SiO₂ Nanoparticles Derived from Arundo Donax L. Ash Composite with TiO₂ Semiconductor for Efficient Photocatalytic Dye Reduction

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Abstract

In this report, the biomass derived silicon dioxide (SiO_2) nanoparticles composite with titanium dioxide (TiO_2) semiconductors used as efficient photocatalyst for degradation of Rhodamine B (RhB) dye molecules under UV-visible light irradiation is proclaimed. At first SiO_2 derived from *Arundo donax* L. ash and TiO_2 synthesized using titanium (IV) isopropoxide by co-precipitation method and then their different compositions prepared by wet impregnation method were exemplified to various optical and atomic level fundamental studies. The amorphous and crystalline nature of SiO_2 and TiO_2 ratify from XRD and here it is found that the crystalline nature decreased in their compositions as compared to TiO_2 . 293 nm UV photons harvesting SiO_2 observed which could be due to more impurity states presence on surface is further accomplished red shift after composition with TiO_2 lead to moving photons harvesting nature towards visible region. The band gap increases in $\text{SiO}_2/\text{TiO}_2$ composites as for TiO_2 composition is rapport well with the aforementioned redshift value. Out of all samples the low recombination rate is procured in 50 wt% SiO_2 /50 wt% TiO_2 composite sample. The separated $\sim 100\text{--}200$ nm sized TiO_2 nanoparticle and aggregated tiny SiO_2 nanoparticles availability in composite sample is authentically substantiated from electron microscopic studies. The presence of Si, O and Ti elements in composite samples probed by XPS. Following the fundamental studies, the photocatalytic degradation ability of the as-prepared samples has been scrutinized against the degradation of Rh B dye in which the pronounced photocatalytic degradation efficiency 93.7% is successfully achieved on 50 wt% SiO_2 /50 wt% TiO_2 nanocomposite photocatalyst.

1. Introduction

From last decade wastewater from various coloring/dying industries is a significant ecological issue, particularly in the developing countries. It is account for that approximately 1–20 % of production of the colors/dyes is directly releasing into natural surface water as material effluents [1, 2]. The angst is that these could be poisonous to microorganisms, oceanic life, and individuals and being a grim bother for both climate and living being [3]. Therefore, globally water pollution turns into a paramount issue in recently due to $\sim 70\%$ of the colors/dyes from the various enterprises straightforwardly mixed with the naturally available water bodies as above said [4]. The basic coloring industries specifically the material industry is widely utilizing textile dyes to coloring their items which are photo catalytically stable and intractable to chemical oxidation process [5][6]. Since this industry likewise utilizes considerable measure of water in their cycles to frame profoundly coloring, the most part of it has been dangerous impact to our biological system because the existence of synthetic compounds [7]. Typically, the traditional physical techniques such as reverse osmosis, ultrafiltration, ion exchange on artificial adsorbent resins, adsorption on the activated carbon materials and coagulation by chemical agents are widely used for the elimination of dye pollutants [8]. These approaches are merely successful in transferring organic mixtures from water phase to another phase, here the secondary effluence formation is inevitable. On the other hand, these methods will need a further step to take care of solid-wastes and renewal of the adsorbent which will

further unnecessarily upsurge the cost to the method. So, it is imperative to implement another approach to eliminate the pollutants prior to releasing them into the water bodies [9].

The different traditional methods presently applied in the evacuation of hued effluents in industrial water. Notwithstanding these could be a classical approach, the prompt entire elimination of the colors is still elusive [10, 11]. Ongoing studies have been dedicated to the utilization of photocatalysis in the expulsion of colors from wastewaters, especially, due to the capacity of this strategy to thoroughly degrade the objective toxins [12, 13]. TiO_2 is a semiconductor with several unique traits such as non-toxicity, acidic and/or basic chemical stability, long-term reliability, and high oxidation power [14]. TiO_2 NPs have been addressing environmental problems as an effective photocatalyst for more than two decades by removing organic pollutants into H_2O and CO_2 in water. As TiO_2 irradiated by photons during dye degradation process it decomposes the water molecules into hydroxyl radicals ($\cdot\text{OH}$), result in degrading of water's organic pollutants [15]. Despite TiO_2 is of a superb photocatalyst, it has a limited photocatalytic activity due to its wide discontinuous energy region in E-K space provides easy and rapid charge carrier separation.

The green synthesis of nanoparticles is an economical, eco-friendly and simple method for preparing metal oxide nanoparticles under mild conditions. The combustion of leaves produces considerable quantities of ultrafine/nanoparticles formed largely by mineral transformation through the high temperature ignition method [16]. Recently research on silica has intensive on the preparation of nanoparticles and this can be derived from either organic chemicals or biomass [17]. Relatively, the usage of biomass is low when compared to chemical precursors due to lack of an assessment and valuation on the reputation of silica from biomass. The natural resources-based silica nanoparticles have gained decisive attention in the field of materials science due to its eminent properties such as large availability, bioactivity, eco-friendliness and cost effectiveness. It is knowing that biomass is an alternate source for commonly used organic precursors. However, the use of biogenic based silica nanoparticles for supporting SiO_2 has been reported by elsewhere [18].

Silica derived from natural plants is one the attractive routes because it is a green synthesis and huge source availability around [19]. Rice husks, sugarcane bagasse, peanut shells and agricultural materials have been widely used till date for deriving silica materials [19, 20]. Just two years back, I. Fatima et al., found that bamboo leaves are also very suitable attractive leaves for extracting SiO_2 material [19]. According to our knowledge, it has never seen before that use of *Arundo donax* L. leaf to produce SiO_2 material. Here it is successfully done for the first time and obtained SiO_2 is composited with TiO_2 in order to find its photocatalytic performance.

So, in this work first SiO_2 derived from *Arundo donax* L. ash, TiO_2 synthesized by simple precipitation method and prepared different compositions of SiO_2 and TiO_2 (25/75, 50/50, 75/25 in wt%) by wet impregnation method. The photophysical and electronic structure properties of samples were investigated continuously using following instrumental analyses XRD, FTIR, UV, PL, SEM, EDX and

mapping, HRTEM and XPS and their properties will be comprehensively described in the upcoming result and discussion's part along with displaying obtained spectra and images. In the end, the photocatalytic degradation performance against the Rh B dye molecules using SiO₂ nanoparticles and TiO₂ semiconductors and their three different composition samples was explored. From this, it has found that 50 wt% SiO₂/50 wt% TiO₂ nanocomposite photocatalyst shows higher photocatalytic degradation activity than other composition ratios as well as TiO₂.

2. Experimental And Section

2.1. Source Materials

Source: Arundo donax L. plant leaves

Reagents: Titanium (IV) isopropoxide (Ti(OCH(CH₃)₂)₃), Sodium hydroxide (NaOH), hydrochloric acid (HCl), ammonia (NH₃), Ethanol (CH₃CH₂OH) and rhodamine B (Rh B) were purchased from Merck.

2.2. Deriving of SiO₂ from Arundo donax L. leaf ash

Fresh Arundo donax L. leaves were collected from the sides of Thenpennai riverbed, Dharmapuri (Dt), Tamilnadu, India. The collected leaves were carefully washed using running tap water and then double distilled (DD) water in order to remove the existed dirt impurities on it. To get well dry leaves, the washed leaves kept in open sunlight atmosphere for 10 days. These were further charred in open air in a small pot to obtain Arundo donax L. leave ash (AD ash). Continuously, this AD ash powder was calcined in muffle furnace at 600°C for 3 h to eradicate carbonaceous materials.

10 g of calcined ash transferred into 250 ml conical flask which already has 2.5 N of sodium hydroxide (NaOH) solution. This reaction solution refluxed at 60°C for 3 h under identical stirring speed. The silica which presents in ash was dissolved and converted into sodium silicate.

The resulting solution was cooled and filtered to get sodium silicate solution. Ash residues was discarded. The sodium silicate solution was acidified using 2 N HCl, here the gel silica formation was formed particularly at around 10–11 pH value. Further, in order to achieve fine particle solution, HCl was added drop by drop into above reaction solution until pH reach 2. Continuously, the ammonia solution was added in the above reaction mixture to increase the pH to 8–9.

Then prepared silica gel was left for 48 h for getting age. This resultant solution was washed using look warm DD water to remove the impurities. Finally, the silica particles were dried in oven at 60 ° C for 24 h to obtain impurity free silica material.

2.3. Preparation of TiO₂ nanoparticles

The TiO₂ NPs were prepared by precipitation method. For that, first 0.2 mole of titanium (IV) isopropoxide and NaOH were dissolved separately in 25 ml of DD water under continuous stirring. These separated solutions were mixed together gently and continued stirring for 240 min with constant stirring rate. The obtained solution was let it be for 24 h to get age. This solution was centrifuged and washed with DD water several times and then dried at 80°C to obtain the TiO₂ NPs. This yield product was calcined at 600°C for 3 h eventually.

2.4. Preparation of different weight percentage of SiO₂/TiO₂ nanocomposite

The various weight ratios of SiO₂-TiO₂ nanocomposites (25:75, 50:50, and 75:25) were prepared through wet impregnation method. These different weight percentages of SiO₂ and TiO₂ nanoparticles are combined into a 30 ml ethanol solution. Following that, the solution was stirred at 50°C for 2 h. After that, the obtained reaction solution was kept at 80°C for 12 h.

2.5. Characterization

The structural, morphological and optical behaviors of the as-prepared samples were thoroughly examined through various instrumental analysis. The X-ray diffraction (XRD) study was used to find the crystalline nature and phase purity of SiO₂, TiO₂ and SiO₂/TiO₂ samples using Rigaku smart lab diffractometer in with Cu K α radiation used ($\lambda = 1.54046 \text{ \AA}$) and recorded data in the 2θ range of 10 to 80°. A Fourier transform infrared spectroscopy (FTIR) was obtained using a Perkin Elmer spectrophotometer in the range of 4000 to 400 cm⁻¹ for studying functional groups which avail in our samples. To know morphology and elemental composition ratio in our prepared samples, Carl Zeiss Supra 55 field emission scanning electron spectroscopies (FESEM) used. Further, the precise morphology of highly photocatalytic sample was investigated by Tecnai T20 high resolution transmission electron microscopy (HR-TEM). The chemical composition was verified by a Thermo Fisher Scientific ESCALAB Xi + X ray Photo Spectroscopy (XPS). The optical absorption spectra were obtained by Ocean Optics USB4000 photo spectrometer. Photoluminescence property of the prepared samples was obtained by Perkin Elmer LS45 photo spectrometer. Finally, UV-visible absorption spectrums of the Rh B solutions were obtained by Epoch-2 microplate reader.

2.6. Evaluation of photodegradation over Rh B dye

The photocatalytic degradation performance of the prepared samples was evaluated against Rh B dye molecule under the visible light irradiation. For that 0.01 g of the prepared samples was first dispersed in 100 ml of Rh B dye molecule (25 ppm) separately. Before light irradiation, the reaction solution stirred and kept in dark room for 1 hr. to attain adsorption/desorption equilibrium. During the irradiation using a xenon lamp (300 W), 1 ml of Rh B solution was collected each 25 min of time break and centrifuged at 6000 rpm to eliminate the photocatalyst particles. The residual absorption changes were measured by UV-visible

spectrophotometer. To investigate the stability of the efficient photocatalyst, the degradation experiments were repeated for 5 times in the identical reaction conditions. Finally, the photocatalyst particles were separated from the dye solution by centrifugation process and dried in an oven. These dry photocatalyst particles were further used for repetitive cycle. To find the role of main active species in the degradation process different scavengers were used in the catalytic process. The triethanolamine (TEOA), benzoquinone (BQ) and isopropyl alcohol (IPA) were used as h^+ , $^*O_2^-$ and *OH scavengers respectively.

3. Results And Discussion

Figure 1. shows the XRD pattern of the SiO_2 , TiO_2 and SiO_2/TiO_2 samples. In Fig. 1 (a), the observed broad diffraction peak at around 20 to 25° infers that SiO_2 is not constructed by periodic repetitive crystal planes which get on with the standard diffraction data (JCPDS: 29–0085) of amorphous SiO_2 . This says that our prepared SiO_2 material for the first time from *Arundo donax* L. ash is being in a state of amorphous nature [1] under certain experimental condition. The XRD pattern of TiO_2 displayed in Fig. 1 (b) reveals sharp diffraction peaks at 2θ values of 25.43°, 37.08°, 37.94°, 38.79°, 48.21°, 54.01°, 55.24°, 62.37°, 62.75°, 68.84°, 70.36°, 72.21° and 76.16° are corresponding to the (101), (103), (004), (112), (200), (105), (211), (213), (204), (116), (220), (215) and (301) crystal planes respectively (JCPDS: 21-1272) [2].

This result is well matched with the anatase phase of body centered tetragonal structure of TiO_2 (19). Here, the recorded diffraction patterns of SiO_2 and TiO_2 are have good harmony association with the previous reports[21]. The calculated d spacing values of (110) and (004) planes of TiO_2 are 0.35 and 0.24 nm respectively and estimated lattice parameter values of a and c are 3.78 and 9.51 Å. Figure 1 (c-e) belongs to the XRD patterns of SiO_2/TiO_2 composite samples. When we increase and decrease the SiO_2 and TiO_2 concentration in wt% respectively in SiO_2/TiO_2 composite samples, the major diffraction peaks of TiO_2 gradually started decreasing clearly tell us the solid-state properties of the amorphous SiO_2 and crystalline TiO_2 mingled clearly.

Figure 2 shows the room temperature FTIR spectra of SiO_2 , TiO_2 and its composition samples displayed in the range of 4000 – 400 cm^{-1} . Figure 2(a), there are three major characteristic peaks were observed below 1200 cm^{-1} which are attributed to the SiO_2 material. These peaks can be assigned to asymmetry Si-O-Si bond stretching (1104 cm^{-1}), SiO_4 tetrahedron ring (796 cm^{-1}) and O-Si-O bond deformation (468 cm^{-1})[22]. Figure 2(b) the peak at 475 cm^{-1} is related to characteristic absorption of Ti-O bond[23]. The availability of both SiO_2 and TiO_2 peaks in SiO_2/TiO_2 composite samples strongly indicates the composite formation between SiO_2 and TiO_2 .

The recorded UV-visible absorption spectra for SiO_2 , TiO_2 and SiO_2/TiO_2 nanocomposite samples is given in Figure. 3 (a-e). In TiO_2 semiconductor, the 2p level of O^{2-} valence band (VB) to 3d levels of Ti^{4+} conduction band (CB) makes energy discontinues region in the crystal structure which allowing the

absorption of photons. The absorption peak observed around 400 nm for TiO_2 indicates the wide band gap nature of the material. Here, the observed sharp absorption peak presumably indicates the good crystalline behavior which strongly confirmed through XRD.

Only the marginal shift in absorption edge of TiO_2 is observed after SiO_2 composition. The separate and magnified absorption spectrum of SiO_2 shows an unclear absorption edge around ~ 293 nm. Generally, amorphous SiO_2 has absorption edge value is at far UV region [24]. The observed absorption edge value is also quietly agreed with the reports[25]. The reason could be more impurity states exist in the prepared SiO_2 sample. It is noticed that the absorbance value in visible region of SiO_2 is high when compare to all other samples reveal that presence of impurity states. The absorption edge shifted to lower energy region as for TiO_2 composition with SiO_2 . This redshift strongly infers that modification of electronic structure by composition making between SiO_2 and TiO_2 . A strong composition formation is also experienced by seeing absorption edges of composition samples exist in between SiO_2 and TiO_2 absorption edges. The energy band gap (E_g) of the as-prepared samples was calculated using Tauc's plots mode [26]. The calculated energy band gap values for SiO_2 , TiO_2 , $\text{SiO}_2/\text{TiO}_2$ (0.25:0.75), $\text{SiO}_2/\text{TiO}_2$ (0.50:0.50) and $\text{SiO}_2/\text{TiO}_2$ (0.75:0.25) samples are 3.82, 3.17, 3.19, 3.21 and 3.22 eV respectively. The energy bandgap of the pure SiO_2 nanoparticles is good agreement with previous report [27]. Moreover, the energy band gap decreases with the increase of TiO_2 composition with SiO_2 , proving that more TiO_2 has been attached onto the surface of SiO_2 by overlapping SiO_2 and TiO_2 conduction and valence band edges. A gradual increasing of TiO_2 band gap value is seen in the graph by 25, 50 and 75% of SiO_2 composition due to overlapping of SiO_2 and TiO_2 electronic structures.

It is well known that the photoluminescence (PL) spectroscopy has been widely used to examine the recombination rate of excited electron-hole pairs and existed defect states in semiconductors[7]. Generally, in wide band gap material the emission band at UV region is attributed to the discontinuous energy region which existed in the Brillouin zone boundary of material leads to exciton formation. Typically, this is called it as material characteristic peak. On the other hand, other emission bands which particularly seen in visible region are recognized as defect states. Figure 5 shows observed relative PL spectra of the prepared samples at an excitation wavelength of 320 nm. In the PL spectrum of pure SiO_2 , the emission band shows a strong, intensified and broad PL peak at 410 nm wavelength indicates the electrons and holes recombine process happen rapidly. The emission peak intensity of 50 % SiO_2 -50 % TiO_2 sample is weakened as compare to other samples strongly demonstrating that the recombination of photogenerated charge carriers is suppressed significantly. Further, the avail deep level emissions from TiO_2 and composite samples belongs to the oxygen, Ti vacancies and surface oxygen vacancies (SOVs). In PL analysis, the eventual conclusion is that the composition of SiO_2 with TiO_2 lead to alter the electronic structure of the material and these changes make favorable photogenerated electron and hole separation which further would be effectively improving the photocatalytic performance when go for the photo degradation of dye molecule applications.

The morphologies of the as-prepared samples were evaluated by FESEM (Fig. 6) In Fig. 6 (a) and (b), the captured SiO₂ and TiO₂ nanoparticles images are shown. The as prepared SiO₂ samples show highly agglomerated tiny nanoparticles this may be due to cluster mechanism involved during synthesis as for identical synthesis conditions.

The TiO₂ nanoparticles are uniform, smooth with average particle size of 80–120 nm. After composite formation (50 wt% SiO₂/50 wt% TiO₂) the nanoparticles are highly agglomerate compared to the pristine TiO₂ sample. Similarly, no morphological changes were observed much more of TiO₂ and SiO₂ indicating the dispersion of SiO₂ and TiO₂ nanoparticles homogeneously in the composite sample. The high distribution of the as-prepared composite may improve its adsorption capacity as well as the active sites of the prepared sample.

The energy-dispersive X-ray spectroscopy (EDX) and mapping study of SiO₂ and 50 wt% SiO₂/ 50 wt% TiO₂ samples were shown in Fig. 7 and Fig. 8 respectively. The observed major elements are related to Si, Ti and O which confirms that presence of TiO₂ and SiO₂ in composition sample.

In order to probe an internal precise morphology, generally the transmission electron microscopy analysis is employed. It can be seen from Fig. 9 (a) and (b) is that tetragonal spherical like TiO₂ and sprinkle like SiO₂ morphologies of TEM images. These have well agreement with the early discussed FESEM analysis. Further, the HRTEM image was taken on spherical like TiO₂ nanoparticle. This shows interplanar spacing value of 0.24 nm which corresponding to the 004-crystal plane. The polycrystalline nature of TiO₂ semiconductor is confirmed by SAED pattern.

XPS analysis was used to investigate the chemical bonds, exact composition, and oxidation state of the compounds. 50 wt% SiO₂/50 wt% TiO₂ nanocomposite sample was subjected to XPS analysis and observed spectra are displayed in Fig. 10. The high-resolution spectra of prepared samples Fig. 10 (c-d) shows characteristic peaks of Ti 2p, Si 2p and O 1s indorsing the presence of Ti, Si and O shown in respectively. The deconvolution peak provides the necessary proof of the synergistic interface of the prepared element in the nanocomposite. Furthermore, the high-resolution spectra of Ti 2p shows two shake-up satellites located at 458.3 and 464.1 eV are corresponding to Ti 2p_{3/2} and Ti 2p_{1/2} peaks respectively representing the existence of Ti⁴⁺ species in the as-prepared sample. The Si 2p spectra was observed at 103.2 eV which can be indorsed to Si-O binding energy [28]. The O 1s spectra exhibit three pecks at 528.9, 531.93 and 532.81 eV ascribed to Ti-O-Ti, Ti-O-Si and Si-O-Si respectively [29–31].

Photocatalytic activity of the prepared samples against Rh B

The Rh B dye is selected here as a model contaminant to study the photocatalytic performance, so that the photocatalytic degradation of Rh B is evaluated to demonstrate the catalytic activity of SiO₂, TiO₂ and its composition (SiO₂/TiO₂) samples. The removal of Rh B dye is examined under UV-visible light

irradiation. Figure 11 demonstrates the photodegradation of Rh B in the certain irradiation time intervals and in the presence of aforementioned photocatalysis.

As shown in Fig. 11 (d), the intensity of the 554 nm absorption peaks decreased rapidly due to the decreases of Rh B chromophore under the UV light irradiation in the existence of our prepared photocatalyst almost disappeared after 150 min signifies that the degradation of Rh B dye molecules. It is noticed that the absorption of the dye decreases with increasing of irradiation time. Compared to pure TiO_2 and SiO_2 , the $\text{SiO}_2/\text{TiO}_2$ nanocomposite samples show high decolorization efficiency of Rh B dye molecules and eventually the observed higher photocatalytic degradation efficiency is in 50 wt% $\text{SiO}_2/50$ wt% TiO_2 nanocomposite photocatalyst which may be due to higher light absorption as well as photogenerated charge carriers' separation. This would be strongly confirmed in the early UV-Visible absorption and Photoluminescence spectra studies.

These photocatalytic reaction rates were found by the pseudo first-order equation by monitoring the absorption of the dye molecules (Fig. 13). The pseudo first-order rate constants (k_{obs}) for the photocatalytic degradation response of Rh B were found by using the plots of $\ln(C/C_0)$ against irradiation time (t), in which C and C_0 are the maximum absorptions of Rh B dye at a certain time and the initial time respectively, which were identified from the sequential absorbance variations in the UV-visible absorption spectra. In Fig. 13, all the calculated R^2 values were larger than 0.95, demonstrating that the data fitted well with the straight lines (Table-1). Among all prepared photocatalysis, the 50 wt% $\text{SiO}_2/50$ wt% TiO_2 composite photocatalysis exhibited the best photo- degradation efficiency.

Table 1
Degradation efficiency, Rate constant (K_{app}) and R^2 values of as prepared photocatalyst samples derived by Pseudo first-order kinetic.

S.No.	Sample name	Degradation efficiency	K_{app}	R^2
1.	SiO_2	34.42	0.0027	0.9919
2.	TiO_2	61.41	0.0066	0.9823
3.	25 wt% SiO_2 -75 wt% TiO_2 nanocomposite	65.42	0.0072	0.9860
4.	50 wt% SiO_2 -50 wt% TiO_2 nanocomposite	93.70	0.0148	0.9516
5.	75 wt% SiO_2 -25 wt% TiO_2 nanocomposite	77.03	0.0101	0.9969

The recyclability is one of the furthestmost features for real use of prepared photocatalysts materials. Hence, the recyclability of the as-prepared 50 wt% SiO_2 -50 wt% TiO_2 nanocomposite was investigated up to four successive cycle and the outcomes are shown in Fig. 14. In each cycle, a fresh Rh B solution was used to examine the stability of the photocatalyst material. The 50 wt% SiO_2 -50 wt% TiO_2 nanocomposite shows its stable photocatalytic action for the four successive runs. Subsequently four repeated cycles,

we noticed only a minor change in the photocatalytic capability compared to the first cycle. This observed loss in degradation efficiency may be due the drops of catalyst particles during the collecting process.

The photocatalytic performance of active species in the degradation process is needs to be recognize the better reaction mechanism of the 50 wt% SiO₂/50 wt% TiO₂ nanocomposite photocatalytic sample.

Therefore, various scavengers were used to found the role of active species in the degradation. So, here we used benzoquinone (BQ), triethanolamine (TEOA), and isopropyl alcohol (IPA) as a $^{\bullet}\text{O}_2^-$, hole and $^{\bullet}\text{OH}$ scavengers, respectively. The experimental results are displayed in Fig. 15. After the addition of the trapping agents to the photocatalysis reaction solution, the Rh B degradation efficiency is in the order of TEOA > BQ > IPA. This trapping experimental results suggest that the degradation of Rh B in the presence of 50 wt% SiO₂/50 wt% TiO₂ composite photocatalysis was most interfered with the existence of IPA it shows that $^{\bullet}\text{OH}$ radicals is the primary active species in the dye degradation process. Also, the adding of BQ scavenges $^{\bullet}\text{O}_2^-$ shows notable decrease in the degradation efficiency it shows that the superoxide radicals also play significant part in the degradation. The radical trapping outcomes reiterate the role of active species in the photocatalytic degradation of Rh B was in the order of $^{\bullet}\text{OH} > ^{\bullet}\text{O}_2^- > \text{h}^+$. This result eventually tells us apart from doping, manipulating different morphologies in TiO₂ semiconductor, the composites formation along with amorphous materials is also a promising route to find out better photocatalytic active materials.

Conclusion

In summary, the SiO₂, TiO₂ and their different compositions (SiO₂/TiO₂) in wt% ratios were successfully synthesized and prepared in which amorphous SiO₂ derived first time from *Arundo donax* L. ash. The result of the structural and morphological analyses demonstrate that strong interactions exist between TiO₂ and SiO₂ when make composition. It would strongly find that formation of the 50 wt% SiO₂/50 wt% TiO₂ composite sample deliver good visible light absorption and separation of charge carrier as for UV-visible and PL analyses. The enhanced photocatalytic degradation activity observed in 50 wt% SiO₂/50 wt% TiO₂ nanocomposite sample due to improved light absorption, charge carrier separation by changing of electronic structure. The scavenging investigation shows that the $^{\bullet}\text{OH}$ radicals contributed more while the degradation of Rh B dye by photocatalytic removal.

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Figures

Figure 1

XRD pattern of (a) SiO₂, (b) TiO₂, (c) 25 % SiO₂-75 % TiO₂, (d) 50 % SiO₂-50 % TiO₂ and (e) 75 % SiO₂-25 % TiO₂,

Figure 2

FTIR spectra of (a) SiO_2 , (b) TiO_2 , (c) 25 % SiO_2 -75 % TiO_2 , (d) 50 % SiO_2 -50 % TiO_2 and (e) 75 % SiO_2 -25 % TiO_2

Figure 3

UV-visible spectra of (a) SiO₂, (b) TiO₂, (c) 25 % SiO₂-75 % TiO₂, (d) 50 % SiO₂-50 % TiO₂ and (e) 75 % SiO₂-25 % TiO₂

Figure 4

Tauc's plots for (a) SiO₂, (b) TiO₂, (c) 25 % SiO₂-75 % TiO₂, (d) 50 % SiO₂-50 % TiO₂ and (e) 75 % SiO₂-25 % TiO₂

Figure 5

PL spectra of (a) SiO₂, (b) TiO₂, (c) 25 % SiO₂-75 % TiO₂, (d) 50 % SiO₂-50 % TiO₂ and (e) 75 % SiO₂-25 % TiO₂

Figure 6

FESEM images of (a) SiO₂, (b) TiO₂ and (c-d) 50 wt% SiO₂-50 wt% TiO₂ nanocomposite

Figure 7

EDX spectra of (a) SiO₂ and (b) 50 wt% SiO₂-50 wt% TiO₂ nanocomposite

Figure 8

elemental mapping images of (a) SiO₂ and (b) 50 wt% SiO₂-50 wt% TiO₂ nanocomposite

Figure 9

(a-c) HRTEM images and (d) SAED pattern of 50 wt% SiO₂-50 wt% TiO₂ nanocomposite

Figure 10

(a) Survey scan spectra of 50 wt% SiO₂-50 wt% TiO₂ nanocomposite high resolution spectrum of (b) Ti 2p, (c) Si 2p and (d) O 1s

Figure 11

Photocatalytic degradation of (a) SiO₂, (b) TiO₂, (c) 25 wt% SiO₂-75 wt% TiO₂ nanocomposite, (d) 50 wt% SiO₂-50 wt% TiO₂ nanocomposite and (e) 75 wt% SiO₂-25 wt% TiO₂ nanocomposite

Figure 12

Plot of dye concentration versus irradiation time for the photocatalytic degradation of Rh B catalyzed (a) SiO_2 , (b) TiO_2 , (c) 25 wt% SiO_2 -75 wt% TiO_2 nanocomposite, (d) 50 wt% SiO_2 -50 wt% TiO_2 nanocomposite and (e) 75 wt% SiO_2 -25 wt% TiO_2 nanocomposite

Figure 13

First-order kinetic plots of photocatalytic degradation of Rh B in the presence of (a) SiO₂, (b) TiO₂, (c) 25 wt% SiO₂-75 wt% TiO₂ nanocomposite, (d) 50 wt% SiO₂-50 wt% TiO₂ nanocomposite and (e) 75 wt% SiO₂-25 wt% TiO₂ nanocomposite

Figure 14

photocatalytic degradation efficiency recycling runs over 50 wt% SiO₂-50 wt% TiO₂ nanocomposite

Figure 15

Scavenger test for degradation of Rh B dye on 50 wt% SiO₂-50 wt% TiO₂ nanocomposite under visible light irradiation