  compound with  $\mathrm{TiO_2}$ : AC aiming at preparing photocatalyst with magnetic core and photoactive encapsulation [234]. They proved that hybrid systems had magnetic properties and could be separated by magnetic materials. Furthermore, the magnetic hybrid photocatalyst can also be magnetically distributed by a discontinuous magnetic field in an interrupted system [304]. But they found

that the photocatalytic activity of the hybrid system declined because the magnetic particles experienced light dissolution [305].

4.6.  $TiO_2$ : AC Nonmetal Doped Photocatalyst System. Doping of nonmetal, such as N, P, B, C, S, F, chlorine, and bromine, has been also widely used to improve the photocatalytic or

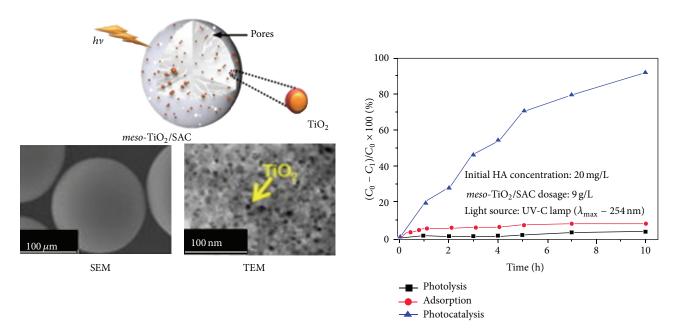


FIGURE 15: Effect of granular and spherical AC on adsorption and photocatalytic activity [252].

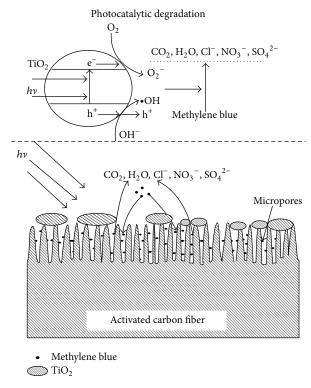


FIGURE 16: Schematic diagram for the adsorption and photocatalytic degradation of methylene blue on the TiO<sub>2</sub>/ACFs [253].

to extend the photoabsorption into visible light of  $\mathrm{TiO}_2$ : AC system [258]. Among them, the P doped  $\mathrm{TiO}_2$ : AC has recently attracted increasing interest due to its enhanced shows a little band gap. It has an adsorption property in the visible light region [306]. Also, it has been found that phosphorous-doped  $\mathrm{TiO}_2$ : AC prepared by the buffer solution method with  $\mathrm{NaH}_2\mathrm{PO}_4$  as precursor showed a greater

photocatalytic activity of acetaldehyde organic burning under visible light absence than the pure sample [307]. It synthesizes the phosphorous-doped  ${\rm TiO_2:AC}$  with high crystallinity and large surface area of hydrothermal process. Methylene blue demoted performance on phosphorous-doped sample was pointedly increased and superior to the commercial Phosphorous 25 [308].

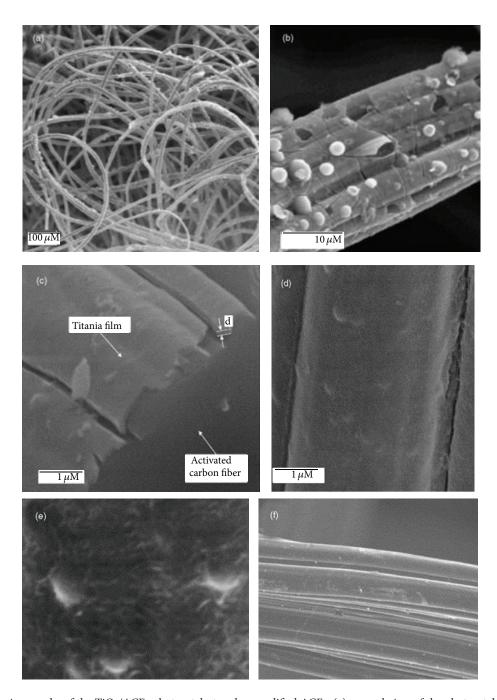


FIGURE 17: SEM micrographs of the  $TiO_2/ACFs$  photocatalyst and unmodified ACFs, (a) general view of the photocatalyst, 100x; (b)  $TiO_2$  coating on single carbon fiber, 2000x; (c) cross-sectional view of  $TiO_2$  coating, 2000x; (d) surface of  $TiO_2$  coating, 2000x; (e) surface of  $TiO_2$  coating, 2000x; and (f) single unmodified activated carbon fiber 4000x [253].

### 5. Conclusion

AC has been employed as an adsorbent for the control of many environmental pollutants due to its high pore density and large uncovered surface area-to-volume ratio. Many AC researches have targeted primarily with concentration in lower to higher concentrations than that associated with indoor aqueous quality. Loading of semiconductors on AC has drawn vast interest while the higher adsorption abilities of

AC can assist in the direction of improvement of organic pollutant about the enzyme, enhancing the toxin transmission method thus enhancing the photocatalytic results. In future, photocatalysis reaction in visible light can be addressed as the main challenge, as reflected by recent intensive scientific endeavors. The state-of-the-art accomplishments in visible light will be induced in selection of organic transformations by AC heterogeneous photocatalysis. In addition, the recent strides are to bridge between AC photocatalysis system

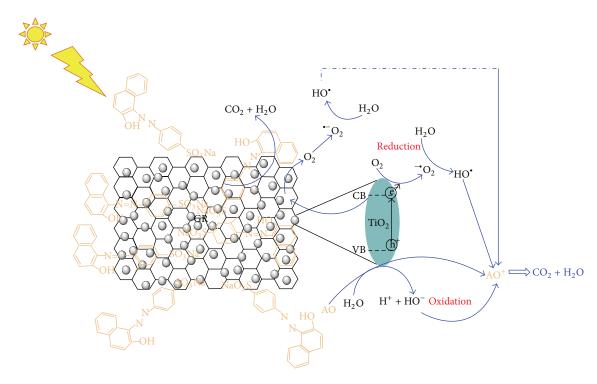


FIGURE 18: Schematic illustrations on the photodegradation mechanism of the synthetic dye on TiO<sub>2</sub>/graphene nanocomposite [248].

and other area of the catalyst with the aim of overcoming the existing limitation of photocatalyst by developing more creative approaches.

### **Abbreviations**

[BMIM]Cl:	1-Butyl-3-meth	ylimidazolium	chloride
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[EMIM]BF<sub>4</sub>: 1-Ethyl-3-methylimidazolium

tetrafluoroborate

[EMIM]Cl: 1-Ethyl-3-methylimidazolium chloride

AC: Activated carbon ACF: Activated carbon fibers

Silver Ag:

 $Al_2O_3$ : Aluminum oxide

Al-MCM-42: Mesoporous aluminium silicate Al-MSU-F: Mesostructure aluminosilicate

AuCl: Gold(I) chloride

C: Carbon

CdSe: Cadmium selenide CeO<sub>2</sub>: Cerium oxide Carbon dioxide  $CO_2$ :  $Co^{2+}$ : Cobalt ions Cobalt(II) oxide  $Co_3O_4$ : Cobalt ferrite  $CoFe_2O_4$ : Cr(VI): Chromium  $Cr^{3+}$ : Chromium ions Chromium(II) chloride CrCl<sub>2</sub>: Chromium(III) chloride CrCl<sub>3</sub>: Chemical Vapor Deposition CVD:

DMA: Dimethylacetate

Dimethylformamide DMF:

Fluoride F:

FCC: Fluid catalytic cracking

Fe<sup>3+</sup>: Iron ions Fe<sub>3</sub>C: Cementite  $H_2O$ : Water H<sub>2</sub>SO<sub>4</sub>: Sulfuric acid

 ${
m H_{3}PW_{12}O_{40}}$ : Tungstophosphoric acids H<sub>3</sub>PW<sub>12</sub>O<sub>46</sub>: Dodeca tungstosilicic acid

H<sub>4</sub>SiW<sub>12</sub>O<sub>40</sub>: Tungstosilicious HCl: Acid hydrochloride Hydroxymethylfurfural HMF: IrCl: Iridium(II) chloride Lanthanum(II) chloride LaCl<sub>3</sub>: LiCl: Lithium chloride

McM 41: Self-bonded zeolite beta

 $Mn^{2+}$ : Manganese ions Nitrogen N:

NaCl: Sodium chloride

Ni: Nickel OH: Hydroxyl P: Phosphate Pd: Palladium

PdCl<sub>2</sub>: Palladium(II) chloride

Pt: Platinum PVB: Polyvinyl butyral Rh: Rhodium Ru: Ruthenium Sulfide S:

Mesoporous silica SBA-15:  $SiO^{2-}$ : Silicon dioxide

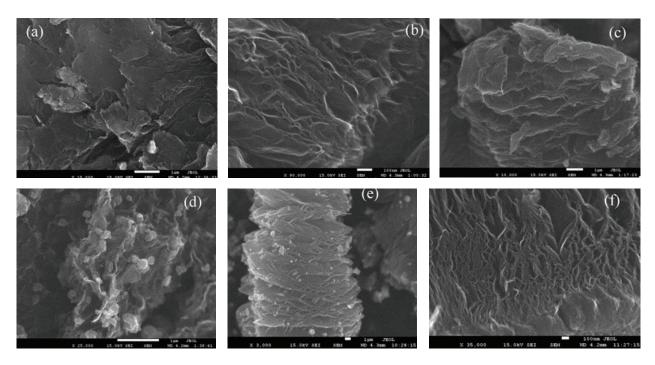


FIGURE 19: FE-SEM images of  $TiO_2$ /graphene nanocomposites at different  $TiO_2$  contents 1 wt% (a), 3 wt% (b), 5 wt% (c), and 10 wt%  $TiO_2$ /graphene heat treated at 450°C under reducing ( $N_2/H_2$ ) (e) and nitrogen (f) [254].

SnCl<sub>4</sub>: Tin(IV) chloride SO<sub>3</sub>H: Sulfonic acid SrTiO<sub>3</sub>: Strontium titanate TiO<sub>2</sub>: Titanium oxide UV: Ultraviolet

V<sub>2</sub>O<sub>5</sub>: Vanadium pentoxide

ZnO: Zinc oxide ZrO<sub>3</sub>: Zirconia

ZrP: Zirconium phosphate ZSM-5: Zeolite Socony mobile 5.

### **Conflict of Interests**

The authors declare that there is no conflict of interests regarding the publication of this paper.

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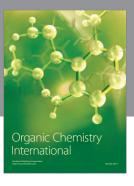
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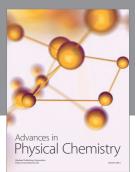
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