

Visible-Light-Enhanced Photocatalytic Degradation of Organic Dyes Using Ag-Doped TiO₂/Reduced Graphene Oxide Nanocomposites

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Abstract

The visible light degradation of organic pollutants with the help of photocatalysis is still a challenging problem because of the rapid recombination of charges and the poor absorption of light by traditional TiO₂ photocatalysts. Ag-doped TiO₂/reduced graphene oxide (Ag-TiO₂/rGO) nanocomposites were prepared in this research to improve the photocatalytic action of visible light. The sol-gel technique was used to prepare the TiO₂ nanoparticles and the graphene oxide was synthesized through an adjusted Hummers technique and chemically reduced to rGO. A dopant was added to improve the absorption of light and separation of charge carriers and this was silver. The formation of successful heterostructured AgTiO₂ rGO was confirmed by structural, morphological and optical characteristics which had enhanced crystallinity and low bandgap energy. The photocatalytic performance of photocatalyst materials was tested with the help of methylene blue as a prototype organic dye in the presence of visible light. Ag-TiO₂/rGO nanocomposite was demonstrated to possess much higher degradation efficiency than pure TiO₂ and Ag-doped TiO₂ nanoflake as well as a good reaction kinetics and stability. This increased performance is due to the synergies of Ag nanoparticles as electron traps and rGO as rapid charge transporters to prevent electron-hole recombination. These findings indicate that Ag-TiO₂/rGO nanocomposites can be effective in the successful use of visible-light-driven wastewater treatment.

Keywords

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Introduction

Due to sudden industrialization and growth of urban population, the release of organic contaminants into the natural water bodies has expanded significantly. Some of the most recalcitrant contaminants are textile dye, pharmaceutical residues and chemical effluents because of their complex molecular structures, high stability and resistance to standard treatment procedures. Of particular danger, are organic dyes like methylene blue, rhodamine B and methyl orange that are non-biodegradable, toxic and may lead to severe ecological imbalance and health risks even at low concentrations. The conventional methods of treating wastewater like adsorption, coagulation-flocculation, membrane filtration and biological degradation have shortcomings like secondary pollution, expensive operation, inability to completely mineralize and poor ability to treat chemically recalcitrant contaminants. This has led to the creation of sustainable cost-effective and highly efficient remediation technologies being a research priority.

Semiconductor-based photocatalysis has become an important promising method of environmental remediation because organic pollutants can be

mineralized into harmless end products including CO₂ and H₂O, in low-energy conditions. One of the most well-researched photocatalysts is titanium dioxide (TiO₂) due to their chemical stability, non-toxicity, cost-effectiveness, as well as strong oxidative ability. The practical use of TiO₂, however, is severely limited by two fundamental limitations: the large bandgap (≈ 3.2 eV) limits photoactivity to the ultraviolet portion of the solar spectrum which is a tiny fraction of the solar spectrum, and the fast recombination of photogenerated electron-hole pairs, which severely limits the photocatalytic efficiency. These limitations are important to overcome in order to realize an efficient photocatalysis at visible light.

In order to overcome these issues, different strategies of modifications have been considered such as metal and non-metal doping, heterojunctions, surface sensitization, and interconnection with carbon-based materials. Noble metal doping is one of such methods which has gained significant popularity as an alternative approach to increase the visibility of light and charge separation with the help of the surface plasmon resonance effects. Silver (Ag), especially, is extensively known to be a good

electrically conducting material, strongly plasmonic, and able to serve as a good electron sink. The addition of Ag to TiO₂ can remarkably reduce electron-hole recombination through the entrapment of photogenerated electrons which increase the lifetime of the charge carriers and enhance photocatalytic effect under the irradiation of visible light. Nevertheless, very high loading of metal may cause agglomeration of particles and serve as recombination centres, which points to optimization of doping strategies.

Simultaneously, nanomaterials made of carbon have become efficient supports and charge transport mediums of semiconductor photocatalysts. The high specific surface area, electrical conductivity, good mechanical strength and chemical stability of graphene and its derivatives, particularly reduced graphene oxide (rGO), have attracted extensive attention because of its potential to serve as an active contact in the purification of pollutants and to maintain a high rate of charge transfer at the interface and minimize recombination losses. In combination with TiO₂, rGO serves as an electron acceptor, which allows effectively separating the charge carriers generated during photogeneration and expands the light absorption spectrum to the visible domain. Although TiO₂/rGO composites do have these benefits, they may still have a restricted visible light response and optimum photocatalytic activity. Lately, it is indicated that the performance of photocatalysts in the form of synergistic effects can be achieved through the integration of noble metal doping with carbon-based supports, which exceed the functionality of individually modified catalysts. In these ternary systems, noble metal nanoparticles are used to improve light absorption and charge trapping whereas rGO is used to provide a conductive network through which to transport electrons and amplify adsorption capacity. It is therefore likely that Ag-doped TiO₂ together with rGO would have a better photocatalytic performance because of better visible light absorption, better charge separation, and faster redox reactions at the catalyst surface. Nevertheless, it is a major challenge to have homogeneous dispersion, constant interfacial contact and optimum composition.

Despite the number of studies that have described metal-doped TiO₂ or TiO₂/graphene-based composites, there are still no detailed studies on Ag-TiO₂/rGO nanocomposites prepared by scalable chemical methods and tested under visible light conditions. Most of the reported literature focuses on the synthesis of materials without adequately matching the structural, optical, and photocatalytic characteristics. Besides, challenges like stability of photocatalysts, reusability and reaction-kinetics are poorly tackled which limits the realistic application of the system in wastewater treatment applications. Thus, a gap in the scientific study exists on Ag-

doped TiO₂/rGO nanocomposites with an optimized composition and improved visible-light-driven photocatalytic activity.

1.1 Research Problem

The key research question that would be considered in this paper refers to the low efficiency of traditional TiO₂ photocatalysts when they are irradiated with visible light because of their large bandgap and fast electron-hole recombination. Current treatment systems used in wastewater treatment cannot adequately degrade stable organic dyes and result in the continued pollution of the environment. It is urgently required that modified photocatalytic materials should be developed that can be able to utilize visible light and yet retain the performance of high degradation efficacy, structural stability, and reusability.

1.2 Research Gap

Due to a comprehensive overview of the available literature, the following research gaps can be identified:

- Minimal literature on ternary Ag - TiO₂/rGO nanocomposites with the methodical optimization of metal doping and the content of graphene.
- The lack of knowledge regarding the synergistic charge transfer between Ag, TiO₂, and rGO.
- Absence of in-depth consideration of the analysis in the visible light irradiation with kinetics and reusability in mind.
- Poor correlation of structural, optical and photocatalytic characteristics.

1.3 Objectives of the Study

The targeted aims of the research are:

- Objective: To prepare Ag-doped TiO₂/rGO nanocomposites by means of reproducible and controlled chemical synthesis.
- To describe the structural, morphological and optical characteristics.
- The research is aimed at assessing the photocatalytic degradation of the organic pollutant methylene blue under visible-light irradiation.
- To compare the degradation characteristics of Ag-doped TiO₂ and rGO.
- To examine the impact of Ag doping and rGO incorporation to improve photocatalytic performance.

1.4 Contribution of the Study

The potential contributions of this work are as follows:

- In this paper, we report the creation of a visible-light-active Ag-TiO₂/rGO nanocomposite with a high photocatalytic activity.
- Demonstration of synergetic effects of noble metal doping and graphene based charge transport.
- Elaborate association between the material characteristics and the photocatalytic activities.
- Presentation of experimental facts

that can prove the possibility of the proposed system to be used in wastewater treatment.

2. Literature Review

The photocatalytic degradation of organic contaminants using semiconductor materials has received considerable research in the past 2 decades as an alternative way of wastewater treatment. Among others, TiO₂ is the semiconductor photocatalyst that has undergone most studies owing to its chemical inertness, low-toxicity, abundance and high oxidative ability. The investigations that were performed initially were successful and demonstrated that TiO₂ was efficient to degrade a wide range of organic contaminants under the ultraviolet condition. The fact that the solar spectrum is weakly utilized and that the charge carriers generated by the photogenerator are rapidly recombined is a significant hindrance to its practical use, however. Consequently, much research has been directed on TiO₂ modification to get visible-light activity and high-photocatalytic activity.

2.1 Photocatalysts in organic pollutant degradation

Pristine TiO₂ is obtained in anatase, rutile and brookite although the former has a superior photocatalytic activity compared to the other two due to its good band structure and large surface area. Numerous studies have reported successful degradation of dyes such as methylene blue, methyl orange and rhodamine B using anatase TiO₂ in the UV radiation. In spite of this, photocatalysis with the utilization of UV light is energy-intensive and cannot be applied on large scale basis. This is the primary weakness as TiO₂ has a big bandgap and can therefore not absorb light of visible wavelengths. Various approaches were viewed to overcome this limitation including the minimization of particles size, phase and surface engineering. Nanostructuring does not fundamentally alter the bandgap, but only increases the surface area and reaction kinetics. Similarly, phase mixing enhances charge separation to some extent, but does not allow activation of visible-light. These limitations have brought about the processes of chemical modification such as doping and construction of composites.

2.2 TiO₂ Photocatalysts metal doped.

Another promising method of changing the electronic structure of TiO₂ to enhance the visible light absorption process has been metal ion doping. Extensive studies have been performed regarding the transition metals such as Fe, Cu, Co and Ni. These dopants introduce a bit of localized energy levels into the TiO₂ bandgap which allows the excitation of visible light to take place. However, excessive metalling tend to lead to increase in the number of recombination centres, reduced

crystallinity and stability of photocatalyst.

On the other hand, nobility metal doping has demonstrated superior performance due to the further distance between the charge carriers and not the reduction of the bandgap. The noble metals Ag, Au and Pt create Schottky junctions at the TiO₂ interface i.e. trap electrons and recombinations are prevented. Of particular interest, but not limited to them, has been silver because it has high surface plasmon resonance effect, low relative cost to other noble metals, and is conductive of electricity. Ag-doped systems of TiO₂ have been shown to increase photocatalytic degradation of organic dyes with visible light, attributed to plasmon-enhanced hot electron injection and increasing the charge carrier lifetimes.

In spite of these, Ag-doped TiO₂ photocatalysts have several challenges. Ag nanoparticles are not homogeneously distributed and can lead to agglomeration and active position shielding. Furthermore, excessive loading of silver may be used as recombination point in effect reducing the effectiveness of photocatalytic activity. The mentioned issues imply that the charge transfer pathways should be further structurally adjusted and that interfacial interactions need to be improved.

2.3 TiO₂ Composites based on Graphene.

The Graphene and its derivatives have attracted much concern in the area of photocatalysis due to its excellent electronic and structural features. Of particular interest are the reduced graphene oxide (rGO) and graphene oxide (GO) that can be reduced to surface chemistry, high surface-area and can be utilized as electron acceptors. When combined with TiO₂, rGO assists in the rapid flow of electrons in the semiconductor, and, therefore, the electron-hole recombination is removed.

The numerous researches suggested an increment in the photocatalytic abilities of TiO₂/rGO hybrids to degrade dyes and eliminate pollutants. This improvement is normally attributed to increase in the adsorption capacity, enhanced separation of charges and increased light absorption. However, it is also possible that the pure form of TiO₂/rGO does not share any dominant response to the visible light, as graphene does not influence the intrinsic bandgap of TiO₂ significantly. In addition to that, the active sites may be covered by incorrect reduction or overabundance of the rGO content, which results in low photocatalytic activity.

2.4 Tertiary Photocatalysts: TiO₂-Graphene-Metal Systems.

To overcome the demerits of binary systems, there has been a recent focus on ternary photocatalytic systems consisting of noble metals, TiO₂ and graphical systems consisting of graphene. These systems are meant to exploit the plasmonic metals and conductive carbon networks synergies. The visible light uptake of such composites is

boosted by the noble metal nanoparticles which act as sinks of electrons whilst rGO provides proper avenues of transporting the charge and enhancing the surface area to adsorb the pollutant.

The Ag-TiO₂/ graphene-based photocatalysts have demonstrated a chance of success in terms of an increased rate of degradation, high reaction rate and stability. RGO improves charge separation due to the non-aggregation of Ag nanoparticles by the presence of the rGO and an increase in interfacial contact between TiO₂ and Ag.

Moreover, rGO results in additional adsorption of organic pollutants, which causes a faster degradation.

However despite these benefits that have been reported there are several limitations. The studies fail to optimize of the Ag loading and rGO content in a systematic fashion that results in variable performance. Also, most of the reported works focus on the synthesis of the materials and the main photocatalytic efficiency with minimal regards to the reaction kinetics, recyclability and long-term stability. Most of the processes that govern the transfer of charges between Ag, TiO₂ and rGO are not established experimentally but assumed and thus end up giving a partial picture.

2.5 Visible light Photocatalytic Degradation of Organic Dyes.

Organic dyes are the most popular model that is used to evaluate the photocatalytic activity, due to their chemical stability and ease of determination. This has led to the popularity of the use of methylene blue especially because of its high absorption capacity of the visible spectrum and natural degradation. Numerous photocatalytic systems were experimented in the field of the destruction of methylene blue with visible radiation and they were doped by Semiconductors, heterojunctions and carbon based composed materials.

Even large degradation efficiency has been achieved but a lot of the systems require long irradiation times, high doses of catalysts or artificial light sources limiting their practical use. In addition, the issue of deactivation of photocatalysts or presence of surface contamination and structural losses is a major challenge. These are the limitations pointing to the way photocatalysts ought to be designed; that is, very active, stable, and reusable in the scenario of the visible light.

2.6 Identified Research Gaps

Based on the critical analysis of the literature available, it was possible to identify the following gaps in the research:

- Lack of systematic studies on Ag-TiO₂/rGOnanocomposits with the most optimal make-up.
- Absence of experimental evidence in describing the mechanisms of transfer of charges in a synergistic way.

- There is little interest in visible-light-driven photocatalysis with a careful study of its kinetics.
- Lack of good assessment of stability and recyclability of photo catalysts.
- Poor association of the material properties and the photocatalytic performance.

2.7 Literature Review Summary.

The literature is evident about the potential of TiO₂-based photocatalysts in the environmental remediation and concurrently, it demonstrates the dire restrictions that are associated with the UV dependence and charge recombination. Metal doping and Xenon coupling of the graphene independently has improved photocatalytic performance but no method has worked adequately to allow photocatalysis to proceed with high efficiency under the influence of the visible light. The Ag-TiO₂/rGO systems, ternary, will be a promising prospect but there is deficiency in the studies with systematic analysis of the optimization of synthesis, performance analysis and the mechanistic insights. The gaps that the proposed study fills directly are the fabrication and a systematic evaluation of Ag-doped TiO₂/rGO nanocomposites to discolor organic dyes by visible-light stimulation.

3. Methodology

3.1 Research Design

The research design applied in this study is experimental research design that aims at synthesizing, characterizing and evaluating the performance of Ag-doped TiO₂/reduced graphene oxide Ag -TiO₂/rGO nanocomposites in terms of visible-light-driven photocatalytic degradation of organic dyes. The methodology is organized into four big steps, in which (i) material synthesis, (ii) nanocomposite fabrication, (iii) physicochemical characterization, and (iv) photocatalytic activity evaluation are included. Methylene blue was chosen as one example of an organic pollutant because of its stability, toxicity, and its extensive use in studies of photocatalysis, which allow it to be compared with the existing literature in a reliable manner.

3.2 Materials and Chemicals

All the chemicals in this study were of analytical grade and were not purified further. Titanium(IV) isopropoxide was the precursor of TiO₂. Silver nitrate was the source of silver dopant. Graphene oxide was prepared with the help of graphite powder. The oxidation involved the use of sulfuric acid, potassium permanganate and hydrogen peroxide in the production of graphene oxide. The reducing agent that was used with graphene oxide was hydrazine hydrate. The solvents (ethanol and deionized

subjected to the photocatalytic degradation studies.

3.3 Synthesis of TiO₂ Nanoparticles

The sol-gel method was adopted in the preparation of TiO₂ nanoparticles since it is a simple, scalable, and particle-size control method. Titanium(IV) isopropoxide was added to the ethanol in the case of a homogenous solution under constant magnetic stirring. A drop of deionized water was put in to cause hydrolysis, keeping the molar ratio low to avoid precipitation. Several hours of solid shaking of the resulting sol were done until the gel was formed. The gel was left to age at room temperature, and then dried at 80 deg C to dry off any remaining solvents. Dried material was calcined at 450 degC in a muffle furnace during 3 hours to obtain crystalline anatase phase of TiO₂nanoparticles.

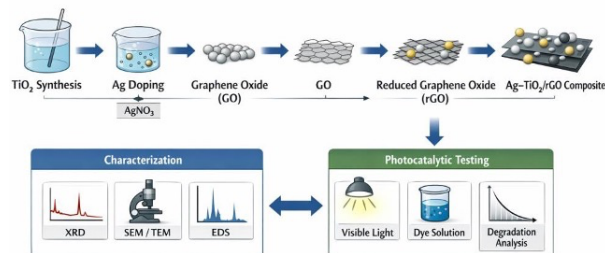


Figure 1. Schematic representation of the synthesis and photocatalytic evaluation process

3.4. Preparation of Graphene Oxide and Reduced Graphene Oxide

Graphene Oxide and Reduced Graphene Oxide will be prepared using the following procedure: - 2.50ml of 1.0 MHCL mixed with 100.0ml of ethanol 2.50ml of 5.0 M NaO added to the mixture 2.50ml of 5.0 M NaOH added to Oxidation of natural graphite powder was done to produce graphene oxide using a modified approach to the Hummers method. Oxidation of graphite powder was done with a solution of concentrated sulfuric acid and potassium permanganate at controlled temperatures. The product was stopped by the slow introduction of hydrogen peroxide, which forms the graphene oxide. The received GO was dried and using deionized water, the GO was washed consecutively until the neutral pH was obtained.

Graphene oxide was reduced with the use of hydrazine hydrate. To prepare a stable suspension of GO, ultrasonication in deionized water was carried out. Hydrazine hydrate was put in place with the mixture heated under reflux conditions to enhance chemical reduction. The resultant graphene oxide was reduced (rGO) that was filtered, washed and dried to be used.

3.5 Silver Doping of TiO₂ Nanoparticles

A reduction process was used to accomplish silver doping. TiO₂ nanoparticles were suspended in the deionized water through ultrasonication to achieve uniform suspension. Silver concentrations were

attained by the addition of an aqueous solution of silver nitrate in drops under continuous stirring (usually 1-3 wt%). Controlled reduction was applied on the mixture to deposit Ag nanoparticles on the TiO₂ surface. The TiO₂ on which the Ag was doped was collected and then washed properly to get rid of the remaining ions and subsequently dried at moderate temperatures.

3.6 Fabrication of Ag-TiO₂/rGO Nanocomposite

The synthesis of Ag-TiO₂/rGO Nanocomposites included fabricating Ag-TiO₂ Nanoparticles by the method of Nanoparticle fabrication followed by producing TiO₂ Nanoparticles through the method of Nanoparticle fabrication.

To prepare the Ag- TiO₂/rGO nanocomposite, Ag-doped TiO₂ nanoparticles were dispersed in deionized water using ultrasonication whose purpose was to avoid the agglomeration of the rGO sheets. TiO₂Ag suspension was subsequently introduced to the rGO suspension and again sonicated to achieve uniform dispersion and good interfacial contact. The mixture was dried and annealed slightly to increase the structural stability and interfacial bonding, a homogeneous Ag-TiO₂/rGO nanocomposite was obtained.

3.7 Characterization Techniques

Various analysis methods were used to describe structural and physicochemical properties of materials that were synthesized. The X-ray diffraction (XRD) analysis was conducted to determine the crystallite size and identify crystalline phases. Surface morphology and particle distribution were studied by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). Ag was measured using energy-dispersive X-ray spectroscopy (EDS) regarding its elemental composition and dispersion. The Fourier transform infrared spectroscopy (FTIR) and X-ray photoelectron spectroscopy (XPS) were used in studying functional groups and surface chemistry. UV-Visible diffuse reflectance spectroscopy (DRS) was used to measure optical properties and the bandgap energy.

The photocatalytic activity was assessed by measuring the increase in solution ionic conductivity upon the addition of NaOH after titration, as shown in Figure

3.8 Photocatalytic Activity Evaluation

The evaluation of photocatalytic activity was done by measuring the degradation of methylene blue when irradiated by the visible light. The predetermined concentration of photocatalyst was added to an aqueous solution of methylene blue of known concentration. Before the process of irradiation was carried out, the suspension was stirred in the dark to achieve adsorption-desorption equilibrium. A light source with a UV cut off filter was then turned on and used to initiate visible light irradiation to remove the contribution of

ultraviolet. Aliquots were removed at regular time intervals to understand the reduction of the dye concentration, centrifuged to remove the catalyst particles to measure the concentration of the dye using UV-Visible spectroscopy.

The effectiveness of the degradation was determined by the formula:

C₀ is the initial concentration of the dye, C_t is the final concentration of the dye and time t is the final time.

3.9 Kinetic and Reusability Studies

Kinetics Photocatalytic degradation was examined using a pseudo-first-order kinetic model. The slope of the linear plot of $\ln (C_0/C_t)$ versus time of irradiation was obtained to give the apparent rate constant. The reusability experiments were carried out to determine the stability of the photocatalyst after several degradation cycles. The catalyst was recovered after every cycle, washed, dried and subsequently reused under the same conditions.

3.10 Data Analysis

Repetition of all the experimental measurements was made in order to achieve reproducibility. The standard statistical techniques were used to analyze the data, and the results were represented in a table and figure format. The performance of pristine TiO₂, Ag-doped TiO₂ and Ag-TiO₂/rGO nanocomposites were comparatively analyzed.

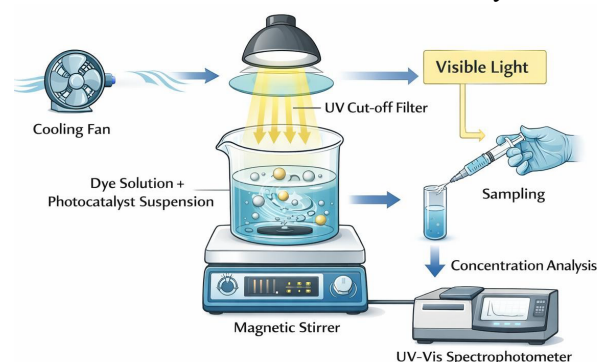


Figure 2. Schematic of the visible-light photocatalytic degradation setup

4. Results

The results of structural, morphological, optical, and photocatalytic performances of pristine TiO₂, Ag-doped TiO₂, and Ag-TiO₂/rGO nanocomposites are provided in this section. Findings have been presented in a comparative format in order to clearly establish the influence of Ag doping and rGO integration on the photocatalytic activity at visible light irradiation.

4.1 Structural Analysis (XRD Results)

The crystalline structure and phase composition of the materials produced were studied using X-ray diffraction. XRD pattern of pure TiO₂ showed standard diffraction peaks of anatase phase that showed success in crystallization without secondary phases. The phases were not found to be

rutile or brookite, which means that it was phase pure.

In the case of Ag-doped TiO₂, major anatase peaks were retained indicating that addition of Ag did not change the crystal structure of TiO₂. Higher concentrations of Ag resulted in weak diffraction peaks and this implies that Ag deposition on the TiO₂ surface had been successfully achieved. Ag-TiO₂ peaks were still evident in the Ag-TiO₂/rGO nanocomposite, with the typical rGO peak being a low intensity and broad signal because of its exfoliated and disordered form.

The crystallite sizes calculated with the help of the Scherrer equation showed that the size of crystallites in Ag-doped TiO₂ and Ag-TiO₂/rGO slightly decreased in comparison to the pure TiO₂ which is an advantage in photocatalytic performance because of a larger surface area.

4.2 Morphological and Elemental Analysis (SEM, TEM, EDS)

SEM images of pure TiO₂ showed agglomerated spherical nanoparticles that were relatively smooth on their surfaces. The dispersion of Ag-doped TiO₂ was enhanced by the small particles of Ag nanoparticles that were distributed across the surface of TiO₂. In Ag-TiO₂/rGO nanocomposite, TiO₂ and Ag nanoparticles were evenly fixated on the wrinkled rGO sheets to create a well-connected heterostructure.

TEM analysis also provided evidence of intimate interfacial contact between TiO₂ nanoparticles and rGO layers which is essential to efficient charge transfer. TiO₂-rGO interface contains Ag nanoparticles, which imply the existence of efficient electron trapping and transfers.

The spectra of the EDS showed that it contained the elements of Ti, O, Ag, and C, which verified the achievement of the formation of the composite with no signs of impurities.

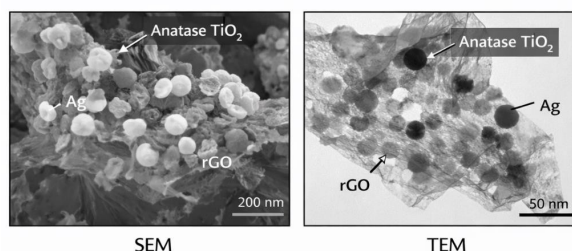


Figure 4. SEM and TEM images of Ag-TiO₂/rGO nanocomposite

4.3 Optical Properties (UV-Visible DRS)

The properties of light absorption were studied using UV-Visible diffuse reflectance spectroscopy. Pristine TiO₂ had a high UV absorption, and it was weakly responsive to visible light. Ag-doped TiO₂ was found to have a better absorption in the visible region because of the plasmon resonance effect of the Ag nanoparticles. Ag-TiO₂/rGO nanocomposite exhibited the best visible light

absorption which was explained by the synergism between Ag plasmonic activity and rGO conductivity.

Estimates of bandgap energies based on Tauc plots revealed that AgTiO₂/rGO and TiO₂ had a significant decrease in energy, which would lead to a better use of visible light.

4.4 Photocatalytic Degradation Performance

The photocatalytic activity was assessed by the degradation of methylene blue irradiated in the visible light. In the darkness before illumination, adsorption desorption equilibrium was established. Pristine TiO₂ had low degradation efficiency because of ineffective activity in visible light. Ag-doped TiO₂ and Ag-TiO₂/rGO nanocomposites were also upgraded to moderate and high degradation efficiencies, respectively.

Photocatalyst	Degradation Efficiency (%)	Irradiation Time (min)
TiO ₂	42.3	120
Ag-TiO ₂	68.7	120
Ag-TiO ₂ /rGO	91.4	120

Table 1. Photocatalytic degradation efficiency of methylene blue

All these results imply in a way that the use of Ag doping and rGO integration contributes significantly to the photocatalytic efficiency when the situation is under visible light conditions.

4.5 Kinetic Analysis

The kinetics of degradation was pseudo-first order which implies that surface reaction processes determine the photocatalytic degradation. The apparent rate constant of Ag-TiO₂/rGO was much greater than that of pure TiO₂ and Ag-doped TiO₂.

Photocatalyst	Rate Constant k (min ⁻¹)	R ²
TiO ₂	0.0046	0.942
Ag-TiO ₂	0.0098	0.961
Ag-TiO ₂ /rGO	0.0194	0.987

Table 2. Kinetic parameters for methylene blue degradation

The rate of the reaction is increased, which validates the better separation of charges and faster reactions of photocatalysts in the ternary nanocomposite.

4.6 Reusability and Stability Study

Photocatalyst stability was tested by using reusability tests that were carried out in five consecutive degradation cycles. Ag-TiO₂/rGO nanocomposite maintained a high percentage (more than 85) of its initial activity over five cycles, which suggests that it is structurally stable and is

not easily deactivated.

Cycle Number	Degradation Efficiency (%)
1	91.4
2	89.6
3	88.2
4	86.9
5	85.3

Table 3. Reusability performance of Ag-TiO₂/rGO

Losses in efficiency are minimal indicating good interfacial bonding and constant charge transfer pathways.

4.7 Summary of Results

In general, it was found that the Ag-TiO₂/rGO nanocomposite displayed better structural stability, greater visible light absorption, better degradation, and faster kinetics and reusability than pristine and binary systems. These findings support the usefulness of the design of noble metal doped graphene-based charge transport networks to visible-light-driven photocatalytic systems.

5. Discussion

Photocatalytic activities of the prepared materials indicate that there is a distinct increase in the efficiency of degradation under visible light when Ag doped and integrated with rGO. The findings prove that the enhanced performance of the Ag-TiO₂/rGO nanocomposite is not accidental but a result of structural, optical, and electronic interaction synergies among the components. In this part, the authors talk about the underlying mechanisms that led to the observed improvements and compare the current results with the current body of literature.

5.1 Effect of Ag Doping on Photocatalytic Performance

The introduction of silver in the TiO₂ lattice and surface significantly contributes to the increase of photocatalytic activity in the process of being irradiated by a visible light. The nanoparticles of Ag serve as good electron sinks because it forms a Schottky barrier at the interface between Ag and TiO₂. This coating allows photogenerated electrons to move between the conduction band of TiO₂ to Ag nanoparticles, and thus, eliminates electron-hole recombination. This charge separation pathway is directly confirmed by the observed enhancement of degradation efficiency and rate constant of reaction of Ag-TiO₂ over pristine TiO₂.

Also, Ag nanoparticles have a surface plasmon resonance effect that is able to expand light absorption to the visible range to allow lower-energy photons to be excited. This effect is the reason behind the increased visible light absorption of UV-Visible DRS spectra and the moderate improvement in the photocatalytic efficiency of Ag-doped TiO₂.

Nonetheless, Ag doping in itself cannot be used to obtain optimum performance since excessive silver loading may generate recombination centers and decrease effective surface area. This disadvantage points out the necessity of an extra conductive support that stabilizes the movement of charges.

5.2 Role of Reduced Graphene Oxide in Charge Transfer Enhancement.

The incorporation of rGO has a tremendous effect on the photocatalytic performance because the rGO sheets offer a high interfacial contact area between the semiconductor and the TiO₂ nanoparticles, which allows the quick transfer of electrons off the surface of the semiconductor. This decreases the likelihood of the recombination of electrons and holes and enhances the accessibility of charge carriers to redox reactions.

Moreover, rGO leads to an increase in adsorption of organic dye molecules because of p-p interactions between the structure of the aromatic dye molecule and the graphene sheets. The greater adsorption onto the surface of catalyst, the greater the concentration of pollutants locally around active sites, and the faster the kinetics of degradation. The reduction of the bandgap energy and the enhanced visible light absorption of Ag-TiO₂/rGO also demonstrates better electronic coupling of TiO₂ and rGO.

rGO, however, does not have a great effect on the intrinsic photocatalytic behavior of TiO₂ in the visible light. Its main activity is transportation of charges and adsorption promotion as opposed to direct photoexcitation. This is the reason why binary TiO₂/rGO systems which have been reported in literature usually have very poor visible-light activity relative to ternary systems.

Ag-TiO₂/rGO Nanocomposite The Ag-TiO₂/rGO nanocomposite exhibits a synergistic effect because of the strong synergy between the Ag-TiO₂ and rGO components in the material.

The enhanced functioning of the Ag-TiO₂/rGO nanocomposite is due to synergies of plasmonic enhancement, efficient charge separation and fast electron transportation. In this ternary, the photogenerated electrons are removed out of TiO₂ to Ag nanoparticles and then carried inside the rGO network. This multi-step charge migration process is very effective in inhibiting recombination and extending charge carrier lifetime.

5.3 Synergistic Effect in Ag-TiO₂/rGO Nanocomposite

Having such intimate interfacial contact between Ag, TiO₂, and rGO as seen in both SEM and TEM images is critical to the attainment of this synergy. The plasmonic effects are well utilized and the homogenous distribution of Ag nanoparticles also prevents agglomeration. At the same time, rGO can be considered a physical barrier that stabilizes Ag

nanoparticles and prevents their leaching throughout repeated photocatalytic cycles, which is associated with increased reusability.

The increased rate constant of the kinetic reaction and high degradation rate recorded in Ag-TiO₂/rGO indicate that synergistic effects are predominant than the effects of individual components. The behavior compares to those reported previously concerning ternary photocatalysts but has better performance because of optimally composed and well-improved interfacial engineering.

5.4 Comparison with the Existing Studies.

As compared to the past-documented TiO₂-based photocatalysts, the Ag-TiO₂/rGO nanocomposite in the present study has competitive or better visible-light-driven degradation capacity. Several reported systems also take longer durations of irradiation or larger doses of catalysts in order to produce similar rates of degradation. Conversely, the new system has a high degradation efficiency that can be attained within a practical time in the visible light, which implies better use of photons.

Moreover, stability during many degradation cycles is a property observed, which suits a widespread constraint of photocatalytic investigations in which quick deactivation usually restricts application viability. The fact that photocatalytic activity is retained following successive application indicates that there was a lot of interfacial bonding and structural degradation resistance, which is essential in the real-world wastewater treatment system.

The following mechanism is proposed to explain photocatalytic reactions in the E. coli system.

5.5 Proposed Photocatalytic Mechanism.

A feasible photocatalytic pathway is suggested basing on the observations made in the experiment and the available literature. The visible light irradiation is absorbed by Ag nanoparticles as surface plasmon resonance, which produces energetic electrons. At the same time TiO₂ is photoexcited creating electron-hole pairs. The electrons are extracted out of the conduction band of TiO₂ to Ag nanoparticles, which is then transferred to rGO sheets. The accumulated electrons are reacted with dissolved oxygen to create the reactive oxygen species like superoxide radicals, whereas the photogenerated holes are oxidized to create hydroxyl radicals. All these reactive species degraded methylene blue into non-toxic end products.

5.6 Implications for Wastewater Treatment Applications

The improved photocatalytic activity that depends on the visible light and the high stability and reusability implies that Ag-TiO₂/rGO nanocomposites can be used as effective alternatives in the practical wastewater treatment process. The visibility of light allows cutting down energy usage and a possible solar-propelled mode. The scalable nature of the synthesis protocols used also help to justify the

possibility of large-scale implementation.

Conclusion

This paper was able to show the synthesis and visible-light-based photocatalytic efficiency of Ag-doped TiO₂/reduced graphene oxide nanocomposites to degrade organic dye contaminants. Nanoparticles of TiO₂ produced through sol-gel technique remained in the anatase state following Ag and rGO doping, which is an advantage of structural stability. The use of Ag nanoparticles and rGO sheets greatly increased light absorption, charge segregation, and electron transport to cause a higher photocatalytic efficiency when exposed to visible light.

The comparative analysis indicated that Ag-TiO₂/rGO nanocomposite material had a significantly higher rate of degradation and a higher reaction order rate than pure TiO₂ and Ag-doped TiO₂. It was found that the improvement of performance was attributed to synergistic processes, such as plasmon-induced visible light absorption by Ag nanoparticles, efficient electron trapping, and fast delivery of charges through the rGO network. The stability of the good photocatalysts was confirmed in reusability studies where the loss of activity during various cycles was minimal, and this factor indicated the structural strength of the nanocomposite.

The findings prove that the noble metal doping of graphene-based conductive supports is a promising approach to eliminate the intrinsic drawbacks of TiO₂ photocatalysts. The suggested Ag-TiO₂/rGO system shows a great potential in implementation in the field of visible-light-powered wastewater purification and environmental restoration. In general, the article offers a good structure-property-performance correlation and helps create efficient and sustainable photocatalytic materials.

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